New precise spectroscopy of the hyperfine structure in muonium with a high-intensity pulsed muon beam

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ABSTRACT

A hydrogen-like atom consisting of a positive muon and an electron is known as muonium. It is a near-ideal two-body system for a precision test of bound-state theory and fundamental symmetries. The MuSEUM collaboration performed a new precision measurement of the muonium ground-state hyperfine structure at J-PARC. The resonance of hyperfine transition was successfully observed, and the muonium hyperfine structure interval of \( v_{\text{HFS}} = 4463302(4) \) GHz was obtained with a relative precision of 0.9 ppm. The result was consistent with the previous ones obtained at Los Alamos National Laboratory and the current theoretical calculation. We present a demonstration of the microwave spectroscopy of muonium with a high-intensity pulsed muon beam and a high-rate capable positron counter.

1. Introduction

Muonium is a bound-state of a positive muon and an electron, which was discovered by V. W. Hughes et al. [1]. In the standard model of particle physics, muonium is a two-body system of structureless leptons. Precision measurement of the hyperfine structure (HFS) of muonium provides one of the most rigorous validations of bound-state quantum electrodynamics (QED) theory, and the most precise method to determine the muon-to-electron mass ratio at present [2].

The muon-to-electron mass ratio is one of the essential parameters to determine the muon anomalous magnetic moment \( g_\mu \), which is known for a discrepancy between an experimental result [3] and theoretical calculations [4, 5, 6]. To examine the discrepancy more precisely, a new experiment at Fermi National Accelerator Laboratory (FNAL) is underway [7], and another one at Japan Proton Accelerator Research Complex (J-PARC) is in preparation [8]. The uncertainty of the \( g_\mu \) resulting from the muonium HFS is 30 ppb out of 540 ppb. This uncertainty is comparable to the major systematic uncertainties expected in the new experiments. Hence, improving the measurement precision of the muonium HFS is important for both experiments at FNAL and J-PARC.

Systems containing second-generation particles amenable to precise spectroscopy are very limited, and thus muonium plays a unique role in the search for physics beyond the standard model and tests of lepton universality. Spectroscopy of the muonium HFS can test the Lorentz invariance via the measurement of a sidereal oscillation [9]. By comparing an experimental result with the theoretical prediction, hypothetical new particles can be searched for [10, 11]. Theory predicts the muonium HFS in the ground-state at \( v_{\text{HFS}} = 4463.302867(271) \) MHz [4]. The uncertainty of the calculation is dominated by measurement precision of the muon mass, which accounts for 253 Hz out of 271 Hz.[1]

From the 1970s to the 1990s, the muonium HFS was measured at the Nevis synchrocyclotron and the Los Alamos Meson Physics Facility (LAMPF). Previous experiments were performed in two ways: direct measurement in a near-zero magnetic field and indirect measurement in a high magnetic field. In the latter, the muonium HFS was determined by using the transition frequencies between Zeeman sub-

\[ h \]

[1] A recent article points out an underestimation of the theoretical uncertainty [12]. The uncertainty given in the reference is 515 Hz, almost double the estimation in Ref. [4].
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levels in a magnetic field. The most precise results for each method were \( \nu_{\text{HFS}} = 4463.3022(1.4) \text{ MHz} \) for direct measurement [13], and \( \nu_{\text{HFS}} = 4463.302765(53) \text{ MHz} \) for indirect measurement [14].

The previous Nevis experiments were performed using a continuous muon beam incoming at random timing. The muon stopping and subsequent emission of decay positron were detected by the scintillation counters to measure the time of events. To measure the time difference between muon stopping and positron emission correctly, only one muon per time window of a few microseconds was allowed by the data-acquisition electronics. Therefore, the measurement precision was statistically limited strictly. In the latest experiment at LAMPF, a continuous muon beam was chopped by an electric-field kicker to separate the measurement time window. Approximately 70% of the beam was lost due to this chopping, so that statistics limited the measurement precision.

