Letter

Rabi-oscillation spectroscopy of the hyperfine structure of muonium atoms

S. Nishimura,[1,2,*] H. A. Torii,[3] Y. Fukao,[1,2,4] T. U. Ito,[2,5] M. Iwasaki,[6] S. Kanda,[6] K. Kawagoe,[7] D. Kawall,[8] N. Kawamura,[1,2,4] N. Kurosawa,[1,2] Y. Matsuda,[9] T. Mibe,[1,2,4] Y. Miyake,[1,2,4] N. Saito,[1,2,4,3] K. Sasaki,[1,2,4] Y. Sato,[1] S. Seo,[6,9] P. Strasser,[1,2,4] T. Suehara,[7] K. S. Tanaka,[10] T. Tanaka,[6,9] J. Tojo,[7] A. Toyoda,[1,2,4] Y. Ueno,[6] T. Yamanaka,[7] T. Yamazaki,[1,2,4] H. Yasuda,[3] T. Yoshioka,[7] and K. Shimomura[1,2,4]

(MuSEUM Collaboration)

[1]*High Energy Accelerator Research Organization (KEK), 1-1 Oho, Tsukuba, Ibaraki 305-0801, Japan*

[2]*J-PARC Center, 2-4 Shirakata, Tokai, Ibaraki 319-1195, Japan*

[3]*School of Science, The University of Tokyo, 7-3-1 Hongo, Bunkyo, Tokyo 113-0033, Japan*

[4]*School of High Energy Accelerator Science, The Graduate University for Advanced Studies (SOKENDAI), 1-1 Oho, Tsukuba, Ibaraki 305-0801, Japan*

[5]*Advanced Science Research Center, Japan Atomic Energy Agency, 2-4 Shirakata, Tokai, Ibaraki 319-1195, Japan*

[6]*RIKEN, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan*

[7]*Kyushu University, 744 Motooka, Nishi, Fukuoka 819-0395, Japan*

[8]*Department of Physics, University of Massachusetts Amherst, 1126 Lederle Graduate Research Tower, Amherst, Massachusetts 01003-9337, USA*

[9]*Graduate School of Arts and Sciences, The University of Tokyo, 3-8-1 Komaba, Meguro, Tokyo 153-8902, Japan*

[10]*Cyclotron and Radioisotope Center (CYRIC), Tohoku University, 2-1-1 Katahira, Aoba, Sendai, Miyagi 980-8577, Japan*

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As a method to determine the resonance frequency, Rabi-oscillation spectroscopy has been developed. In contrast to conventional spectroscopy which draws the resonance curve, Rabi-oscillation spectroscopy fits the time evolution of the Rabi oscillation. By selecting the optimized frequency, it is shown that the precision is twice as good as conventional spectroscopy with a frequency sweep. Furthermore, the data under different conditions can be treated in a unified manner, allowing more efficient measurements for systems consisting of a limited number of short-lived particles produced by accelerators such as muons. We have developed a fitting function that takes into account the spatial distribution of muonium and the spatial distribution of the microwave intensity to apply this method to ground-state muonium hyperfine structure measurements at zero field. It was applied to the actual measurement data, and the resonance frequencies were determined under various conditions. The result of our analysis gives $\nu_{\mathrm{HFS}} = 4\,463\,301.61 \pm 0.71$ kHz.

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I. INTRODUCTION

Atomic spectroscopy is a reliable tool for precision studies of the properties of elementary particles. Measurement of the transition energy of an atom undergoing electric and magnetic transitions can provide information on the mass and magnetic moment of the particle constituting the atom. Conventional spectroscopy plots signal intensity as a function of a frequency swept over the range of the resonance frequency. The resonance frequency is obtained by finding the center of the curve shaped by fitting an appropriate resonance function to the resonance curve. In this paper, we introduce an analysis method in which the resonance frequency can be obtained by fitting the simulated function to the time evolution of the Rabi oscillation directly without any frequency sweep. This method, named Rabi-oscillation spectroscopy, can essentially eliminate systematic uncertainties due to power fluctuations. We report the application of this method to the spectroscopy of the spin resonance of muonium atoms.

