

# Muonic helium hyperfine structure measurements at J-PARC MUSE

**P. Strasser,<sup>a,b,\*</sup> S. Fukumura,<sup>c</sup> Y. Goto,<sup>d</sup> K. Asai,<sup>d</sup> M. Fushihara,<sup>d</sup> T. Ino,<sup>a,b</sup> S. Kanda,<sup>a,b</sup> S. Kawamura,<sup>d</sup> M. Kitaguchi,<sup>d,e</sup> S. Nishimura,<sup>a,b</sup> T. Oku,<sup>f,g</sup> T. Okudaira,<sup>d,f</sup> H. M. Shimizu,<sup>d,a</sup> K. Shimomura,<sup>a,b</sup> H. Tada<sup>d</sup> and H. A. Torii<sup>h</sup>**

(MuSEUM Collaboration)

<sup>a</sup>*Institute of Materials Structure Science (IMSS), High Energy Accelerator Research Organization (KEK), 1-1 Oho, Tsukuba, Ibaraki 305-0801, Japan*

<sup>b</sup>*Materials and Life Science Division, J-PARC Center, 2-4 Shirakata, Tokai-mura, Naka-gun, Ibaraki 319-1195, Japan*

<sup>c</sup>*Department of Physics, Niigata University, 8050 Ikarashi 2-no-cho, Nishi-ku, Niigata City, Niigata 950-2181, Japan*

<sup>d</sup>*Graduate School of Science, Nagoya University, Furo-cho, Chikusa-ku, Nagoya, Aichi 464-8601, Japan*

<sup>e</sup>*Kobayashi-Maskawa Institute, Nagoya University, Furo-cho, Chikusa-ku, Nagoya, Aichi 464-8601, Japan*

<sup>f</sup>*Advanced Science Research Center, Japan Atomic Energy Agency (JAEA), 2-4 Shirakata, Tokai-mura, Naka-gun, Ibaraki 319-1195, Japan*

<sup>g</sup>*Graduate School of Science and Engineering, Ibaraki University, 2-1-1 Bunkyo, Mito, Ibaraki 310-8512, Japan*

<sup>h</sup>*School of Science, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan*

*E-mail:* [patrick.strasser@kek.jp](mailto:patrick.strasser@kek.jp)

Muonic helium is a hydrogenlike atom composed of a helium atom with one of its two electrons replaced by a negative muon. Its ground-state hyperfine structure is a sensitive tool for testing the theory of three-body atomic systems and bound-state quantum electrodynamics and determining fundamental constants of the negative muon magnetic moment and mass. New precise measurements are now in progress at J-PARC Muon Science Facility (MUSE). Zero-field measurements have already been carried out, and the results are more precise than previous measurements 40 years ago. High-field measurements are now in preparation. Furthermore, a new experimental approach to recover the negative muon polarization lost during the muon cascade process in helium is being investigated, which could drastically improve the measurement accuracy. The first laser repolarization experiments have recently been performed. The status of these new muonic helium HFS measurements and the latest results are presented.

*International Conference on Exotic Atoms and Related Topics and Conference on Low Energy Antiprotons (EXA-LEAP2024)  
26-30 August 2024  
Austrian Academy of Sciences, Vienna.*

---

\*Speaker

## 1. Introduction

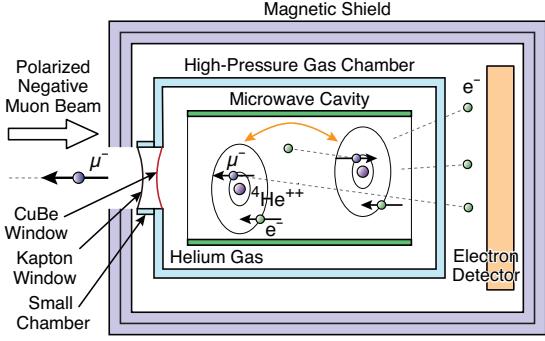
New precision microwave spectroscopy measurements of the ground-state hyperfine structure (HFS) of muonic helium atoms are now being carried out at the Muon Science Facility (MUSE) of the Japan Proton Accelerator Research Complex (J-PARC) [1, 2]. Muonic helium is composed of a helium atom with one of its electrons replaced by a negative muon ( $\mu^-$ ). The Bohr radius of the bound  $\mu^-$  in helium is  $\sim$ 400 times smaller than that of a hydrogen atom. The muon is so closely bound to the helium nucleus that it nearly completely screens one proton charge, producing a “pseudonucleus”  $[\mu^-{}^4\text{He}^{++}]^+$  with a positive effective charge and a magnetic moment nearly equal to that of a negative muon  $\mu_{\mu^-}$ . Thus, it can be regarded as a heavy hydrogen isotope, similar to muonium, another hydrogenlike atom made of a bound state of a positive muon and an electron. The ground-state hyperfine structure of muonic helium results from the interaction of the negative muon magnetic moment and the remaining electron and is very similar to that of muonium but inverted because of the different signs of their respective muon magnetic moments. A comprehensive review of theory and experiments on muonic helium atoms is given in [3].

