## Precision Measurements of the ${}^{138}Ba^+$ $6s^2S_{1/2} - 5d^2D_{5/2}$ Clock Transition

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(Received 21 June 2019; revised manuscript received 11 March 2020; accepted 27 April 2020; published 11 May 2020)

Measurement of the <sup>138</sup>Ba<sup>+</sup>  ${}^{2}S_{1/2} - {}^{2}D_{5/2}$  clock transition frequency and  $D_{5/2}$  Landé  $g_J$  factor are reported. The clock transition frequency  $\nu_{Ba^+} = 170126432449333.31 \pm (0.39)_{stat} \pm (0.29)_{sys}$  Hz, is obtained with accuracy limited by the frequency calibration of the maser used as a reference oscillator. The Landé  $g_J$  factor for the  ${}^{2}D_{5/2}$  level is determined to be  $g_D = 1.20036739(24)$ , which is a 30-fold improvement on previous measurements. The *g*-factor measurements are corrected for an ac-magnetic field from trap-drive-induced currents in the electrodes, and data taken over a range of magnetic fields underscores the importance of accounting for this systematic.

DOI: 10.1103/PhysRevLett.124.193001

Singly ionized Barium has been considered for a variety of applications in atomic physics including parity nonconservation investigations [1], optical clocks [2], and quantum information [3–5]. Being a monovalent atom, it is also amenable to precise theoretical predictions of atomic properties [6,7], making it a testbed for theoretical and experimental techniques. This has led to high accuracy measurement of a variety of atomic properties including Landé *g* factors [8–11], matrix elements [12,13], branching ratios from the  $P_{1/2}$  and  $P_{3/2}$  levels [14–16], lifetime measurements of the lower lying metastable *D* states [17,18], fine-structure splittings [19], and hyperfine structure of odd isotopes [20,21].

The <sup>138</sup>Ba<sup>+</sup>  $S_{1/2} - D_{5/2}$  clock transition shares many of the advantages of the corresponding transitions in <sup>88</sup>Sr<sup>+</sup> and <sup>40</sup>Ca<sup>+</sup>. In addition, the upper-state lifetime of ~30 s would allow long interrogation times. Moreover, dynamic-decoupling, as recently demonstrated in Sr<sup>+</sup> [22], is directly applicable to Ba<sup>+</sup> and opens the door to multi-ion clock implementations [23]. Our objective in this study is threefold: (i) determination of the clock transition frequency for which an accurate value has not yet been reported, (ii) an improved accuracy measurement of the  $D_{5/2}$   $g_J$  factor, and (iii) validation of Ba<sup>+</sup> as a trap diagnostic tool for characterizing rf-induced ac-magnetic fields. The ac-magnetic field assessment is relevant for subsequent Lu<sup>+</sup> clock experiments in the same apparatus.

In this Letter, the clock frequency is measured with sub-Hertz level accuracy, which is accomplished by averaging over six Zeeman transitions to eliminate dominant linear Zeeman shifts and tensor shifts. From a combination of four transitions, the ratio of Zeeman splittings between the  $S_{1/2}$  and  $D_{5/2}$  levels is extracted. The  $S_{1/2}$  Landé g factor,  $g_S$ , is known to the  $10^{-8}$  level [8], so this ratio provides an accurate determination of the  $D_{5/2} g$  factor,  $g_D$ .

As ac-magnetic fields arising from rf currents in the electrodes can compromise the accuracy of this determination, an Autler-Townes splitting on the clock transition is used to accurately characterize this systematic [24].

The experiments are carried out using a four-rod linear Paul trap with axial end caps described in previous work [25,26]. The rf potential at a frequency near  $\Omega_{\rm rf} = 2\pi \times 20.7$  MHz is delivered via a quarter-wave helical resonator. Together with static potentials of 5 V on the end caps and -0.5 V on two of the diagonally opposite rods, the measured trap frequencies for a single <sup>138</sup>Ba<sup>+</sup> ion are ~(890, 770, 240) kHz, with the lowest frequency along the trap axis, defined as  $\hat{z}$ . The quantization axis is defined by a magnetic field applied in the xz plane at angle  $\phi$  with respect to  $\hat{x}$  [Fig. 1(e)]. For the absolute frequency and  $g_D$ measurements,  $\phi$  is set to 33(2)° and 0°, respectively.