Muonium is a bound state comprising a positive muon and an electron, being as such one of several hydrogenlike atoms. Spectroscopy of muonium atoms is a promising method in the search for new physics in particle-physics research. In the standard model of particle physics, the positive muon and the electron are pointlike lepton particles; therefore, the contribution of the strong interaction is relatively small and well understood compared to hydrogen. The muon-to-electron mass ratio can be found from the muonium hyperfine structure (HFS) [1]. The electron mass has been measured precisely, so the spectroscopy of the muonium HFS provides the most precise estimation of the muon mass, which is an essential input parameter for determinations of the standard model parameters, such as the Fermi coupling constant. Also, the precise measurement of the muonium HFS is important for the determination of the muon anomalous magnetic moment $g - 2$ [2–4].

In previous experiments [5–7], the muonium HFS was measured by sweeping either the frequency of the microwave field or the strength of the external magnetic field. The dominant source of the measurement uncertainty was the statistical nature of the experiment. It is necessary to ensure the most

*shoichiro.nishimura@kek.jp

efficient use of the data. Considering that the microwave power can change during the frequency sweep, it will be an effective use of the data if we no longer need to limit the data to those with stable microwave-power conditions.

The complexity of the muonium resonance experiment stems from the fact that a muon decays in a very short lifetime of about 2.2 μs. In the conventional method, the muonium HFS interval is determined by drawing a resonance curve of the time-integrated signal as a function of the microwave frequency; the width of the resonance curve limits the obtainable precision of the resonance frequency. The key to reaching higher precision is to reduce the resonance width, attributed to the natural width and power broadening.

In general, in the case of the homogeneous natural width or power broadening, the resonance frequency can be obtained precisely by fitting a Lorentzian function to the resonance curve. Even if an ideal resonance curve is constructed, it is still challenging to determine the resonance-center frequency with an uncertainty smaller than one hundredth of the resonance width. If there is inhomogeneous broadening, such as a Doppler width or a particular systematic factor that makes the resonance curve asymmetric, the precision of the resonance-frequency measurement becomes even lower. In particular, asymmetry in the microwave power across a resonance line would lead to difficulties in extracting the line center. The natural width of the muonium resonance line is 145 kHz due to the finite lifetime of the muon, so even achieving 1-kHz precision on a line center is difficult.

One technique to improve the measurement precision is to reduce the resonance width. It is possible to extend an apparent lifetime by using the time information of the signal. Namely, the width in the vicinity of the resonance center becomes narrower by selecting the later signals, i.e., signals from those muonium atoms that have interacted for a longer period of time with the microwave. However, this method leaves the envelope curve wide because of sidelobes that appear in the frequency space in the process of the Fourier transformation. Also, events occurring in later times are selected at the cost of the statistics. Therefore, it is necessary to select an optimal time range to reconcile the advantages of the narrower width of the center peak with the disadvantages of limited statistics. In an experiment where the time range is not selected by hardware but all signals are acquired with the corresponding time information, it is possible to analyze data by selecting an appropriate time range after the measurement. For muonium atoms, this method, which is called the old muonium method, was also applied to improve the measurement precision [8].

II. RABI-OSCILLATION SPECTROSCOPY

The Rabi-oscillation spectroscopy presented here is a technique in which the detuning frequency is directly obtained from the Rabi oscillation. Let us consider the time evolution, known as Rabi oscillation, of transition signals from a two-state atom interacting with a microwave field. When the microwave frequency is detuned away from the resonance center, the peak of the signal becomes lower, and the oscillations become faster. Therefore, the detuning frequency can be obtained from the time evolution of the Rabi oscillation if the microwave field strength is known.

In the case of the muonium HFS measurement, the time evolution of muon polarization can be obtained by measuring the change in the number of decay positrons at a downstream or upstream counter because the positron emission direction is preferentially along the muon spin direction. The muon spin-flip signal due to the Rabi oscillation can be extracted if we define this signal as the ratio of the number of positrons with the microwave present N_{ON} and the number of positrons without the microwave N_{OFF} minus 1.

