High precision measurement of muonium hyperfine structure

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Abstract. MuSEUM (Muonium Spectroscopy Experiment Using Microwave) collaboration aims to measure the muonium hyperfine structure (MuHFS, $\nu_{\text{HFS}}$) with a few ppb (parts per billion). MuHFS spectroscopy is a stringent test of the bound-state QED. From this measurement, the muon-proton magnetic moment ratio ($\mu_{\mu}/\mu_{p}$) and the muon-electron mass ratio ($m_{\mu}/m_{e}$) can also be determined by applying a high magnetic field. In the previous MuHFS measurement carried out at Los Alamos Meson Physics Facility (LAMPF), the main uncertainty was caused by the lack of the statistics. MuSEUM collaboration uses the high intense pulsed muon beam at Japan Proton Accelerator Research Complex (J-PARC) to improve the statistical uncertainty. We are also developing our own experimental setups for the improvement of the systematic uncertainties. From June 2016, MuSEUM collaboration have carried out the MuHFS measurement in an extremely low magnetic field within 100 nT and now we are preparing for the measurement in a 1.7 T high magnetic field.
1. Introduction

Muonium is a bound state of a positive muon and an electron. As muonium has the lifetime of $2.2 \mu$s which is sufficiently long, the measurement of its hyperfine structure (MuHFS) can be performed with high precision. And also muonium can be regarded as a hydrogen-like atom free from the finite size effect of the nucleon, the theoretical calculation can be performed with high precision. Therefore by comparing the experimental and theoretical precision, the MuHFS is a good probe to test the bound-state QED. The hyperfine structure of a hydrogen is measured most precisely by MASER spectroscopy with 2.1 ppt (parts per trillion) [1], however the precision of the theoretical calculation is limited to 1.2 ppm (parts per million) [2] because of the internal structure of the proton.

About the MuHFS, both the theoretical calculation and the experimental results can be decided with high precision. The theoretical value of the MuHFS is $\nu_{\text{HFS}} = 4.463\,302\,891(272)$ GHz [3], determined with 61 ppb (parts per billion) precision. The main uncertainty comes from the experimental precision of the muon-to-electron mass ratio ($m_\mu/m_e$).

The muonium hyperfine structure is measured by the spectroscopy of the muon and the electron spin states. The Hamiltonian of the ground state muonium is expressed as

$$H = h \nu_{\text{HFS}} s_\mu \cdot s_e - \mu_\mu g'_\mu s_\mu \cdot B + \mu_e g'_e s_e \cdot B$$

(1)

where $h$ is the Planck constant, $\nu_{\text{HFS}}$ is the frequency of the muonium hyperfine structure, $\mu_\mu$ ($\mu_e$) is the muon (electron) magnetic moment, $g'_\mu$ ($g'_e$) is the bound g-factor of the muon (electron) in the muonium, and $s_\mu$ ($s_e$) is the spin of the muon (electron). When $B = 0$, the muonium hyperfine structure is determined simply by measuring the energy level between the spin singlet and triplet. When $B$ is non-zero, the spin triplet split to three sublevels, which is called the Zeeman effect. Figure 1 shows the Breit Rabi diagram of the muonium $1S_{1/2}$ state.

**Figure 1.** The Breit-Rabi energy level diagram of the muonium $1S_{1/2}$ state in magnetic field. The vertical axis is the energy level $W$ normalized by the hyperfine energy shift $h\nu_{\text{HFS}}$. The horizontal axis $x$ is a dimensionless parameter which is proportional to the external magnetic field $H$ (see equation (6)). $M_\mu$ and $M_e$ are the spin states of muons and electrons. The state 1 to 4 are distinguished from $(M_\mu, M_e)$. $\nu_{12}$ ($\nu_{34}$) is the spin transition frequency between state 1 and 2 (3 and 4).

The most precise measured values of the MuHFS are

$$\nu_{\text{HFS}}(ZF) = 4.463\,302\,2(14)\ \text{GHz (0.3 ppm)}$$

(2)

$$\nu_{\text{HFS}}(HF) = 4.463\,302\,765(53)\ \text{GHz (12 ppb)}$$

(3)
where $\nu_{\text{HFS}}(\text{ZF})$ is from the measurement with an extremely low magnetic field [4] and $\nu_{\text{HFS}}(\text{HF})$ is from the measurement with a high magnetic field [5]. Both experiments were conducted at Los Alamos Meson Physics Facility (LAMPF). The high precision MuHFS spectroscopy will also contributes to other physics, such as exotic particle search [6][7], proton radius puzzle [8][9], and test of CPT or Lorentz invariance [10].