Muonic helium HFS has only been measured twice in the 1980s directly at zero magnetic field [4] and indirectly at high field [5]. New precision measurements of the muonium HFS interval using a microwave magnetic resonance technique are now in progress at J-PARC by the MuSEUM Collaboration [6–8]. The same method can be used to precisely measure the muonic helium HFS interval  $\Delta\nu$ , which is a sensitive tool to test three-body atomic systems and bound-state quantum electrodynamics (QED) theories. Measurements at high field also let us to determine the fundamental constants of the negative muon magnetic moment and mass. The world’s most intense pulsed negative muon beam at J-PARC MUSE allows improving previous measurements and testing further *CPT* invariance by comparing the magnetic moments and masses of positive and negative muons (second-generation leptons). Moreover, a more precise determination of the muonic helium atom HFS will be beneficial to test and improve the theory of the three-body atomic system.

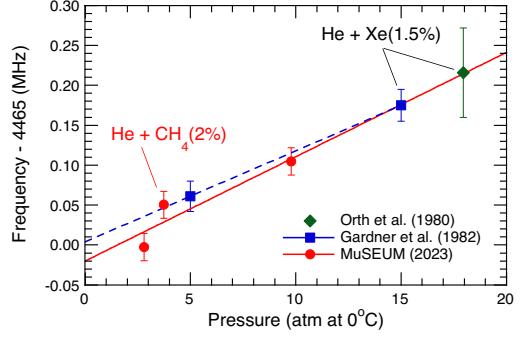
Already, new precise measurements of the muonic helium HFS performed at zero magnetic field have better precision than previous measurements in the 1980s [2]. High-field measurements are now in preparation. Also, a new experimental approach to improve HFS measurement accuracy is being investigated by repolarizing muonic helium atoms using a spin-exchange optical pumping technique. An overview of the different features of these new muonic helium atom HFS measurements and the latest results are reported.

## 2. New muonic helium HFS measurements a zero field

What makes muonic helium HFS measurements more challenging is that the initial  $\mu^-$  polarization ( $\sim$ 100%) is strongly reduced down to 2–5% [9, 10] during the muon cascade process in He due to Auger transition and collisional Stark mixing. This value should be compared to the case of muonium (50%), thus making it more challenging to measure the HFS interval as we need to expand the measurement period by 100 times to get similar statistics with muonic helium. Also, when a He atom captures a  $\mu^-$  ejecting both electrons via Auger transitions in the process, and a  $(\mu^-{}^4\text{He})^+$  ion in its ground  $1s$  state is formed, it cannot capture an electron from the neighboring He atoms because its electron binding energy is similar to that in hydrogen (13.6 eV). Thus, to form a



**Figure 1:** Schematic view of the experimental arrangement enclosed in a magnetic shield box made of three layers of permalloy to measure muonic helium HFS at zero-field.



**Figure 2:**  $\Delta\nu$  as a function of  $\text{He} + \text{CH}_4$  (2%) gas pressure (closed circle). The solid line shows the linear fit to the data to determine  $\Delta\nu(0)$ . Previous results measured with  $\text{He} + \text{Xe}$  (1.5%) from [4, 5] are shown.

neutral  $\mu\text{He}$  atom, the prerequisite to measuring HFS, a collision with a foreign gas atom acting as an electron donor is necessary. Here,  $\text{CH}_4$  was preferred to  $\text{Xe}$  used previously [4, 5] because of its reduced total charge ( $Z = 10$ ), which makes it less likely to capture muons (Fermi-Teller  $Z$  law), a similar ionization potential (12.5 eV), and  $\text{CH}_4$  is also believed to give a larger residual polarization compared to  $\text{Xe}$  [11], making it advantageous for the measurements.