The Hamiltonian of muonium in a static magnetic field $\vec{B}$ is expressed as

$$\mathcal{H} = h\nu_{\mathrm{HFS}}\vec{S}_\mu \cdot \vec{S}_e + \left(g'_e\mu_e^{\mathrm{B}}\vec{S}_e - g'_\mu\mu_\mu^{\mathrm{B}}\vec{S}_\mu\right)\cdot\vec{B}, \tag{1}$$

where $\vec{S}_\mu$ is the muon spin operator, $\vec{S}_e$ is the total angular momentum operator of the electron, g'_μ and g'_e are the bound-state g factors of the muon and electron in muonium, respectively [9], $\mu_e^{\mathrm{B}} = e\hbar/2m_e$, $\mu_\mu^{\mathrm{B}} = e\hbar/2m_\mu$, m_e is the electron mass, and m_μ is the muon mass.

A microwave field with an appropriate frequency excites muonium. The time-dependent term of the Hamiltonian $\mathcal{H}'$ is given by

$$\mathcal{H}' = \left(g'_e\mu_e^{\mathrm{B}}\vec{S}_e - g'_\mu\mu_\mu^{\mathrm{B}}\vec{S}_\mu\right)\cdot\vec{B}_1\cos 2\pi\nu_{\mathrm{mw}}t, \tag{2}$$

in which $\vec{B}_1$ is the vector amplitude of the microwave field and ν_{mw} is the microwave frequency. Solving Eq. (2) under the condition of zero magnetic field yields the time dependence of state amplitudes, and the time evolution of the signal is expressed as

$$S(t) = \frac{N_{\mathrm{ON}}(t)}{N_{\mathrm{OFF}}(t)} - 1 = A\left(\frac{G^+}{\Gamma}\cos G^- t + \frac{G^-}{\Gamma}\cos G^+ t - 1\right), \tag{3}$$

in which A is a constant dependent on the detector acceptance and energy threshold of decay positrons and

$$G^\pm = \frac{\Gamma \pm \Delta\omega}{2}, \tag{4}$$

$$\Gamma = \sqrt{(\Delta\omega)^2 + 8|b|^2}, \tag{5}$$

where $\Delta\omega$ is the detuning angular frequency and $|b|$ is a parameter proportional to the strength of the applied microwave field. The time dependence of the signal is expressed by the summation of cosine functions. Their oscillation frequencies and amplitudes are related to the detuning frequency and the field strength of the stored microwave. When the microwave frequency is further detuned from the HFS resonance, the frequency G^- decreases, and G^+ increases, while the signal amplitude G^+/Γ increases and G^-/Γ decreases. When the microwave power increases, both frequencies, G^- and G^+, increase, while the signal amplitude G^+/Γ decreases and G^-/Γ increases. The response to the signal form is different for the microwave frequency and the stored energy. Hence, the Rabi-oscillation analysis can extract the muonium HFS and the microwave power at the same time, both from the time evolution of the resonance signal. This also indicates that there is no need to sweep the microwave frequency. Therefore, the Rabi-oscillation analysis does not suffer from the uncertainty

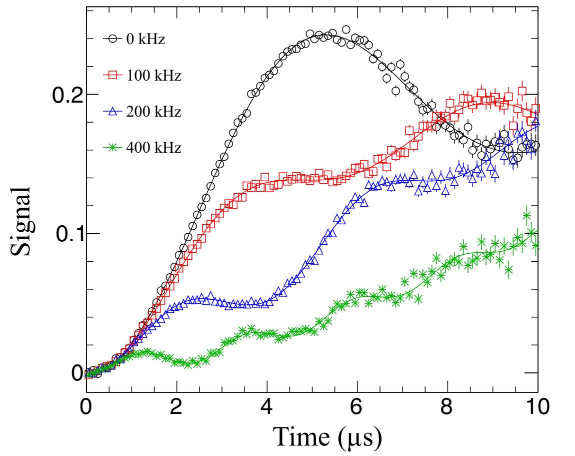

FIG. 1. Simulation results of the time evolution of resonance signals for muonium atoms at different detuning frequencies of 0, 100, 200, and 400 kHz. Error bars represent the statistical uncertainties.

due to a systematic factor that makes the resonance curve asymmetric, such as the frequency dependence of energy stored in the cavity.