1.1. Muon-proton magnetic moment ratio

In the MuHFS measurement with high magnetic field, muon-proton magnetic moment ratio can be derived from the transition frequencies between the Zeeman sublevels $\nu_{12}$ and $\nu_{34}$. They are expressed as

$$\nu_{12} = -\frac{\mu_{\mu} g'_{\mu} H}{\hbar} + \frac{\nu_{\text{HFS}}}{2} [(1 + x) - \sqrt{1 + x^2}]$$

(4)

$$\nu_{34} = +\frac{\mu_{\mu} g'_{\mu} H}{\hbar} + \frac{\nu_{\text{HFS}}}{2} [(1 - x) + \sqrt{1 + x^2}]$$

(5)

$x$ is a dimensionless quantity proportional to the magnetic field $H$ expressed as

$$x = \frac{(g'_{\mu} e + g_{\mu} \mu_{\mu}) H}{\hbar \nu_{\text{HFS}}}$$

(6)

$H$ can be measured with the nuclear magnetic resonance (NMR) precession frequency of a free proton $\nu_{p}$ as $h \nu_{p} = 2 \mu_{p} H$. Including the proton NMR frequency and in the limit of strong magnetic field ($x \gg 1$), $\nu_{12}$ and $\nu_{34}$ become

$$\nu_{12} = -\frac{g'_{\mu} \mu_{\mu} \nu_{p}}{\mu_{p}} + \frac{\nu_{\text{HFS}}}{2}$$

(7)

$$\nu_{34} = +\frac{g_{\mu} \mu_{\mu} \nu_{p}}{\mu_{p}} + \frac{\nu_{\text{HFS}}}{2}.$$  

(8)

Therefore, the muonium hyperfine structure can be measured by $\nu_{\text{HFS}} = \nu_{12} + \nu_{34}$ and the muon-proton magnetic moment ratio is derived from $\nu_{12}$, $\nu_{34}$ and $\nu_{p}$ as

$$\frac{\mu_{\mu}}{\mu_{p}} = \frac{1}{g'_{\mu}} \frac{\nu_{34} - \nu_{12}}{\nu_{p}}$$

(9)

in the limit of strong magnetic field. This value was measured at LAMPF with 120 ppb precision as $\mu_{\mu}/\mu_{p} = 3.183 \ 345 \ 24(37)$ [5].

The muon-proton magnetic moment ratio is essential for the measurement of the muon anomalous magnetic moment $a_{\mu} = (g_{\mu} - 2)/2$, or muon $g - 2$. It is known that the theoretical and the experimental values have about 3.6$\sigma$ discrepancy [11][12]. $a_{\mu}$ is measured by a muon storage ring and derived as follows,

$$a_{\mu} = \frac{R}{\lambda - R} (R = \frac{\omega_{\mu}}{\omega_{p}}, \lambda = \frac{\mu_{\mu}}{\mu_{p}})$$

(10)

where $R$ is the muon-proton spin precession frequency ratio decided by the muon storage ring experiment, and $\lambda$ is the muon-proton magnetic moment ratio decided by other experiments. From the previous results, $\lambda$ is quoted from the MuHFS measurement. At the measurement at Brookhaven National Laboratory (BNL) [11], the precision of $\lambda$ was derived from the quotation of the $\nu_{\text{HFS}}$ 12 ppb result [5] and estimated as $\lambda = 3.183 \ 345 \ 13(39) \ (30 \ \text{ppb})$, instead of using the measured value with 120 ppb precision [5]. This extrapolation is based on a assumption that the theoretical QED calculation is correct. However, considering the discrepancy of $a_{\mu}$, it is important to determine $\lambda$ only by the measurement. Therefore, the results of high magnetic field MuHFS measurement is essential to the muon anomalous magnetic moment experiment.
1.2. Muon-electron mass ratio

Muon-electron mass ratio can also be derived from the HF MuHFS measurement as

\[ \frac{m_\mu}{m_e} = \left( \frac{g_\mu}{g_e} \right) \left( \frac{\mu_\mu}{\mu_p} \right) \left( \frac{\mu_e}{\mu_p} \right)^{-1}. \]  

(11)

The major uncertainty of this formula is caused by the measured value of the \( \mu_\mu/\mu_p \). Therefore the precision of the muon-electron mass ratio is determined as \( m_\mu/m_e = 206.768 \pm 0.277 \) (24) (120 ppb) from the LAMPF measurement [5].

