# Refined determination of the muonium-deuterium 1*S*-2*S* isotope shift through improved frequency calibration of iodine lines

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We report a refined determination of muonium 1*S*-2*S* transition frequency and its isotope shift with deuterium by recalibrating the iodine reference lines using an optical frequency comb. The reference lines for the muonium and deuterium 1*S*-2*S* transitions are determined with a precision of $2.4 \times 10^{-10}$ and $1.7 \times 10^{-10}$, respectively. An updated muonium-deuterium 1*S*-2*S* isotope-shift frequency is derived from these references to be 11 203 464.9(9.2)(4.0) MHz, in agreement with an updated bound-state quantum-electrodynamics prediction based on 2010 adjustments of Committee on Data for Science and Technology and 2.3 times better in the systematic uncertainty than V. Meyer *et al*. [Phys. Rev. Lett. **84**, 1136 (2000)].

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## I. INTRODUCTION

The deviation of the proton charge radius between the muonic hydrogen ($\mu$H) Lamb shift measurement [1] and the recommended values in Committee on Data for Science and Technology (CODATA) [2,3] has recently been reinforced by the $2S_{1/2}$-$2P_{3/2}$ measurement in the same $\mu$H system [4]. One possible theoretical explanation for the size puzzle is unknown quantum-electrodynamics (QED) corrections on the order of 310 $\mu$eV causing $\mu$H results to be wrongly attributed to the nuclear size effect [5]. Other theories for the new interactions have also been suggested [6,7].

Experimentally, there is another long-standing $3.3\sigma$ experiment-theory discrepancy in the muon anomalous magnetic moment $(g-2)_\mu$ [8–11]. In addition, the isotope-shift measurement of 1*S*-2*S* transition between muonium and deuterium has a 1.4-ppm deviation from the current theory. In comparison, the deviation for the case of hydrogen and deuterium is only 0.6 ppb [12,13], which is better by at least three orders of magnitude. It is tentative, therefore, to speculate that new interactions may be of muon-related origin.

Microwave and laser spectroscopy of the muonium atom, a purely two-body leptonic bound state (Mu, $\mu^+e^-$), can offer stringent experimental tests for the bound-state QED without the finite-size effect due to the structureless muon nucleus [14–16]. This removes the main limiting factor caused by the hadronic structure in $\mu$H or H when comparing the theory and the experiment.

Among the lower-lying levels of non-Rydberg-state Mu atoms, the electromagnetic $1S_{1/2}$-$2S_{1/2}$ transition is of particular importance because the fundamental property of a muon (e.g., mass) can be inferred from it [16]. Currently, the most accurate value of the muon mass is $m_\mu/m_e = 206.768\,284\,3(52)$, suggested by CODATA [3], which derived its small uncertainty from the Mu ground-state (1*S*) hyperfine splitting [15]. The natural linewidths of the Mu 1*S* hyperfine and the 1*S*-2*S* transitions are both 145 kHz, limited by the ≈2.2-$\mu$s lifetime of the muon. Therefore, the optical (higher-frequency) 1*S*-2*S* transition should, in principle, offer orders of magnitude higher accuracy than the microwave (lower-frequency) ground-state hyperfine transition.

In the last 1*S*-2*S* isotope-shift measurement done at the ISIS muon facility of the Rutherford Appleton Laboratory in the United Kingdom, however, this optical advantage was not obvious because the measurement was statistically limited by the low vacuum yield of the muonium source ($\Delta f = 9.2$ MHz) [16] and was systematically limited by the low accuracy of the deuterium reference line ($\Delta f = 9.3$ MHz) [17].

In this paper, we carry out a frequency comb calibration of the iodine reference cell used in the ISIS experiment with a Doppler-free saturation spectrometer. Combining our calibration with the experimental parameters of the ISIS measurement, we can reduce the systematic uncertainty by 2.3 times. The calibration leads to an updated Mu-D 1*S*-2*S* isotope shift of 11 203 464.9(9.2)(4.0) MHz, where the first set of parentheses indicates the statistical uncertainty and the second indicates the systematic uncertainty. This is in better agreement with the current QED theory.

## II. EXPERIMENT

Our experimental setup is shown in Fig. 1. The 730-nm light source (≈500 mW) was from a titanium:sapphire laser (Technoscan TIS-SF-07e) pumped by a 10-W diode-pumped solid-state laser (Coherent Verdi V10). The light beam was subsequently diverted into (a) the Fabry-Pérot cavity [free spectral range (FSR) ≈1 GHz] for frequency stabilization and scanning, (b) the wavemeter (resolution of ≈1 GHz) for monitoring, (c) the optical frequency comb (OFC) for the absolute frequency calibration, and (d) the cell area for resolving the Doppler-free spectral features, which was partially based on our previous frequency-modulation saturation spectrometer [17].