To exceed the limits of previous experiments and realize spectroscopy with higher precision, we proposed a new experiment using a high-intensity pulsed muon beam at J-PARC [15]. In contrast to an experiment using a continuous beam, no muon trigger is required because the arrival of the beam is synchronized to the accelerator repetition. Bunches of muons are periodically injected, and the Rabi oscillation of muonium is observed as an ensemble average over muonium atoms.

At the Materials and Life Science Experimental Facility (MLF) of J-PARC, the Muon Science Establishment (MUSE) facility delivers the world’s highest-intensity pulsed muon beam [16]. However, its benefit involves difficulties in positron counting due to the high instantaneous event rate. The novelty and significance of the experiment described in this paper are the application of the high-intensity pulsed muon beam to precise spectroscopy using a high-rate capable particle detector.

2. Theory

The HFS \( A = \hbar \nu_{\text{HFS}} \) of muonium in the ground-state is the energy splitting between the spin-triplet state and the spin-singlet state. The Hamiltonian of muonium in a magnetic field is described as

\[
\mathcal{H} = A S_z \cdot S_e + (g'_{\mu} \mu_B S_e - g'_{\mu} \mu_B S_\mu) \cdot B,
\]

where \( S_\mu \) is the spin operator of muon or electron \( (l = \mu, e, \text{ the same shall apply hereinafter}) \), \( g'_\mu \) is the bound-state g-factor in muonium, \( \mu_B = e\hbar / 2m_l \), \( m_l \) is the mass, and \( B \) is the external magnetic field.

Microwave irradiation at an appropriate frequency excites muonium from the singlet-state to the triplet-state. The associated time-dependent Hamiltonian is represented as

\[
\mathcal{H}_t(t) = (g'_{\mu} \mu_B S_e - g'_{\mu} \mu_B S_\mu) \cdot B_1 \cos \omega t.
\]

Here \( B_1 \) is the magnetic field of the applied microwave, and \( \omega \) is its angular frequency.

When the external magnetic field is sufficiently weak, the energy eigenstates of muonium are classified by the total angular momentum \( F \) and the associated magnetic quantum number \( m_F \) as \((\psi_1, \psi_2, \psi_3, \psi_4) = (\{1, 1\}, \{1, 0\}, \{1, -1\}, \{0, 0\}) \), where the first and the second number indicates \( F \) and \( m_F \), respectively. The Hamiltonian based on the energy eigenfunctions of muonium is explicitly written as

\[
\mathcal{H}' = \mathcal{H} + \mathcal{H}_t(t) = \hbar \begin{pmatrix}
\Omega_L & 0 & 0 & 2\Omega_R \cos \omega t \\
0 & 0 & 0 & 0 \\
0 & 0 & -\Omega_L & -2\Omega_R \cos \omega t \\
2\Omega_R \cos \omega t & 0 & -2\Omega_R \cos \omega t & -2\pi \nu_{\text{HFS}}
\end{pmatrix},
\]

where \( \Omega_L \) is the Larmor frequency, \( B \) is the static field strength along the \( z \)-axis, \( \Omega_R \) is the Rabi frequency, and \( B_1 \) is the microwave field strength. The Rabi frequency describes the time evolution of the muon spin polarization. In the experiment, the muon spin polarization is observed as an ensemble average over muonium atoms via the decay positron asymmetry. To obtain a theoretical expression of the signal, calculation of the state amplitudes using the density matrix for a statistical mixture of muonium states is necessary. The theoretical expression of the Rabi oscillation and the resonance curve are obtained in the references [17, 18, 19].