\( m_\mu/m_e \) is measured in other methods. From the muonium 1S–2S interval measurement by the Doppler-free two-photon laser spectroscopy, \( m_\mu/m_e \) was extracted as 206.768 ± 0.38 (17) (822 ppb) [13]. However, as the most precise result comes from the MuHFS measurement at present, this will be the most stringent test of the bound state QED.

2. Experimental method

Figure 2 shows the setup of the MuSEM experiment. The MuSEUM experiment utilizes the pulsed muon beam line of Japan Proton Accelerator Research Complex (J-PARC), Materials and Life science experimental Facility (MLF), MUon Science Establishment (MUSE). We use the surface muon beam which muon spins are almost 100% polarized and The initial muon momentum is 29.8 MeV/c.

The positive muon (\( \mu^+ \)) beam is injected into a cylindrical aluminum krypton gas chamber. Inside the gas chamber, a muon forms into muonium by capturing an electron from the krypton gas. The krypton noble gas is used because of the low energy threshold for muonium formation, which leads to the high muonium production efficiency [14]. The gas pressure is controlled to 1.0 atm or below. Also the purity of the krypton gas is ensured to avoid the muonium depolarization due to the electron spin exchange interaction with oxygen after the muonium formation [15].

A copper RF cavity is placed inside the gas chamber. The resonance frequency of the cavity is tuned to the spin transition frequency and 1 W microwave power can be applied. When the microwave matches to the spin transition frequency, the muon spin flips to the downstream side and the decay positron is likely to hit the backward positron counter. The positron counter is formed by segmented plastic scintillator, silicon photomultiplier, and the fast read out electronics, which is designed to utilize in the condition at high intense pulsed muon beam [16]. By sweeping the RF drive and counting the the number of positrons, one can determine the HFS resonance frequency.

1.7 T magnetic field is applied from a superconducting solenoid magnet. In the MuHFS measurement with an extremely low magnetic field, the superconducting magnet will be substituted with permalloy magnetic shields to suppress the backgrounds magnetic field in the experimental area.
2.1. MuHFS experiment in an extremely low magnetic field

The MuSEUM collaboration is now performing the MuHFS measurement with an extremely low magnetic field. The magnetic field is suppressed below 100 nT by the three layer permalloy magnetic shield. Figure 3 is the observed resonance curve of the MuHFS measurement. The details of the analysis and the first result are explained at [17]. From this measurement, the systematic uncertainties due to the gas pressure, temperature and impurity, RF power and muon beam distribution are considered, and these studies will give feedback to the high field MuHFS measurement.
2.2. MuHFS experiment with high magnetic field

The MuSEUM collaboration is planning a high-field MuHFS measurement in future and the study for the high magnetic field measurement is ongoing. The list of uncertainties in the previous research [5] are shown in table 1. The main uncertainty was caused by the lack of the statistics. Their experiment was performed at LAMPF using a continuous (DC) muon beam being chopped to use the “old muonium method” [18]. The old muonium method is a method that uses only the long-lived muons for the analysis to make the resonance linewidth narrower, which is generally constrained by the muon lifetime of 2.2µs. We are planning to improve the systematic uncertainty by using the new high-intensity pulsed muon beamline, H-line, which will soon be constructed at J-PARC [19]. With 100 days of measurement, the number of muons available for the MuSEUM experiment will be about 10^{15}. This improves the statistical uncertainty by a factor of ten from the previous measurement at LAMPF [5], which used about 10^{13} muons for statistics. As pulsed muons already have a time structure, the “old muonium method” can be easily used by setting a time window in the analysis program.