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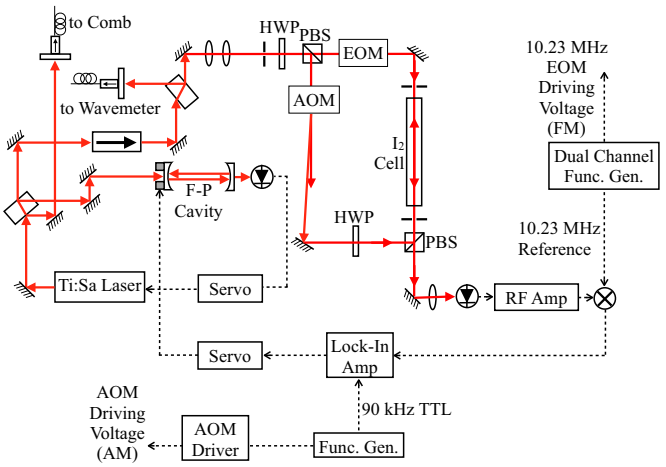


FIG. 1. (Color online) Schematic of this experiment. HWP: half-wave plate; PBS: polarizing beam splitter; F-P cavity: Fabry-Pérot cavity; AOM: acoustic-optical modulator; EOM: electro-optical modulator.

The beam delivered to the cell area was collimated (≈1 mm in radius) and further branched into the pump (≈185 mW, or, equivalently, $\approx 5.89 \times 10^4$ W/m$^2$) and the probe (≈2 mW), whose powers were adjusted independently. The pump beam was amplitude modulated (AM) at 90 kHz with an acoustic-optical modulator (IntraAction ATM-801A2). The probe beam was phase modulated (PM) with a resonant-type electro-optical modulator at 10.23 MHz (LC resonant circuit plus a Thorlabs EO-PM-NR-C2) before passing through the cell and being detected by a fast photoreceiver (125 MHz New Focus 1801). The phase-modulation depth was set such that the carrier amplitude is ≈6 times bigger than the amplitudes of the 10.23-MHz sidebands. These amplitude and phase modulation parameters are identical to the conditions used in the ISIS measurement [16,17].

The mixer (Mini-Circuits ZAD-1-1) demodulated the preamplified (HP 8447D) signal, which then was subsequently processed by the lock-in amplifier (Stanford Research SR830) to demodulate the 90-kHz signal with an adjustable phase, sensitivity, and time constant. The laser frequency was servoed (10 MHz Precision Photonics LB1005) to the center of the demodulated hyperfine signal and was calibrated by fiber coupling some laser light to the OFC. The usage of our OFC has been described previously [18]. The accuracy of our OFC system was verified by measuring the $a_{10}$ line of the $^{127}I_2$ molecular iodine R(56)32-0 transition (≈532 nm) with an error limit less than 200 kHz [19].

## III. IMPROVED IODINE REFERENCES AT 732 NM

The 54.1-cm-long iodine absorption cell used for the ISIS measurement was manufactured by the University of Heidelberg (code named HEID4 in Ref. [20]) in the 1990s, which we retrieved recently for this comb calibration project. The cell was made of quartz that can withstand temperatures up to 700 °C. The heating of iodine vapor to above 500 °C was necessary in order to access the rovibrational transitions of the *B-X* system originating from high-lying vibrational levels of the electronic ground state [17,21,22]. Our oven-temperature-dependence study showed that while the population of the high-lying levels of the ground state depended on the oven temperature (evidenced by a change in the signal-to-noise strength at different oven temperatures), the center frequencies of these hyperfine transitions remained unchanged with a standard error of 50 kHz (or ≈0.1 ppm) in the 425 °C to 650 °C range.

Two weak molecular $^{127}I_2$ transitions, not reported in the iodine atlas at 13 350–13 920 cm$^{-1}$ [23], were of interest, corresponding to the Mu and D 1*S*-2*S* transitions. One important criterion for these reference lines was that they should be located within 1 GHz of the target transition to allow the use of acoustic-optical modulators (AOM) to bridge the frequency offset. For the Mu 1*S*-2*S* reference, the $a_{15}$ line of the R(26) 5-13 transition was calibrated. For the D 1*S*-2*S* reference, the $a_{19-21}$ line of the R(137) 5-12 transition was calibrated. Figure 2 shows both reference lines.