III. SIMULATION

The signals observed in the measurement are, in fact, more complicated because of the spatial distribution of the muonium atoms and the variance of the magnetic field strength of the microwave. The distribution of the microwave power felt by muonium atoms was evaluated by referring to a calculated field map in the microwave cavity [10] and summing up over the muon-stopping distribution estimated with a particle-tracking simulation program based on the GEANT4 tool kit [11–13]. Then, the time evolution of the resonance signal was calculated by estimating the time distribution of the number of detected positrons in each case with and without microwaves. Statistical fluctuations were calculated according to the Poisson distribution. Simulation results of the time-evolution signals for several detuning frequencies are plotted in Fig. 1. These simulated signals were then used as a data set for the Rabi-oscillation analysis to see if the correct detuning frequencies can be extracted back by the data fitting. The fitting function is expressed as

$$f(t;\ A,\ |b|,\ \Delta\omega) = A\sum_i N_i\left(\frac{G_i^+}{\Gamma_i}\cos G_i^- t + \frac{G_i^-}{\Gamma_i}\cos G_i^+ t - 1\right), \tag{6}$$

where i represents the index of summation over the discrete muon-stopping position and N_i is the fraction of stopping muons at that position, with A, $|b|$, and $\Delta\omega$ being free parameters.

It must be noted that the absolute value of the detuning frequency $|\Delta\omega|$ can be obtained from the Rabi-oscillation analysis but not its sign; the reason is that a negative detuning gives exactly the same Rabi oscillation as a positive detuning, as is evident from Eqs. (3)–(5). The muonium HFS frequency