The Rabi oscillation at a certain microwave field strength is written as follows,

\[
S(t) = \left[ \frac{\Gamma + \Delta \omega}{\Gamma} \cos \frac{\Gamma - \Delta \omega }{2} t + \frac{\Gamma - \Delta \omega}{\Gamma} \cos \frac{\Gamma + \Delta \omega }{2} t \right] e^{-\beta t}.
\]

where \( \Delta \omega = \omega - 4463.302867 \times 2\pi \text{ MHz} \) is the microwave frequency detuning, \( s \) is the scaling factor depending on the acceptance of the positron detector and the minimum energy of detected positrons, and \( \beta \) is the damping constant, which represents muon spin depolarization. Since the strength of the microwave field is position-dependent, the spin-precession signal is observed as the sum of multiple oscillation components. The time-integration of the oscillations yields the resonance curve as a function of the varying microwave frequency.

3. Experiment

The experiment was conducted at J-PARC MLF MUSE D-Line. A pulsed 3 GeV proton beam was injected into a graphite target, and pions were produced by hadronic interactions. Decay of pion at rest on the target surface yielded
spin-polarized positive muon ($\mu^+$). A muon beam having a momentum of 27.4 MeV/c irradiated krypton gas at a pressure of 1 bar to form muonium atoms after muon thermalization in the gas target. The beam intensity was $2 \times 10^6 \, \mu^+/s$ with the accelerator operation power of 200 kW. The beam was pulsed and repetitive at 25 Hz, resulting in $8 \times 10^4 \, \mu^+/\text{pulse}$. The momentum spread of the beam was $\pm 5\%$.

The initial state population of muonium is statistically distributed in the spin-singlet state (25\%) and the spin-triplet states (75\%). Irradiation of microwave induces transitions between the states. This hyperfine-state transition causes muon spin precession. The time evolution of the muon spin was observed via the angular asymmetry of positrons from muonium decays. A segmented plastic scintillation counter detected the decay positrons. Figure 1 illustrates the experimental setup.

![Figure 1](image_url)

**Figure 1:** Cutaway drawing of the experimental apparatus: (1) three-layers of magnetic shield, (2) the cylindrical gas chamber made of aluminum, (3) the cylindrical microwave cavity made of copper, (4) the aluminum absorber for background suppression, (5) the segmented positron counter. The inner axial length of the gas chamber and the cavity was 450 mm and 230 mm, respectively.

Krypton gas with the purity of 99.999\% was confined in a cylindrical aluminum vessel with an inner diameter of 280 mm. The gas pressure was measured by a capacitance gauge (ANELVA M-342DG) with 0.2\% accuracy. The upstream end of the chamber has a thin aluminum beam window with a thickness of 100\,\mu m and a diameter of 100 mm. At the beam window, the muon beam profile was a two-dimensional Gaussian with a standard deviation of 2 cm (1\sigma).

A cylindrical cavity made of oxygen-free copper with an inner diameter of 81.8 mm was used to apply microwaves to the muonium atoms. The microwave resonated in TM110 mode with a quality factor of 5000 at 4463.302 MHz. The quality factor was frequency-dependent, smoothly decreasing from 7000 to 4000 as the frequency increased from 4462 MHz to 4465 MHz. The microwave from a signal generator (Hewlett-Packard 8671B) was input to the cavity through amplifiers (Mini Circuit ZVE-8G). The microwave power was monitored by a thermal power sensor (Rohde&Schwarz NRP-Z51), and typical input power was about 0.7 W. The resonance frequency was tuned by moving an aluminum rod inserted into the cavity with a piezoelectric actuator (attocube ANPz101eXT12). The adjustable frequency range was 4463 ± 1.5 MHz.

A Monte-Carlo simulation using GEANT4 toolkit [20, 21, 22] was performed, and the fraction of muon stopping in the cavity was estimated to be 30\% of the total incident.

A three-layer box-shaped magnetic shield made of an alloy of iron and nickel was used against the geomagnetic field and the static magnetic field generated by surrounding devices. The three-dimensional magnetic field distribution in the cavity was measured by a fluxgate probe (MTI FM3500) with 0.5 nT resolution. The static magnetic field inside the cavity was less than 60 nT.