An estimate of the frequency stability $\Delta f$ can be obtained by dividing the linewidth by the signal-to-noise ratio (SNR), leading to $\Delta f_{Mu} \approx 270$ kHz and $\Delta f_D \approx 230$ kHz. The

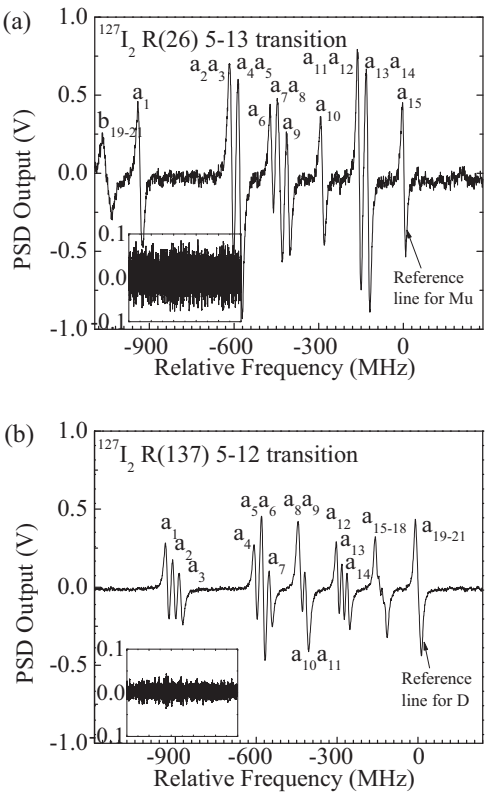


FIG. 2. Hyperfine spectra for (a) Mu and (b) D (along with frequency markers) obtained at an oven temperature of 650 °C following Ref. [16]. The insets are the stabilized phase-sensitive detection (PSD) outputs for a 10-s duration. The cold-finger temperatures of the cell are set at 43 °C and 33 °C for Mu and D, respectively. The experimental linewidth is 11.2 MHz for Mu and 23.0 MHz for D. The raw signal-to-noise ratio (SNR) is 41.2 for Mu and 98.3 for D. A time constant of 10 ms and 6 dB/octave slope are used in both spectra, which lead to an equivalent noise bandwidth (ENBW) of 25 Hz. Therefore, the calibrated 1-Hz noise bandwidth SNR are 206 $\sqrt{\text{Hz}}$ and 491 $\sqrt{\text{Hz}}$ for Mu and D, respectively.

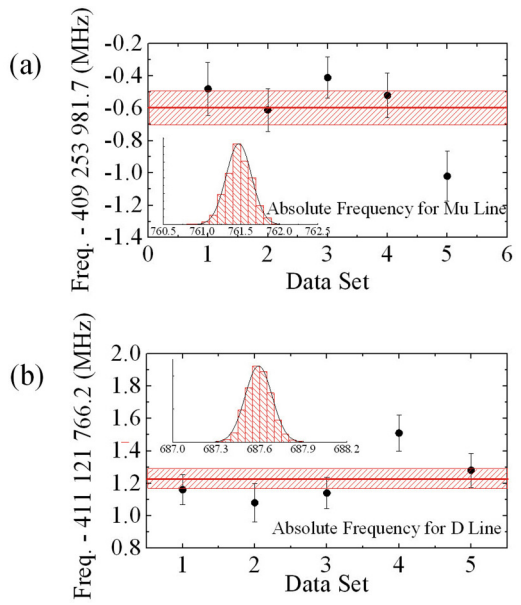


FIG. 3. (Color online) Absolute frequency comb measurements for (a) Mu and (b) D. The red (gray) lines represent constants from the zero-slope linear fitting of each data set. The shaded areas indicate the standard error of the fits. The fitted values are −0.6(0.1) MHz for Mu and 1.23(07) MHz for D. The insets are typical histograms of the beat frequencies in megahertz.

nonzero means of the Gaussian fits to the frequency-stabilized phase-sensitive detection (PSD) outputs (insets of Fig. 2) result in an offset frequency of −70 kHz for both Mu and D. In addition, due to the fact that the zero of the servo locking electronics does not necessarily coincide with the mean of the dispersive signal due to its asymmetric line shape, the voltage difference between them contributes an additional offset frequency for both Mu and D. Using the discriminant (in units of MHz/V), which is derived from the concurrent Fabry-Pérot frequency marker as the hyperfine signal is scanned, it is estimated that the lineshape asymmetry of the hyperfine signal contributes an additional offset of −450 and −80 kHz for Mu and D, respectively. The net systematic offset is thus −520 and −150 kHz for Mu and D, respectively, indicating the distance of measured frequencies away from the true hyperfine centers.