The systematic uncertainty caused by the magnetic field is the second largest part. At the muonium spectroscopy with high magnetic field, spin transition frequencies ν_{12} and ν_{34} are measured. The MuHFS is determined by ν_{HFS} = ν_{12} + ν_{34} and from the equations (4) and (5), the term of the magnetic field is cancelled out. Therefore the systematic uncertainty caused by the magnetic field was considered as zero for the MuHFS at the previous research [5]. However, μ_µ/μ_p is proportional to ν_{34} − ν_{12} and the term of the magnetic field remains. Therefore the inhomogeneity of the magnetic field relates to the systematic uncertainty. To improve this uncertainty, ameliorating the magnetic field homogeneity is critical to the experiment. The development of the MRI magnet and the NMR probes for the improvement of magnetic field are described in the next section.

As the resonance frequencies are different at zero field and high field, the design of the RF cavity is also different. The cavity for zero-field measurement uses the TM110 (or TM220) mode to resonate at ν_{HFS} = 4.463 GHz. While for the high-field measurement at 1.7 T magnetic field, the RF cavity is designed so that the TM110 mode resonates at ν_{12} = 1.906 GHz and the TM210 mode resonates at ν_{34} = 2.556 GHz, respectively.

| Table 1. The major uncertainties of the previous MuHFS experiment with 1.7 T [5] |
|---------------------------------|----------|--------|
|                                  | MuHFS [ppb] | μ_µ/μ_p [ppb] |
| Statistics                       | 10.9      | 107    |
| Magnetic field                   | 0         | 56     |
| Gas pressure/impurity            | 4.4       | 11     |
| Muon stopping distribution       | 1.0       | 13     |
| RF power                         | 0.96      | 9.6    |
| **Total**                        | **12**    | **120**|

3. Requirements of the magnetic field and the field detection system

The magnetic field inhomogeneity can be classified to two factors, the spacial inhomogeneity of the magnetic field applied by the magnet, and the magnetic field fluctuation per time. These inhomogeneities in the region where the muonium formed are crucial. Therefore, to improve the systematic uncertainty related to magnetic field, the requirements in our measurement are as follows.

- 1 ppm magnetic field homogeneity in the muonium formation volume (z = 300 mm, r = 100 mm ellipsoid).
• magnetic field stability below 0.1 ppm/h drift.
• 10 ppb order sensitivity for NMR magnetometer.

The former two requirements are fulfilled by the development of the superconducting solenoid magnet and the passive shimming method. To evaluate the magnetic field, high precision NMR magnetometers are also required. In MuSEUM collaboration, continuous wave NMR (CW-NMR) method is used for the magnetometer.

### 3.1. Evaluation of the MRI magnet

The MuSEUM collaboration possess a superconducting MRI magnet (Hitachi, Ltd.) with a maximum magnetic field of 2.9 T that will be operated at 1.7 T. The muonium formed volume will be at the center of the magnet, and the magnetic field will be measured at the surface of that volume. To improve the magnetic field homogeneity inside the magnet, passive shimming method is used. This method uses the insertion of iron shim plates at designated positions inside the superconducting magnet according to the singular value decomposition SVD calculations and improves the homogeneity of the magnetic field [20]. As a result of the passive shimming method, the magnetic field homogeneity at the muon formed ellipsoid surface \((z = 300 \text{ mm}, r = 100 \text{ mm})\) was measured within 0.8 ppm peak-to-peak. This magnetic field mapping was obtained by measuring 576 points with a single NMR probe, including uncertainties of the magnetic field drift during the measurement and the probe misalignment.

The ideal situation for the measurement is that the magnetic field is identical during \(\nu_{12}\) and \(\nu_{34}\) measurements. Therefore the magnetic field stability per time is also important. To evaluate this stability, the magnetic field drift at the MRI magnet center was monitored by a NMR probe. By a measurement with nine days, the drift was observed as 3 ppb/h, which is sufficient to fulfill our requirement.

### 3.2. Principle of the NMR probe

As \(\nu_{12}\) and \(\nu_{34}\) are dependent to the magnetic field, precise magnetometers are required in the measurement. For the magnetometer, CW-NMR probe is used. The NMR is the method to measure the spin precession frequency of the atom induced by the external magnetic field. Typically, the proton NMR frequency \(\nu_0\) inside the water molecule is measured. This is expressed as

\[
2\pi\nu_0 = \gamma_p B_0, \tag{12}
\]

where \(\gamma_p\) is the gyromagnetic ratio of the proton known as \(\gamma_p = 2\pi \times 42.577 478 92(29) \text{ MHz} \ T^{-1}\) [3]. In 1.7 T, the proton NMR frequency is about 72.382 MHz. In practice, the measured proton NMR frequency is affected by the surrounding water molecules and other materials. Therefore, the measured NMR frequency \(\nu_p\) deviates from \(\nu_0\) as

\[
\nu_p = (1 - (\sigma(\text{H}_2\text{O}) + \delta_b + \delta_p + \delta_s))\nu_0. \tag{13}
\]