The level structure of  ${}^{138}\text{Ba}^+$  is shown in Fig. 1(a) along with the relevant transitions used in this work. Doppler cooling is provided by driving the 493- and 650-nm transitions, where  $\approx 2.7\%$  of the fluorescence at 650 nm is detected on a single photon counting module for state determination [27]. The  $D_{5/2}$  level is populated by driving the  $S_{1/2} - D_{5/2}$  clock transition at 1762 nm and depopulated by driving the  $D_{5/2} - P_{3/2}$  transition at 614 nm. Cooling and repumping light at 493, 614, and 650 nm are all collinear and propagate at 45 degrees to the  $\hat{z}$  axis. For  $\phi = 0$ , the 493-nm cooling beam is  $\pi$  polarized, whereas the 614-, and 650-nm beams are polarized perpendicular to the magnetic field (> 0.2 mT) to avoid dark states in the D levels. State preparation into the  $m = \pm 1/2$  states of the  $S_{1/2}$  levels is facilitated by two additional 493-nm beams that are polarized  $\sigma^+$  and  $\sigma^-$ .

Spectroscopy of the clock transition is implemented using an external-cavity-diode laser, which is phase locked



FIG. 1. (a) Low-lying level structure of  ${}^{138}\text{Ba}^+$  showing transitions relevant to this work. (b) Optical clock transitions used. The absolute frequency of the  $S_{1/2} - D_{5/2}$  transition,  $\omega_0$ , is obtained from the average of transitions labeled 2–4 and their Zeeman-symmetric counterparts. The ratio of the Zeeman splittings,  $\omega_D/\omega_S$ , is found from transitions 1–2 and their Zeeman-symmetric counterparts. (c)–(e) End, side, and top views of the ion trap electrodes. Polarizations and geometric orientations of lasers indicated. The magnetic field  $\vec{B}$  is oriented at angle  $\phi = 33(2)^\circ$  for the absolute frequency measurement, and  $\phi = 0$  for the  $g_D$  measurement.

to an optical frequency comb (OFC). The short term (<10 s) stability of the OFC is derived from a  $\sim1$  Hz linewidth laser at 848 nm which is referenced to a 10 cm long ultralow expansion cavity with finesse of  $\sim 4 \times 10^5$ . For longer times ( $\gtrsim 10$  s), the OFC is steered to an active hydrogen maser (HM) reference. The frequency of the maser is calibrated to the SI (International System of Units) second by continuous comparison to a GPS timebase [28] in combination with the circular-T reports from the Bureau International of Weights and Measures (BIPM) [29,30].

For the absolute frequency measurements, the 1762 nm laser is continuously on and intensity stabilized by an acousto-optic modulator. The transition of interest is driven by the lower sideband ( $\approx$ 357.58 MHz) of a wideband electro-optic modulator (EOM) which is frequency shifted near to resonance for clock interrogation. The clock laser propagates at an angle of  $\theta = 3.0(1.5)^\circ$  with respect to  $\hat{\mathbf{x}}$  [Fig. 1(e)] and is linearly polarized in the plane spanned by the propagation vector and the magnetic field. Control of the rf power driving the EOM enables equal  $\pi$  times for each transition while maintaining the same total laser intensity at the ion.

A typical experiment consists of four steps: 500  $\mu$ s of Doppler cooling, optical pumping for 10  $\mu$ s to either  $|S_{1/2}, m = \pm 1/2\rangle$ , a 600  $\mu$ s clock interrogation pulse on a  $|D_{5/2}, m'\rangle$  transition, and finally detection for 1 ms. The



FIG. 2. (a) Allan deviation demonstrating the stability of the comparison between the barium clock and the HM (orange points). At shorter times the stability is consistent with projection noise limit of  $Ba^+$  servo (black line), and at longer times approaches the  $1.5 \times 10^{-15}$  frequency flicker noise floor of the HM (black dashed line). (b) Absolute frequency measurements given relative to  $\nu_{Ba^+} = 170126432449333.31$  Hz.

initial Doppler cooling step includes the 614-nm beam to facilitate repumping from the upper clock state. Secondorder integrating servos [31] independently track the average and difference frequency [32] for three pairs of Zeeman transitions  $m = \pm \frac{1}{2} \rightarrow \pm m'$  for  $m' = \{\frac{1}{2}, \frac{3}{2}, \frac{5}{2}\}$  [33]. With a servo update period of  $\approx 0.8$  s and probe duty cycle of 30%, each Zeeman pair is servoed for approximately 1.5 minutes before cycling to the next. Coherence times are limited by the background magnetic field noise such that the resonant transfer probabilities for  $|m'| = \{\frac{1}{2}, \frac{3}{2}, \frac{5}{2}\}$  transitions are approximately  $\sim \{0.98, 0.93, 0.80\}$  for the 600  $\mu$ s probe time.