The experiment was performed in the D2 area of MUSE D-line. The apparatus enclosed in a magnetic shield box made of permalloy for zero-field measurements is shown in Fig. 1. In brief, pulsed polarized  $\mu^-$  were stopped into a microwave cavity located inside a gas chamber containing pressurized He gas with an admixture of 2%  $\text{CH}_4$  as an electron donor to form neutral  $\mu\text{He}$  atoms efficiently. The muon spin was flipped by applying a microwave magnetic field in the cavity. Electrons ( $e^-$ ) from  $\mu^-$  decay were emitted preferentially in the direction antiparallel to the  $\mu^-$  spin. At the resonance, the microwave field induced the  $\mu^-$  spin flip, changing the angular distribution of decay electrons, which were detected with segmented scintillation counters placed downstream. Muonic helium HFS measurements were performed by scanning the microwave frequency and measuring the decay  $e^-$  asymmetry with and without microwave ( $N_{\text{ON}}/N_{\text{OFF}} - 1$ ) to determine the resonance frequency  $\Delta\nu$ . Muonic helium HFS resonance curves were measured at three different He gas pressures of 3.0, 4.0, and 10.4 atm. The resonance curve centers were determined by fitting a theoretical resonance line shape using the “old muonium” method [12]. The three frequency values  $\Delta\nu$  measured with  $\text{He} + \text{CH}_4$  (2%) are shown in Fig. 2 as a function of the gas density in atmosphere at 0°C and corrected for nonideal gas behavior (closed circle). Previous results from [4, 5] measured with  $\text{He} + \text{Xe}$  (1.5%) are also shown for comparison. The HFS frequency at zero pressure  $\Delta\nu(0)$  of a free  $\mu\text{He}$  atom was obtained by fitting a linear pressure shift to the data (solid line). By fitting our measured values with  $\Delta\nu(p) = \Delta\nu(0) + Ap$ , we obtained  $\Delta\nu(0) = 4464.980(20)$  MHz (4.5 ppm). The details are reported in [2].

After nearly 40 years, new precise measurements of the muonic helium HFS were performed, and 3 times better precision than the previous measurement at zero field was achieved, and without relying on muonium data to determine the HFS frequency at zero pressure [4]. Our result is also more precise than the high field measurement [5], improving the current world record by a factor

of 1.5, and the first performed with  $\text{CH}_4$  admixture to efficiently form neutral  $\mu\text{He}$  atoms. It should be noted that muonic helium HFS is the only available experimental data for three-body muonic atoms. Unfortunately, the latest theoretical value [13] is still 30 times less precise. However, there is hope that new theoretical calculations developed for HFS in  $^3\text{He}$  [14] could be applied to muonic helium HFS. This would improve the current theory to the same level as the present experimental accuracy and give the first opportunity to test QED effects in three-body muonic atoms.

### 3. Muonic helium atom repolarization

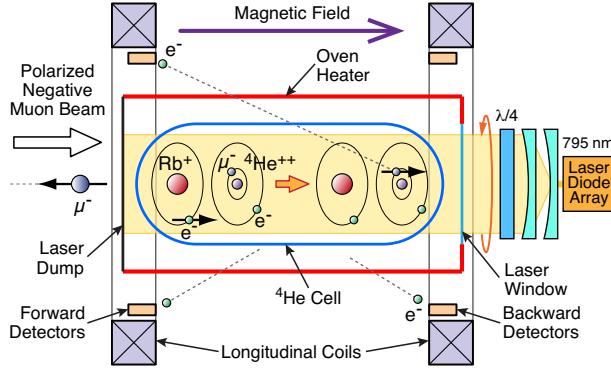
As already introduced earlier, during the muon cascade process in He to form a  $\mu\text{He}$  atom in its ground  $1s$  state, the initial muon polarization ( $\sim 100\%$ ) is strongly reduced due to Auger transition and collisional Stark mixing, leading to a remaining residual polarization of only 2–5% [9, 10]. Therefore, a key factor in improving muonic helium HFS measurements is to efficiently produce neutral muonic helium atoms while restoring the initial muon polarization. Incidentally, the residual polarization in  $\mu\text{He}$  can be improved nearly tenfold by applying the technique described in [15] to produce highly polarized muonic helium atoms ( $\sim 44\%$  for  $^4\text{He}$ ). Muonic helium atoms are polarized by collision with alkali metal vapors using a spin-exchange optical pumping (SEOP) technique. This method is very similar to the one used in  $^3\text{He}$  neutron spin filters for neutron scattering experiments at J-PARC Materials and Life Science Experimental Facility (MLF) [16].