\(\sigma(\text{H}_2\text{O})\) is the diamagnetic shielding effect of the water molecule which is dependent on the temperature of the water sample [21]. \(\delta_b\) is the bulk diamagnetism of the water sample, which is decided by the shape of the glass tube to fill the water sample. \(\delta_p\) is the effect of paramagnetic impurities in water and \(\delta_s\) is the effect of paramagnetic or diamagnetic materials near the sample. Therefore the systematic shift by the probe material or shape should be considered to estimate the external magnetic field.

CW-NMR is a method to measure the NMR frequency by sweeping the microwave and detecting the signal when the microwave power is absorbed by the proton inside the water sample. Figure 4 shows the schematic of the CW-NMR probe. The NMR signal is measured
when the NMR frequency by the external magnetic field and the microwave frequency are matched. To match these frequencies, we mandatory impose a tiny magnetic field sweep by the modulation coil. The NMR signal is detected by the RF pickup coil, and passes through the amplification and detection circuit. In this circuit, the output is the amplitude of the input microwave passed through the RF pickup coil and be observed as an envelope signal. Here, the NMR signal is observed as a voltage drop (See figure 5).

Figure 4. The schematic of the CW-NMR probe. The glass tube with the water sample is placed inside the probe, covered by the aluminum and teflon pipes. The proton NMR signal is detected by the RF pickup coil connected to a circuit for amplification and detection to read out. To make a tiny sweep of the magnetic field, a saddle type modulation coil winded outside the probe.

Figure 5. Detected signals of the CW-NMR probe. The horizontal axis corresponds to the time span of the magnetic field modulation swept periodically. The black points are the raw data and the blue points are the averaged data. The peak of the enveloped signal is detected by the red line peak fitting.

There are two types of NMR probes are required for the high-field MuHFS measurement, a fixed NMR probe for monitoring the magnetic field during the measurement and a multi-channel field mapping probe to measure the magnetic field homogeneity of the surface of the muonium formed volume. Fixed NMR probes will be placed around the apparatus to evaluate the magnetic field stability during the measurement. The Multi-channel field mapping NMR probe will be installed inside the apparatus to measure the spatial distribution of the magnetic field. This is formed by a half oval circuit board with multi-channel NMR probes which can rotate and scan. Figure 6 shows the prototype of the field mapping NMR probe.
Figure 6. The schematic and the prototype picture of the field mapping probe. 24 RF pickup coils will be placed around the board (one is placed at this figure) and all NMR signals will be detected through the circuit. The probe will be inserted into the RF cavity and the magnetic field of the surface of the muonium formation volume will be scanned by the iteration of rotation and scanning. The field map will be measured before and after the MuHFS measurement.

To evaluate the accuracy of our CW-NMR probe, cross calibration tests were performed with the pulse probe which is used for the FermiLab muon storage ring experiment [11] at Argonne National Laboratory. The cross calibration test are performed by measuring the magnetic field in the same position with the CW-NMR probe and the pulse-NMR probe. As designs and measurement principles of the two probes are different, the systematic uncertainties are also different. Therefore, the accuracy of both probes will be reliable when the results including the uncertainties have an agreement. The cross calibration tests are performed at March 2017 and March 2018 with a 1.45 T magnetic field (the magnetic field required in the Fermi muon storage ring experiment) and the analysis is in progress.

4. Summary
MuSEUM collaboration succeeded in the muonium hyperfine spectroscopy with an extremely low magnetic field [17] and the results are under analysis. We are now continuing our research with upgrading the apparatus and the detection system. With the construction of the MLF H-line, the development for the experiment with a high magnetic field is ongoing in parallel. The major part of the research and development required for the this experiment is the high precision NMR probe. With the R&D of the NMR probes, we are also evaluating our CW-NMR probes with the pulse NMR probes used at Fermilab muon $g-2$ experiment to ensure the accuracy.

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