The absolute frequency of the  $S_{1/2} - D_{5/2}$  transition is obtained from the average of all six transitions, which is free of linear Zeeman and tensor shifts [34]. A total of ten measurements were taken over one month, with total measurement time of 70 h. The fractional instability of a typical measurement is shown in Fig. 2(a) (orange points). The statistical uncertainty of individual measurements shown in Fig. 2(b) is taken from the quadrature sum (orange line) of the Ba<sup>+</sup> servo instability (solid black line) and HM instability (black dashed line). The HM frequency is evaluated using GPS comparison data from a 20-day interval centered on the respective optical measurement, and with the assumption the HM has only a linear frequency drift over that time window. The total uncertainty in the HM calibration, including the link to the SI second via the BIPM Circular T, is evaluated to be  $2.3 \times 10^{-15}$  [35]. Given the substantial overlap of the 20-day averaging windows used, the uncertainty in the maser calibration is not averaged down

TABLE I. Systematic frequency shifts and uncertainties for the  ${}^{138}\text{Ba}^+ S_{1/2} - D_{5/2}$  transition.

Description	Shift(Hz)	$\sigma$ (Hz)
Black body radiation	0.66	0.09
ac Stark (1762 nm)	0.53	0.27
Quadratic Zeeman	0.05	< 0.01
Gravitational redshift	0.32	0.06
Total	1.56	0.29

but taken as the statistical uncertainty in the final value. The mean result of all optical frequency measurements is  $\nu_{Ba^+} = 170126432449333.31 \pm (0.39)_{stat} \pm (0.29)_{sys}$  Hz, including correction for the systematic effects listed in Table I.

The leading systematic shifts and uncertainties for the  $^{138}Ba^+$  clock transition are given in Table I and each briefly discussed in the following paragraphs. Averaging over all m' states eliminates tensor shifts which include the electric quadrupole shift, and tensor components of the quadratic Zeeman shift and ac-Stark shift from the 1762-nm laser. Other systematic effects which are considered, but omitted from Table I because they are well below the final uncertainty, include the following: shifts arising from excess micromotion, ac-Stark shifts due to the leakage of 493-nm or 615-nm laser light, second-order Doppler shifts from the thermal motion of the ion, and the ac-Stark shift due to off resonant quadrupole couplings of the clock laser. For completeness, a discussion of these shifts is given in the Supplemental Material [35].

As reported in the Ref. [38], the static and dynamic differential scalar polarizabilities are evaluated to be  $\Delta \alpha_0(0) = -73.56(42)$  a.u. and  $\Delta \alpha_0(\omega_0) = -78.51(42)$  a.u., where a.u. denotes atomic units,  $\omega_0$  is the clock transition frequency, and we have taken the  $2\sigma$  uncertainties as recommended in Ref. [38]. The blackbody radiation (BBR) shift is evaluated for a temperature at ion of T = 303(10) K, 5 degrees above the ambient temperature in the laboratory. The BBR shift uncertainty is dominated by the temperature contribution.

The ac-Stark shift due to the 1762-nm clock laser is inferred from the observed Rabi coupling on the weakest transitions,  $m = \pm \frac{1}{2}$  to  $m' = \pm \frac{5}{2}$ , for which the EOM modulation depth is set to maximize the first order sideband. We evaluate the ac-Stark shift to be 0.53(27) Hz accounting for the laser polarization, which is linear in the xz plane, the laser incidence angle relative to the magnetic field,  $\phi - \theta = 30(2)^\circ$ , and the reduced electric-quadrupole matrix element,  $\langle D_{5/2} || r^2 || S_{1/2} \rangle = 15.80(11)$  a.u. [6]. Although we expect this procedure to be accurate to 22% when allowing for 10% uncertainty in determination of the  $\pi$  time, we conservatively use 50% uncertainty as it is not measured directly.

The quadratic Zeeman shift arises from Zeeman coupling between  $D_{5/2}$  and  $D_{3/2}$  fine structure states.

The magnetic dipole matrix element is calculated, in the LS-coupling regime, to be  $\langle D_{5/2} || M1 || D_{3/2} \rangle = 1.549$  (a.u.), which is consistent with other perturbative theory methods [39] to better than 1%. The quadratic Zeeman sensitivity for the average of all  $|D_{5/2}, m'\rangle$  states is evaluated to be 1.09(2) Hz mT<sup>-2</sup> and all measurements were performed at a magnetic field near 0.225 mT.