We have started a collaboration with a neutron group at MLF to develop an SEOP system dedicated to muonic helium atoms to confirm its repolarization using the conventional SEOP technique with pure Rb vapors as used in [15]. Furthermore, improvements can be expected using the more recent hybrid-SEOP technique with K/Rb vapors. Indeed, from SEOP experiments with  $^3\text{He}$  neutron spin filters, in a hybrid cell, K/Rb enhances the spin-exchange efficiency where Rb is used as a spin-transfer agent to K to prevent depolarization of Rb due to Rb-Rb collisions. Also, the spin-exchange collisions between K and He atoms transfer the angular momentum with much greater efficiency (nearly ten times) than directly Rb-He [17], and by optimizing the K/Rb vapor density ratios, high polarizing rates with high polarization can be achieved [18], which is very important for HFS measurements.

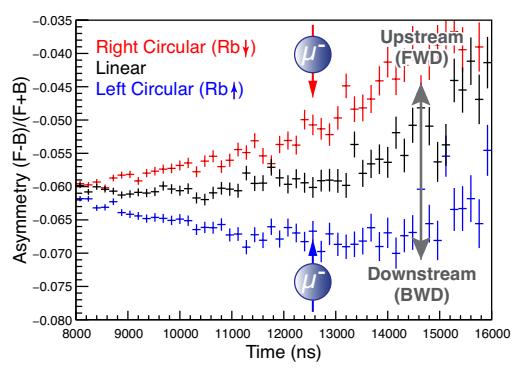
An experimental apparatus was designed to perform muonic helium repolarization measurements using the  $\mu\text{SR}$  spectrometer in the D1 area of MUSE D-line. A removable laser enclosure was fabricated that can be easily assembled around the  $\mu\text{SR}$  spectrometer to allow laser experiments to be performed safely. A dedicated laser diode array system operating at 795 nm ( $\sim 80$  W, CW) was built for optical pumping (Fig. 3). The laser light polarization can be adjusted by rotating the quarter-wavelength plate. The experimental arrangement installed inside the  $\mu\text{SR}$  spectrometer is shown in Fig. 4. Pulsed polarized negative muons ( $\sim 32$  MeV/c) are stopped into a glass cell containing  $^4\text{He}$  gas with few Rb or K/Rb vapors. A heating oven surrounding the target heat the glass cell to 150–250°C to vaporize the alkali metals, thereby increasing the gas pressure in the cell from 3 atm (room temperature) to about 5 atm. A laser diode array illuminates the cell from downstream and polarizes Rb atoms by optical pumping when the laser light is right- or left-handed circularly polarized. Optically pumped Rb atoms subsequently polarize K atoms via spin-exchange collision. Then, the outermost electron of the K/Rb atom is polarized. A longitudinal magnetic field of  $\sim 0.4$  mT (from the  $\mu\text{SR}$  spectrometer) provides the quantization axis for the K/Rb spins.



**Figure 3:** Laser arrangement for repolarizing muonic helium atoms using spin-exchange optical pumping. Rotating the quarter-wavelength plate makes right- or left-handed circularly polarized laser light.



**Figure 4:** Schematic view of the experimental arrangement for muonic helium SEOP measurements inserted in the  $\mu$ SR spectrometer at D1 area. The longitudinal coils provide the magnetic field for the spin quantization axis.