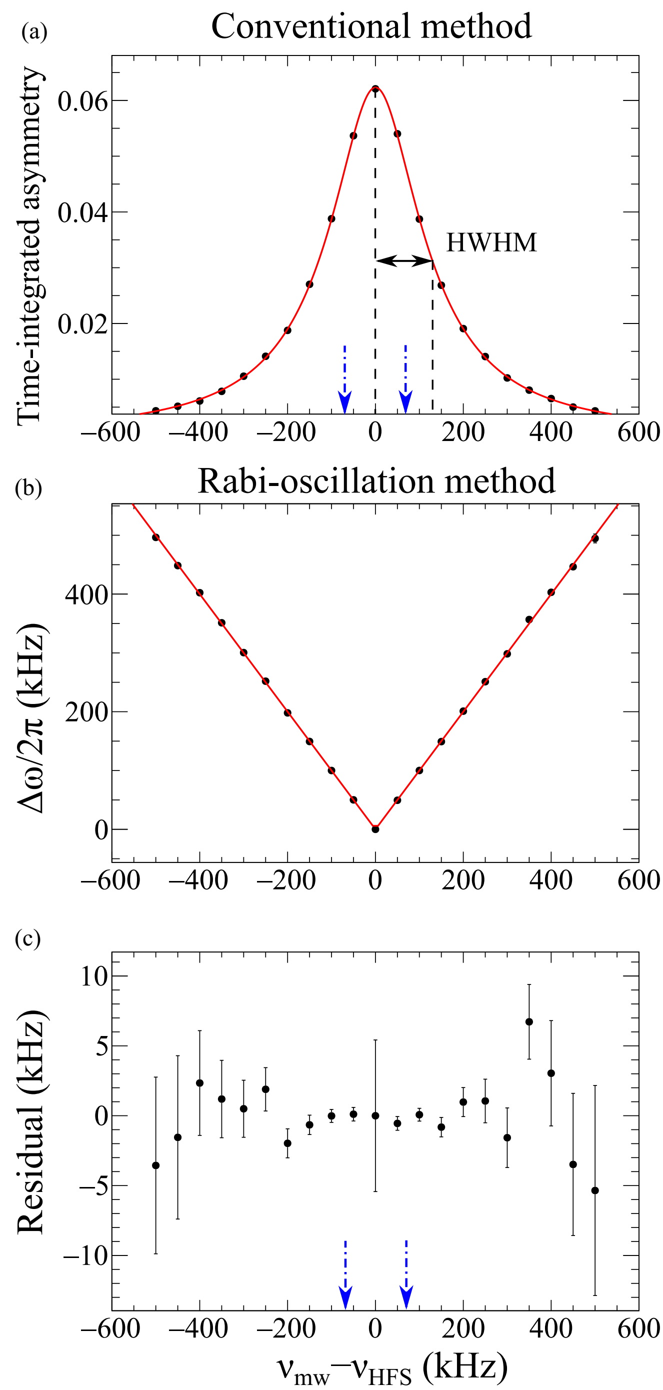

FIG. 2. Simulation results of (a) the conventional and (b) the Rabi-oscillation methods. The solid lines indicate the fitting results. (c) The residuals of the Rabi-oscillation method. The black dashed lines and the horizontal arrow in (a) indicate the HWHM of the resonance curve, and the blue dash-dotted arrows in (a) and (c) represent the detuning frequency with the lowest uncertainty in the Rabi-oscillation spectroscopy.

ν_{HFS} is obtained from the following equation:

$$|\Delta\omega|/2\pi = |\nu_{\mathrm{mw}} - \nu_{\mathrm{HFS}}|. \tag{7}$$

The results of simulated measurements at various microwave frequencies are shown in Fig. 2. The

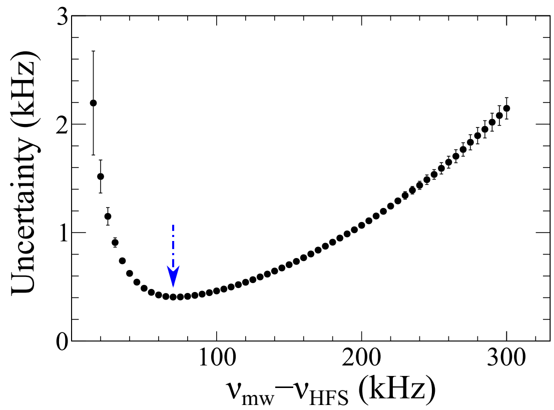

FIG. 3. The detuning-frequency dependence of fitting uncertainties of the resonance frequency obtained by repeating 500 times for each frequency the same simulation of the Rabi-oscillation spectroscopy as in Fig. 2. The error bars represent standard deviations.

spatial discretization was 1 mm, fine enough to calculate the Rabi-oscillation signals from the power distribution in the cavity. The resonance frequency can be obtained by both the conventional and Rabi-oscillation methods. The fitting results of the simulation ensure that we can successfully obtain the resonance frequency from the Rabi-oscillation analysis for different microwave frequencies.

The plot of the residuals in Fig. 2(c) shows that the uncertainty depends on the detuning frequency in the Rabi-oscillation spectroscopy. The uncertainty in determining the detuning frequency becomes large when the microwave is tuned on resonance because fitting parameters $|b|$ and $\Delta\omega$ are correlated and difficult to separate. The uncertainty is also large when the microwave frequency is far away from the resonance because most muons decay before the spin flip due to the slow Rabi oscillation. The same simulation as in Fig. 2 was repeated 500 times for each frequency to investigate the detuning-frequency dependence of the uncertainty in determining the resonance frequency by the Rabi-oscillation method, and the results are shown in Fig. 3. The best precision is achieved for a detuning frequency of about ± 70 kHz, corresponding to half of the half-width at half maximum (HWHM) of the resonance curve in the frequency domain. According to our simulation study, the Rabi-oscillation method can improve precision nearly twice as much as the conventional method by concentrating all data at a detuning frequency of about ± 70 kHz.

IV. EXPERIMENT AND RESULTS

There were experiments in the past that tried to optionally use time-dependent information [14–16] in order to confirm the results of the conventional method. The Rabi-oscillation spectroscopy, which makes full use of time information, requires high statistics and a good time-accurate detector, which was not technically possible in the past.