To determine the gravitational redshift we have taken a local geoid height of 7.9 m relative to the World Geodetic System (WGDS84) ellipsoid from the 2008 Earth Gravitation Model (EGM 2008) datum. The elevation of the ion in WGDS84 is determined from its relative position to the rooftop GPS receiver. From this, we estimate the local height above the geoid to be 17(3) m. Given the gravitational acceleration of 9.780 m s<sup>-2</sup> at equatorial latitude, the gravitational redshift is evaluated to be 0.32(6) Hz.

For determination of  $g_D$ , four transitions are used:  $|S_{1/2}, m = \pm 1/2\rangle \equiv |S, \pm\rangle$  to  $|D_{5/2}, m = \pm 1/2\rangle \equiv |D, \pm\rangle$ , and  $|S, \pm\rangle \leftrightarrow |D, \mp\rangle$  [see Fig. 1(b)]. The following differences for the  $g_D$  experiments are noted:  $\theta = -18^{\circ}$ ,  $\phi = 0$ , the clock probe times used were between 0.3–0.4 ms, the clock laser EOM is operated near the rf frequency  $\approx 167.55$  MHz, and the 1762 nm laser intensity was not actively stabilized with the AOM which was instead used to switch the intensity during clock interrogation.

In a static magnetic field,  $B_0$ , the Zeeman splittings for the  $S_{1/2}$  and  $D_{5/2}$  levels are given by  $\omega_S = g_S \mu_B B_0/\hbar$  and  $\omega_D = g_D \mu_B B_0/\hbar$ , respectively. The ratio  $r = \omega_D/\omega_S = g_D/g_S$  can be inferred from measured frequency differences between the four transitions  $|S, \pm\rangle \leftrightarrow |D, \pm\rangle$  and  $|S, \pm\rangle \leftrightarrow$  $|D, \mp\rangle$  from which  $g_D = rg_S$  can be obtained using  $g_S =$ 2.00249192(3) given in [8]. However, an rf-magnetic field, which arises from currents in the electrodes driven by the rf-trapping potentials, results in a measured ratio,  $\tilde{r}$ , given by [24]

$$\tilde{r} = \left[ \frac{1 + \frac{1}{2} \frac{\omega_D^2}{\omega_D^2 - \Omega_{rf}^2} \frac{\langle B_{\perp}^2 \rangle}{B_0^2}}{1 + \frac{1}{2} \frac{\omega_S^2}{\omega_S^2 - \Omega_{rf}^2} \frac{\langle B_{\perp}^2 \rangle}{B_0^2}} \right] r$$
(1)

where  $B_{\perp}$  is the amplitude of the rf-magnetic field perpendicular to the applied static field. For a given  $B_{\perp}$ , the correction factor in square parentheses can, to a good approximation, be inferred from the measured values of  $\omega_S$ , and  $\omega_D$ , using the measured  $g_S$  to determine  $B_0$ .

An accurate determination of  $B_{\perp}$  can be obtained by setting  $\omega_S = \Omega_{\rm rf}$ , and driving the optical transition  $|S, +\rangle \leftrightarrow |D, +\rangle$  with a coupling strength  $\Omega_{\rm o}$ . When  $\Omega_{\rm o} \ll \Omega_{\rm m} = g_S \mu_B B_{\perp} / (2\hbar)$ , an Autler-Townes splitting on the optical transition is observed, as illustrated in Fig. 3(a), with the two peaks separated by  $\Omega_m$ . When  $\Omega_o \gtrsim \Omega_m$ , precession of the ground-state population is



FIG. 3. (a) Autler-Townes splitting due to resonant ( $\omega_s = \Omega_{\rm rf}$ ) transverse ac-magnetic field. A fit (orange line) is obtained by  $\chi^2$  minimization of the exact Hamiltonian solution from which we find the coupling strength is  $\Omega_m/2\pi = 14.10(4)$  kHz with reduced  $\chi^2 = 0.94$ . (b) When  $\Omega_o \gtrsim \Omega_m$ , Zeeman precession is observed. A fit (orange line) to a cosine function yields a precession frequency of 14.05(1) kHz. (c) Allan deviation for the measured Autler-Townes splitting.

observed, as illustrated in Fig. 3(b), with the oscillation frequency given by  $\Omega_m$ . Both approaches give good agreement, with the data from Fig. 3 giving an inferred coupling  $\Omega_m/(2\pi)$  of 14.10(4) kHz from the Autler-Townes splitting and 14.05(1) kHz from the Zeeman precession frequency. Although these are in agreement, an Autler-Townes splitting is a better method as an off-resonant Zeeman coupling appears as an imbalance in the two peaks and can be accounted for in data analysis.