**Figure 5:** Asymmetry spectra obtained with a pure Rb cell at 180°C (preliminary). The muon spin direction changes when the laser light is right- or left-handed circularly polarized.

After the helium atom captures a muon and a  $(\mu^4\text{He})^+$  ion in its ground  $1s$  state is formed, it will capture an electron from K/Rb atoms to form a neutral muonic helium atom by charge exchange (no  $\text{CH}_4$  needed here). If the captured electron is polarized, then an electron-polarized muonic helium atom is formed. However, if the captured electron is unpolarized, it will subsequently be polarized by spin-exchange collision with K/Rb atoms. After a short-lived collision affecting only the valence electron spin, the polarization is shared with the pseudonucleus  $(\mu^4\text{He})^+$  via hyperfine interaction, thus polarizing the muon. The muon asymmetry, proportional to the muon polarization, is measured with the  $\mu$ SR spectrometer forward and backward detectors using longitudinal  $\mu^-$  SR measurements. Moreover, the muon polarization can be reversed by flipping the helicity of the laser light (right- or left-handed).

In our first muonic helium SEOP experiment in February 2023, we successfully observed muonic helium atoms being repolarized with their spin direction changing when the laser light circular polarization was right- or left-handed (Fig. 5). It should be noted that these asymmetry spectra should be first corrected for decay electrons from the background, mainly Si and O of the glass cell having different muon lifetimes and with amplitudes slightly different for forward and backward detectors before the muonic helium polarization time spectrum can be extracted. A detailed analysis is in progress. In this experiment, prototype cells made of pyrex glass and tested for muonic helium measurements [19] were used. The cell outer dimensions were  $\phi 75$  mm, 150 mm long, and 1.0 mm thick at the center of both ends. However, based on the time spectrum of decay electrons, it was estimated that more than 90% of the muons stopped inside the glass constituting the cell. The cell diameter was also too large compared to the laser beam size of  $\sim \phi 40$  mm.

In the following experiments, narrower and longer cells with thinner walls were fabricated from aluminosilicate glass (GE-180), which is resistant to corrosion by alkali metals. The new cell outer dimensions were  $\phi 50$  mm, 180 mm long, and 0.5 mm thick at the center of both ends. This maximized the laser overlapping region in the cell and more efficiently stopped negative muons, increasing the S/N ratio. The temperature dependence (160–240°C) of two cells, pure Rb (conventional SEOP) in December 2023 and K/Rb (hybrid SEOP) in May 2024, was studied in detail. Distinct effects were observed between both cells. With pure Rb, the asymmetry first increased and then decreased at higher temperatures, indicating a decline in polarization efficiency due to spin relaxation from Rb-Rb collision. Meanwhile, with K/Rb, the polarization efficiency remained, and the polarization rate kept increasing. These are promising results for the application of SEOP to HFS measurements. Muonium repolarization measurements were also obtained for a detailed comparison. The data analysis is in progress. In the next experiment in January 2025, the effect of the magnetic field on the spin-exchange rate will be measured to investigate whether the SEOP technique can be used in a high-magnetic field.

The next step will be to demonstrate that the SEOP technique can be applied to muonic helium HFS measurements. A new experimental apparatus will be conceived, combining a cell with a microwave cavity. Usually,  $^3\text{He}$  neutron spin filters use a glass cell to achieve a very long spin relaxation time ( $>100$  h). However, for muon experiments, Rb atoms can be polarized *in situ* only during or before the muon injection. Therefore, we are currently considering using a metal cell. Such a metal cell has already been developed for an experiment to polarize  $^3\text{He}$  and measure spin-dependent  $\mu^-$  capture reaction [20]. The main advantages are less background from high-Z nuclei due to muon capture (shorter lifetime) compared to Si and O of a glass cell. Also, the metal cell can be directly used as a microwave cavity, a higher pressure can be applied, and it can be reused. The challenges will be selecting materials that are not corroded by alkali metals, installing laser entry windows that are hermetically sealed and pressure resistant, and preventing damage due to heating.