New precise measurements of the muonium ground-state HFS are now in progress using the high-intensity pulsed muon beam from the Muon Science Establishment (MUSE) in the Materials and Life Science Experimental Facility (MLF) at the Japan Proton Accelerator Research Complex (J-PARC) by the Muonium Spectroscopy Experiment Using Microwave (MuSEUM) Collaboration [17,18]. In our recent experiment, a surface muon beam, which has a polarization of 100% and a momentum of 27.4 MeV/c, was injected into a krypton gas target magnetically shielded to approximately 100 nT, resulting in the formation of muonium. The muon spin-flip signal induced by the microwave was measured with downstream positron detectors that use a silicon strip sensor which

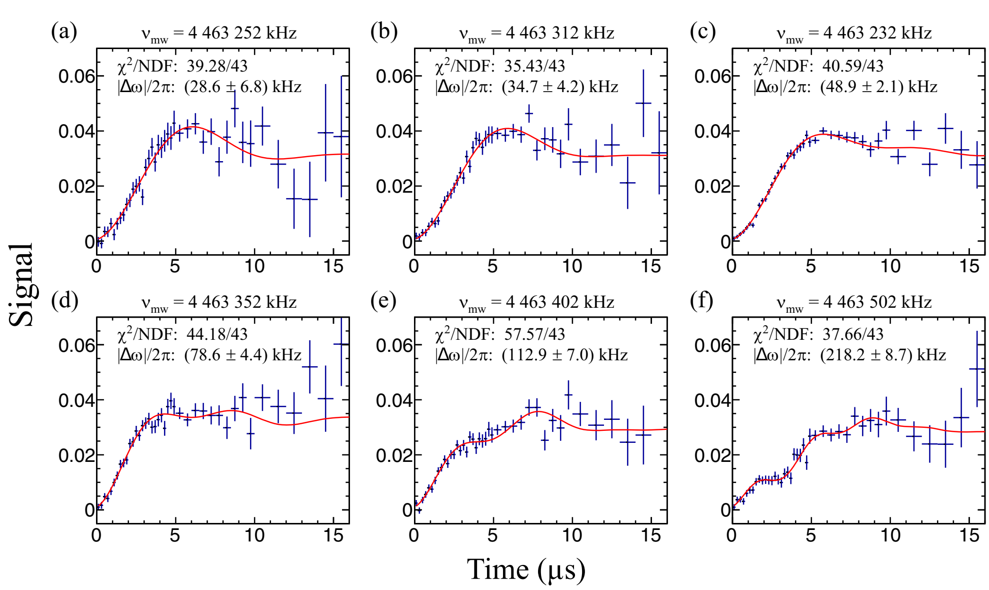

FIG. 4. Experimental results with the Rabi-oscillation spectroscopy. (a)–(f) show the time evolution of resonance signals with microwave frequencies from 4 463 232 to 4 463 502 kHz and are shown in ascending order of the absolute value of the detuning frequency $|\Delta\omega|/2\pi$. The solid lines indicate the fitting results.

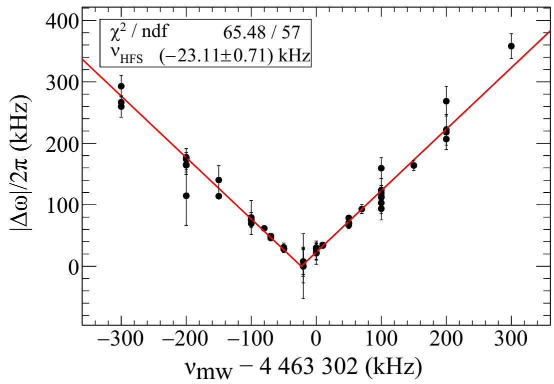

FIG. 5. Determination of ν_{HFS} from the fitting results of the Rabi-oscillation analysis for different microwave frequencies. The data were for a krypton gas target with a pressure of about 7×10^4 Pa.

digitizes positron hit time at 5 ns and has a time resolution of about 3 ns [19]. The sensor was optimized for the pulsed-beam experiment and exhibited a high-rate capability to reduce inefficiency due to pileup events. The use of a high-intensity beam and a detector with a good time response, as described above, were important keys in the technical development needed to make the Rabi-oscillation spectroscopy successful.

We have analyzed part of the experimental data obtained in June 2018 using the Rabi-oscillation method. At J-PARC, the muon beam has a double-pulse structure with a 100-ns width separated by 600 ns and a repetition rate of 25 Hz. The beam flux was 7.5×10^6 muons/s at 500-kW proton-beam-power operation [20]. The target gas was krypton with a pressure of 7.003×10^4 Pa at room temperature. The measurement time at that condition was about 42 h. The results of the Rabi-oscillation analysis are shown in Fig. 4. The time of the muon injection was calibrated by a time histogram of detected positrons without the microwave. We obtained data sets with different frequencies and applied the Rabi-oscillation analysis to extract the muonium HFS frequency. The results plotted in Fig. 5 show the microwave frequency in the horizontal axis and the detuning frequency obtained by the Rabi-oscillation analysis in the vertical axis. The data contain measurements at different microwave powers, yet the data points are well fitted together in a straight line, revealing the advantageous features of the Rabi-oscillation spectroscopy, which can make use of all the data available regardless of difference in the microwave input power.