We test the stability of  $\Omega_m$  by tracking the Autler-Townes splitting with a servo. The half-width half-maximum of the both lines in the Autler-Townes feature [such as shown in Fig. 3(a)] are sequentially interrogated on both  $\Delta m = 0$ transitions,  $|S, \pm\rangle \leftrightarrow |D, \pm\rangle$ . An outer servo loop adjusts the magnetic field to maintain the condition  $\omega_S = \Omega_{\rm rf}$ . Measured continuously for over four hours, the observed splitting is projection noise limited, as illustrated in Fig. 3(c), and shows a fractional stability of  $\lesssim 2 \times 10^{-4}$ at one hour.

Measurements of  $\omega_D/\omega_S$  were taken over a range of magnetic fields spanning ~0.4–1.6 mT, and two data sets taken approximately one month apart are shown in Fig. 4. For each set, solid circles are the measured values of  $\omega_D/\omega_S$  with the magnetic field deduced from the measured ground-state splitting and the previously specified value of  $g_S$ ; diamonds are corrected using values of  $\Omega_{\rm rf}$  and  $B_{\perp}$ measured at the time the data was taken. Each point is a result of servoing on the four transitions for approximately  $10^3$  servo updates with one servo update derived from 100 measurements on either side of each of the transitions. The



FIG. 4. (a),(b) Two data sets measuring the ratio  $\omega_D/\omega_S$  over a range of magnetic fields. The red circles are the uncorrected measurements of the ratio ( $\tilde{r}$ ). Black diamonds are ratios (r) corrected for the rf-magnetic field effects. The blue line (shaded region) indicates the correction (uncertainty) evaluated by Eq. (1) given the measured  $B_{\perp} = 0.906(35) \ \mu\text{T}$  and  $1.006(3) \ \mu\text{T}$  for (a) and (b), respectively.

error bars for the uncorrected data are the statistical errors from the servo, and the corrected points include the error arising from the uncertainty in  $B_{\perp}$  as specified in the caption.

Using a  $\chi^2$  minimization to determine the mean, the data in Fig. 4(a) gives a reduced  $\chi^2$  of 1.95 indicating that the measurements are not limited by the statistical errors. At that time, it was noted that the rf-resonator was not optimally coupled, which resulted in a degraded stability of the rf-confinement and hence  $B_{\perp}$ . This was fixed for the second data set, Fig. 4(b), which gives a statistically acceptable reduced  $\chi^2$  of 1.4. Nevertheless we take as the ratio result the mean of the second data set with the full standard deviation as the uncertainty, 0.59943681(12), noting that this is also consistent with 0.59943672(11) from the first dataset. The ratio combined with  $g_S =$ 2.00249192(3) [8] yields  $g_D = 1.20036739(24)$ .

In summary, we have carried out precision spectroscopy of the  $S_{1/2} - D_{5/2}$  clock transition in <sup>138</sup>Ba<sup>+</sup>. The measurements have provided an absolute frequency determination of the clock transition with sub-Hertz level accuracy and a 30-fold improvement in the Landé  $g_I$  factor for the  $D_{5/2}$  level. To our knowledge this is the first direct measurement of the optical transition frequency. A recently reported measurement of 146 114 384.0(1) MHz for the  $S_{1/2} - D_{3/2}$  transition [40], together with the fine structure splitting of 24 012 048 319(1) kHz [19], is consistent with our result albeit limited by the 100 kHz uncertainty of [40]. Our value for  $g_D$  is in agreement with the value reported in [10]. The discrepancy with the result of [11] is potentially due to quadrupole shifts, which were not considered in that analysis. We have also demonstrated the influence that acmagnetic fields arising from rf currents in the electrodes can have on  $g_J$ -factor measurements. Similar to the microwave demonstration reported in [24], the observation of an Autler-Townes splitting on the clock transition has allowed an accurate characterization of the ac-magnetic field component orthogonal to the applied dc field. This will be an important diagnostic tool for characterizing the acmagnetic field shift for the  $Lu^+$  clock transitions [24].

This work is supported by the National Research Foundation, Prime Ministers Office, Singapore and the Ministry of Education, Singapore under the Research Centres of Excellence programme. This work is also supported by A\*STAR SERC 2015 Public Sector Research Funding (PSF) Grant (SERC Project No: 1521200080). It is also supported by the National Research Foundation, Singapore, hosted by National University of Singapore under the Quantum Engineering Programme (Grant No. QEP-P5).

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