Figure 3 shows the absolute frequency calibrations of the two isotopic $1S$-$2S$ reference lines. Each data point shown in Fig. 3 is a normal distribution-fitted mean of 1000 beat frequencies (gated 0.1 s, $\geqslant$30 dB in strength) with the error bar indicating $1\sigma$ (one standard deviation) of the distribution. The standard deviation is higher for the Mu reference line, in agreement with the relative SNR sizes of the observed hyperfine spectra (Fig. 2).

The absolute frequency is calculated using the following equation:

$$f_{\text{measure}} = N \times f_{\text{rep}} \pm f_{\text{off}} \pm f_{\text{beat}} - f_{\text{AOM}}/2,$$

where $f_{\text{rep}}$, $f_{\text{off}}$, and $f_{\text{beat}}$ are the repetition rate, the offset frequency, and the beat frequency of the OFC. $f_{\text{AOM}}$ is the frequency shift of the AOM incurred during the amplitude modulation of the pump beam. The recalibrated reference values are (taking systematic offsets into account)

$$a_{15},\ \text{R(26) 5-13 (Mu)} = 409\,253\,981.6(0.1)\ \text{MHz},$$
$$a_{19-21},\ \text{R(137) 5-12 (D)} = 411\,121\,767.58(07)\ \text{MHz}.$$

## IV. IMPLICATION FOR THE ISIS EXPERIMENT

An explanation of how iodine reference lines were used in the ISIS measurement [16] is briefly given here. On average, there were 3500 $\mu^+$ particles per muon pulse in the ISIS setup. After hitting a $SiO_2$ target, a small fraction of them formed Mu (≈80 Mu per pulse). An alexandrite ring amplifier, seeded by an iodine-stabilized cw Ti:sapphire laser at 732 nm, produced laser pulses which were synchronized in time with the muon pulses. These laser pulses were first frequency doubled with lithium triborate (LBO) crystals and subsequently mixed with beta barium borate (BBO) crystals to give frequency-tripled output at 244 nm for the $1S$-$2S$ two-photon absorption. The same pulsed light source was also used for the deuterium $1S$-$2S$ measurement.

Detailed information regarding the light field with which the muonium and deuterium resonances were taken can be found in Ref. [24]. The pulsed light source was based on an alexandrite laser where the chirp compensation was disciplined and investigated using deuterium. Therefore, common systematics among the muonium and deuterium resonances cannot be excluded even though the authors of Ref. [16] did their best to avoid such. Specifically, the analysis of the spectra was done

TABLE I. Summary of QED theoretical predictions for the H, Mu, and D $1S$-$2S$ frequencies and their comparison to experimental values. The CODATA-10 adjustments of basic constants are used. Only the dominating uncertainty contributions from the electron and nucleon masses and sizes are included in the calculation. Wherever applicable, the first uncertainty set of parentheses indicates the statistical uncertainty, and the second indicates the systematic uncertainty.

| | $1S$-$2S$ Contributions | Hydrogen (MHz) | Muonium (MHz) | Deuterium (MHz) |
|---|---|---|---|---|
| Theory (CODATA-10) | Dirac eigenvalue | 2466068541.00571(75) | 2455535991.79(40) | 2466739545.08835(37) |
| | Lamb shift | −7126.786097(13) | −7056.046727(17) | −7131.300102(81) |
| | Finite-size effect | −1.054(14) | 0 | −6.288(14) |
| | Total | 2466061413.165(15) | 2455528935.74(40) | 2466732407.500(15) |
| Experiment | This work | | 2455528940.6(9.1)(3.7) | 2466732405.5(1.1)(1.5) |
| | Ref. [16] | | 2455528941.0(9.1)(3.7) | 2466732397.2(1.1)(8.5) |
| | Ref. [25] | 2466061413.187035(10) | | |
| | Refs. [13,25] | | | 2466732407.521641(18) |

differently (see Ref. [16]) than what is traditionally done in typical spectroscopic experiments. It was concluded that such a frequency chirp could be limited to an ≈6.5-MHz swing by high-voltage rampings of the electro-optical modulators in the alexandrite ring cavity [16].