By correcting for the gas density shift of the resonance frequency of 22.72 kHz due to atomic collision [5], the extrapolated resonance frequency at zero density ($\rho = 0$) was determined as

$$\nu_{HFS}(0) = (4\,463\,301.61 \pm 0.71)\text{ kHz (160 ppb)}, \quad (8)$$

where the uncertainty shown is of a statistical nature.

Dominant systematic uncertainties are listed in Table I. The systematic uncertainties consist of the uncertainty due to the precision of the gas pressure determination, the uncertainty due to the frequency or power of the microwaves, and the uncertainty due to the performance of the detector. The precision of the gas pressure determination was caused by the precision

TABLE I. Summary of systematic uncertainties.

Contribution	June 2018	Prospects
Precision of pressure gauge (Hz)	46	5
Pressure fluctuation (Hz)	25	5
Precision of microwave frequency (Hz)	45	< 1
Microwave power drift (Hz)	10	< 1
Detector pileup (Hz)	1	< 1
Time accuracy of detector (Hz)	1	1

of the pressure gauge and the pressure fluctuation due to the gas temperature change and can be improved up to 5 Hz by using a pressure gauge with a precision of 0.02% and a temperature control device, respectively. One of the microwave-induced uncertainties derived from the lack of accuracy of the microwave-frequency reference of the signal generator (R&S SMB 100A), of which the frequency precision was approximately 10 ppb, but can be reduced to less than 1 Hz by using a GPS clock system as a reference. The other was the effect of the cavity warming which changed the Q value, typically by 1%, during the same frequency measurement. It can be reduced to less than 1 Hz by using a water-cooling system. Determination of the Rabi-oscillation frequency is dependent on the clock calibration of the time-to-digital converter used for the positron detector. The resultant uncertainty is estimated to be less than 1 Hz and negligibly small in our current overall precision, as the accuracy of the clock is better than 1 ppm and the detuning frequency is 1 MHz at most. A detailed description will be given in a separate paper.

Table II shows a comparison between our result and previous experiments. The obtained result is consistent with the two previous experiments at the Los Alamos Meson Physics Facility [6,7] within two standard deviations and 1.8 times as precise as the previous experiment at zero magnetic field [6]. By using Rabi-oscillation spectroscopy, we estimate that the precision of 4 ppb will be reached after 1 month of measurements at the new muon beamline under construction at J-PARC.

V. SUMMARY AND PROSPECTS

We have successfully established a spectroscopic method, which we named Rabi-oscillation spectroscopy, and have obtained the resonance frequency directly from the time evolution of the Rabi oscillation without drawing a resonance curve, eliminating the need for a frequency sweep. By using a fitting function that can treat the measurement data with different conditions in a unified manner, we have also established a method for determining the resonance frequency stably from

TABLE II. Comparison with previous experiments.

	Field	Muonium HFS (kHz)
Our result	Zero	4 463 301.61(71)
Thompson *et al.* [5]	Zero	4 463 308(11)
Casperson *et al.* [6]	Zero	4 463 302.2(14)
Liu *et al.* [7]	High	4 463 302.765(53)

data under various experimental conditions, which cannot be handled together by conventional methods.

We also plan to make measurements of muonium HFS at high magnetic fields. Rabi-oscillation spectroscopy can be applied there, with an expected precision of 2 ppb or better within 1 month of measurement time.

Rabi-oscillation spectroscopy finds its application in the measurement of resonance frequencies of atoms and molecules that, in general, contain unstable particles, especially those produced in limited quantities at accelerators, such as exotic atoms and isotopes with short-lived nuclei. The beam time is finite, and this technique is clearly effective as it does not require adjustment of microwave power or frequency sweeping.

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