The segmented positron counter consisted of an array of plastic scintillator tiles and silicon photomultipliers (SiPMs) [23]. Figure 2 depicts the positron counter. The detector had two layers 4 cm apart, consisting of 24-by-24 scintillator tiles. A SiPM (Hamamatsu Photonics MPPC S12825-050P-01) having an active area of 1.3 mm square was connected to the center of each scintillator (Eljen Technology EJ-212). The scintillator tiles of 1 cm square and 3 mm thick were two-dimensionally arranged. Reflector films (3MESR) were inserted between tiles. The upstream layer of the detector was placed 20 cm away from the downstream end of the cavity.

![Figure 2](image_url)

**Figure 2:** Drawing of the positron counter: (a) enlarged view of the scintillator tile and SiPM, (b) overall view seen from the beam, (c) view from the side.

The signals from the SiPMs were processed by the Kalliope front-end electronics consisting of an ASIC-based amplifier-shaper-discriminator and a multi-hit time-to-digital converter implemented in FPGA\(^4\), where the leading edge time was recorded [24]. Photon yield of a positron from muon decay was represented by a Landau distribution.
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with a peak at 55 photons\(^5\). The threshold for the discriminator was set at 1.5 photon equivalent (p.e.) level so that the positron detection efficiency was almost 100%. The typical dark count rate of each SiPM at 1.5 p.e. threshold was 17 kHz. The time resolution of the detector was 8 ns (1\(\sigma\)). A Monte-Carlo simulation estimated that about 1% of the decay positrons from muonium decays were detected.

A large number of prompt positrons from the muon production target and positrons from muon decay during transport were incident on the apparatus. The momentum of these background positrons was similar to that of the transported muons, 27.4 MeV/c. To prevent these positrons from causing background events, an aluminum plate with a thickness of 40 mm was placed between the target chamber and the detector. This plate served as an absorber to block positrons with momentum below 40 MeV/c. The background events were suppressed by a factor of five. In addition, the energy threshold selected the positrons emitted preferentially along the muon spin direction. The loss due to the absorber was estimated by a GEANT4 simulation to be 40%.

When the microwave field induces the Rabi oscillation, the muon spin polarization changes with time, and it is observed as an oscillation in the detection time spectrum of the decay positrons. The spin-precession signal was extracted by taking the ratio of the spectra with and without the microwave field to suppress the microwave independent systematic uncertainties. Microwave switching was repeated at several-minute intervals to reduce the thermal load due to power absorption in the cavity.

4. Analysis

Data for 24 hours of beam time was analyzed. In data analysis, the background events due to dark counts of SiPMs were suppressed by selecting coincidence events detected at the same time in the two detector layers. The time window of the coincidence analysis was set to 24 ns, which corresponded to three times the time resolution. Simultaneous hits in adjacent segments on the layer were merged as a hit-cluster having the same origin. The number of coincidence events per beam pulse was about 110, which was consistent with the expectation considering pileup counting loss described later.

A bunch of pulsed muons makes multiple hits on the detector simultaneously. The simultaneous overlap of multiple positrons causes signal counting loss. This pileup event occurs more frequently when the instantaneous count rate is higher. Figure 3 shows the time spectrum.

In an ideal situation without pileup counting loss, the spectrum is exponential with the muon mean lifetime. The effect of the pileup was evaluated by taking the difference between the observed spectrum and the extrapolated fitting result obtained in the low rate region, where pileup loss is negligible. Figure 4 shows the relative efficiency considering pileup loss. The measurement result is well explained by a pulse-height analyzer (PHA) windowing model [25]. The dead-time of the detector obtained by the fitting analysis was 500 ns, which was consistent with the analog signal observations. The counting loss due to pileup was about 20% of the total detection.

The Rabi-oscillation signal was obtained by taking the ratio between times spectra with and without microwave irradiation. Figure 5 shows the result with the microwave frequency of 4463.302 MHz.