From the recalibrated iodine reference lines, the $1S$-$2S$ energy intervals are updated in Table I along with theoretical values. Since the $1S$-$2S$ spectroscopy in Mu and D involves a frequency tripling from the baseband followed by two-photon absorption, the uncertainty in the $1S$-$2S$ reference lines would be 6 times that of the iodine reference lines.

Major systematic contributions in the ISIS measurement for Mu and D are detailed in the Table I of Ref. [16]. Among the various systematic contributions of the Mu $1S$-$2S$ frequency measurement, the residual linear Doppler shift and line fitting contributed 3.4 and 1.2 MHz, respectively. Although our OFC calibration improves the Mu $1S$-$2S$ reference to 0.6 MHz ($6 \times 0.1$ MHz), it is too small of a correction to influence the final systematic uncertainty significantly due to the dominating residual linear Doppler shift. This is the reason that our updated Mu $1S$-$2S$ frequency, denoted as $f_{\mathrm{Mu}}$, remains essentially the same as in the ISIS measurement.

On the other hand, our OFC calibration has improved the uncertainty of the iodine reference for D $1S$-$2S$ from 8.4 to 0.42 MHz, a 20 times improvement. This systematic reduction improves the final uncertainty significantly. Taking uncertainties in the lock stability (0.6 MHz), the residual linear Doppler shift (0.4 MHz), and the line fitting (1.2 MHz) into account, the final systematic uncertainty of the D $1S$-$2S$ frequency $f_{\mathrm{D_{NTHU}}}$ is updated to 1.5 MHz. In addition, the difference with the most accurately determined D $1S$-$2S$ experimental value (extracted from Refs. [13,25]; $f_{\mathrm{D_{MPI}}}$) is reduced to 0.82 ppb in comparison to 4.2 ppb in the ISIS measurement.

The Mu-D $1S$-$2S$ isotope shifts extracted from various measurements are summarized in Table II along with the theoretical value. The 400-kHz uncertainty of the theoretical Mu-D isotope shift mainly comes from the electron-muon mass ratio measurement [15]. The good agreement between our updated Mu-D isotope-shift interval and the theory indicates that the systematic effect in the ISIS measurement has been correctly taken into account. For the case of $f_{\mathrm{D_{NTHU}}} - f_{\mathrm{Mu}}$, the isotope-shift derivation is based on a differential type of measurement using the same pulsed excitation source where the common systematics have been canceled as far as possible. The systematic uncertainty is improved by 2.3 times, and the difference with the theoretical value is changed from 1.4 to 0.62 ppm, in comparison to the ISIS measurement. The uncertainties are now only limited by the Mu $1S$-$2S$ interval. A slightly higher accuracy can be derived from $f_{\mathrm{D_{MPI}}} - f_{\mathrm{Mu}}$, assuming negligible common systematics in Ref. [16]. In this case, the systematic uncertainty in the Mu-D isotope shift is improved by 2.5 times.

TABLE II. List of Mu-D $1S$-$2S$ isotope-shift intervals. The uncertainty of $f_{\mathrm{D_{MPI}}}$ has been treated as a statistical uncertainty during the $f_{\mathrm{D_{MPI}}} - f_{\mathrm{Mu}}$ calculation.

| | $1S$-$2S$ Mu-D isotope shift (MHz) |
|---|---|
| Theory | 11203471.8(0.4) |
| Ref. [16] (expt.) | 11203456.2(9.2)(9.3) |
| $f_{\mathrm{D_{NTHU}}} - f_{\mathrm{Mu}}$ | 11203464.9(9.2)(4.0) |
| $f_{\mathrm{D_{MPI}}} - f_{\mathrm{Mu}}$ | 11203466.9(9.1)(3.7) |

## V. CONCLUSION

In summary, we have calibrated the $^{127}I_2$ reference cell, used in the last Mu-D $1S$-$2S$ isotope-shift measurement, with a frequency comb for possible unknown systematics which might shift the Mu $1S$-$2S$ interval. We determine a refined value for the Mu-D $1S$-$2S$ isotope-shift frequency which agrees with the updated value calculated with CODATA fundamental constants. At the present level of experimental accuracy, no inconsistency with the current bound-state QED theory can be found.

Further improvement in the experimental accuracy will require increasing the statistics. Recent technological advancement in increasing the Mu vacuum yield to 38% at 250 K with mesoporous silicon [26], which may permit cw laser spectroscopy in an enhancement cavity, shows great promise in alleviating this statistical limitation.

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