Monte Carlo simulations were also performed. The polarization of the pseudonucleus ( $\mu^4\text{He}$ )<sup>+</sup> with electron-polarized Rb atoms is maximized in about 2  $\mu\text{s}$  after  $\mu^-$  injection with almost 100% polarization [21, 22]. The final polarization depends on the available laser power, cell geometry, and amount of alkali metal vapors. However, the gain seems to be reduced when the microwave is applied due to the competition between SEOP and microwave resonance. A pulsed laser system may be required to polarize Rb before muon injection. Further simulations are in progress.

#### 4. Summary and future plans

Muonic helium atom HFS measurements are of the utmost importance for testing and improving the theory of three-body atomic systems and bound state QED and for determining the fundamental constants of the negative muon magnetic moment and mass. New measurements at zero-field were already performed with a precision better than both previous experiments in the 1980s. Further measurements at higher pressure are planned at the D-line to improve the determination of the pressure shift (quadratic dependence). D-line is used under a user program, and obtaining a long measurement period is difficult. Measuring the HFS interval  $\Delta\nu$  at 1 ppm level could be achieved depending on the allocated beamtime. The high-field measurements are in preparation at the H-line

after muonium HFS measurements, using higher  $\mu^-$  intensity than at the D-line, decay electrons being more focused on the detectors due to the high magnetic field and longer measurement time, we aim to improve the determination of  $\Delta\nu$  and the negative muon magnetic moment and mass nearly 100 times from the previous measurement [5]. The investigation of repolarizing muonic helium atoms using the SEOP technique is progressing smoothly. This could drastically improve the measurement precision by nearly 1 order of magnitude if successful. Entirely new systematics will arise that will require careful study.

## Acknowledgements

The muon experiment at the Materials and Life Science Experimental Facility of J-PARC was performed under a user program (Proposals No. 2020B0333, 2021B0169, 2022A0159, 2022B0314, 2023A0317, 2024A0075). This work was supported by the JSPS KAKENHI Grant No. 21H04481.

## References

- [1] P. Strasser, S. Shimomura and H. A. Torii, JPS Conf. Proc. **21**, 011045 (2018).
- [2] P. Strasser *et al.*, Phys. Rev. Lett. **131**, 253003 (2023).
- [3] V. W. Hughes and G. zu Putlitz, in *Quantum Electro-dynamics*, ed. T. Kinoshita (World Scientific, Singapore, 1990), pp. 822–904; refer to subsection VII - Muonic Helium Atom, pp. 892.
- [4] H. Orth *et al.*, Phys. Rev. Lett. **45**, 1483 (1980).
- [5] C. J. Gardner *et al.*, Phys. Rev. Lett. **48**, 1168 (1982).
- [6] S. Kanda *et al.*, Phys. Lett. B **815**, 136154 (2021).
- [7] S. Nishimura *et al.*, Phys. Rev. A **104**, L020801 (2021).
- [8] K. S. Tanaka *et al.*, Prog. Theor. Exp. Phys. **2021**, 053C01 (2021).
- [9] P. A. Souder *et al.*, Phys. Rev. A **22**, 33 (1980).
- [10] H. Orth, Hyperfine Interact. **19**, 829 (1984).
- [11] D. J. Arseneau *et al.*, J. Phys. Chem. B **120**, 1641 (2016).
- [12] W. Liu *et al.*, Phys. Rev. Lett. **82**, 711 (1999).
- [13] D. T. Aznabayev *et al.*, Phys. Part. Nucl. Lett. **15**, 236 (2018).
- [14] V. Patkóš *et al.*, Phys. Rev. Lett. **131**, 183001 (2023).
- [15] A. S. Barton *et al.*, Phys. Rev. Lett. **70**, 758 (1993).
- [16] T. Ino *et al.*, J. Phys.: Conf. Ser. **862**, 012011 (2017).

- [17] E. Babcock *et al.*, Phys. Rev. Lett. **91**, 123003 (2003).
- [18] W. C. Chen *et al.*, Phys. Rev. A **75**, 013416 (2007).
- [19] T. Ino *et al.*, JPS Conf. Proc. **37**, 021208 (2022).
- [20] P. A. Souder *et al.*, Nucl. Instum. Meth. A **402**, 311 (1998).
- [21] S. Fukumura *et al.*, EPJ Web Conf. **262**, 01012 (2022).
- [22] S. Fukumura *et al.*, Physics **2024**, 6, 877 (2024).