Figure 3: Time spectrum of the number of decay positrons and background events without microwave irradiation. The ordinate is normalized by the incident of the muon beam pulse. The black solid curve shows the fitting result with an exponential function on a constant background. The fitting exponent gives the muon lifetime of 2198(5) ns. The red dashed line indicates extrapolation of the fitting function.

Figure 4: Pileup counting loss as a function of the instantaneous event rate. The black curve indicates the result of fitting with the model function [25].

\(^{5}\text{SiPM is an array of avalanche photodiodes (APDs), and the output signal is discretized by the number of APDs that detected photon.}\)
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Figure 5: The Rabi oscillation of muonium under the microwave field with a frequency of 4463.302 MHz. The solid curve shows the fitting result with the theoretical expression of the oscillation. In the fitting function, the frequency detuning was fixed at zero. The reduced chi-square is $\chi^2/NDF = 50/45$, which gives the $p$-value of 0.28.

Figure 6: Result of the frequency scan measurement. The upper panel shows the resonance curve. The vertical axis corresponds to the time integration of the Rabi-oscillation signal. The horizontal axis represents the frequency detuning from 4463.302 MHz. The solid curve shows the fitting result with a Lorentz function. The reduced chi-square is $\chi^2/NDF = 16.9/15$, which gives the $p$-value of 0.32. The normalized fitting residuals are shown in the lower panel.

Table 1
Systematic uncertainties in the experiment.

| Source                      | Contribution (Hz) |
|-----------------------------|-------------------|
| Gas pressure measurement    | 46                |
| Microwave power             | 37                |
| Detector pileup             | 19                |
| Static magnetic field       | negligible        |
| Gas pressure fluctuation    | 6                 |
| Gas impurity buildup        | 12                |
| Muon beam intensity         | negligible        |
| Muon beam profile           | negligible        |
| Total                       | 63                |

Figure 5: The Rabi oscillation of muonium under the microwave field with a frequency of 4463.302 MHz. The solid curve shows the fitting result with the theoretical expression of the oscillation. In the fitting function, the frequency detuning was fixed at zero. The reduced chi-square is $\chi^2/NDF = 50/45$, which gives the $p$-value of 0.28.

5. Result

The time-integration of the Rabi-oscillation signal yields the spin-precession signal at a particular frequency. The resonance curve is obtained by sweeping the microwave frequency. Figure 6 shows the resonance curve as a result of the experiment. The frequency dependence of the cavity quality factor was corrected. Pressure-dependent frequency shift due to atomic collisions [26] was corrected for using the past experimental result, which amounted to the shift of 36 kHz at the krypton gas pressure of 1 bar [19]. The resonance frequency was determined by fitting the curve with a Lorentz function. The analysis gave the muonium HFS of

$v_{\text{HFS}} = 4463.302(4) \text{ MHz}$, 

where the uncertainty is statistical. The result was consistent with the theoretical prediction and the previous experimental results with the continuous muon beams. No significant systematic deviation from the fitted curve was observed.

The systematic uncertainties in the experiment are summarized in Table 1. With the present apparatus, the systematic uncertainty was dominated by the uncertainty of the gas pressure, making the correction of the pressure shift ambiguous (46 Hz). The second-largest contribution was due to the instability of the microwave power (37 Hz). Both systematic uncertainties were far less than the statistical uncertainty (4 kHz). In a future experiment, quantitative evaluation of the systematics will be essential after the completion of a brand-new beamline construction that enables long-term measurement [27].

6. Conclusion

A new precision measurement of the muonium HFS using the high-intensity pulsed muon beam was performed at J-PARC MLF MUSE. Measurement principle was proven...
under a precisely-controlled near-zero magnetic field. The segmented positron counter was employed to maximize the advantage of the high-intensity beam. This measurement is a milestone for a future long-term measurement with a further enhanced beam-intensity to surpass the results of the previous experiments.

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