Magic wavelength of the $^{138}\text{Ba}^+$ $6s^2S_{1/2} - 5d^2D_{5/2}$ clock transition

S. R. Chanu,$^1$ V. P. W. Koh,$^2$ K. J. Arnold,$^1$ R. Kaewuam,$^1$ T. R. Tan,$^1,2$ Zhiqiang Zhang,$^1$
M. S. Safronova$^{3,4}$ and M. D. Barrett$^{1,3,*}$

$^1$Centre for Quantum Technologies, National University of Singapore, 3 Science Drive 2, 117543 Singapore
$^2$Department of Physics, National University of Singapore, 2 Science Drive 3, 117551 Singapore
$^3$Department of Physics and Astronomy, University of Delaware, Newark, Delaware 19716, USA
$^4$Joint Quantum Institute, National Institute of Standards and Technology and the University of Maryland, College Park, Maryland 20742, USA

(Received 28 October 2019; revised manuscript received 5 March 2020; accepted 26 March 2020; published 22 April 2020)

The zero crossing of the dynamic differential scalar polarizability of the $S_{1/2} - D_{5/2}$ clock transition in $^{138}\text{Ba}^+$ has been determined to be 459.1614(28) THz. Together with previously determined matrix elements and branching ratios, this tightly constrains the dynamic differential scalar polarizability of the clock transition over a large wavelength range ($\gtrsim 700$ nm). In particular, it allows an estimate of the blackbody radiation shift of the clock transition at room temperature.

DOI: 10.1103/PhysRevA.101.042507

I. INTRODUCTION

Singly ionized barium has been well studied over the years with a wide range of precision measurements [1–8] that have provided valuable benchmark comparisons for theory [9–14]. It was recently proposed that some of these measurements, specifically high accuracy measurements of transition matrix elements and branching ratios, could be used to construct an accurate representation of the dynamic differential scalar polarizability, $\Delta \alpha_0(\omega)$, of the $S_{1/2} - D_{5/2}$ clock transition over a large wavelength range [15]. Crucial to that proposal was a determination of a zero crossing, $\Delta \alpha_0(\omega_0) = 0$, near 653 nm, which bounds significant contributions to $\Delta \alpha_0(\omega)$ from the ultraviolet (uv) spectrum. Here we determine $\omega_0$ with an inaccuracy of a few GHz.

Together with measurements of the $P_{3/2}$ branching ratios [16] and reduced matrix elements [4], our result allows us to construct a model of $\Delta \alpha_0(\omega)$ for the $S_{1/2} - D_{5/2}$ clock transition that is accurate at the sub-% level for wavelengths above 700 nm. This will allow accurate measurement of differential polarizabilities in other ions through comparison with $\text{Ba}^+$. In particular, it will enable more than an order-of-magnitude improvement in the polarizability assessments for the $^{176}\text{Lu}^+$ clock transitions [17].

II. METHOD

To find $\omega_0$, a linearly polarized laser beam near 653 nm is focused onto the ion to induce an ac Stark shift of the clock transition and the shift measured as a function of the laser frequency $\omega$. The ac Stark shift, $\delta_2(M_J)$, induced by the 653-nm laser is given by

$$\delta_2(M_J) = - \frac{1}{2} \Delta \alpha_0(\omega) \langle E^2 \rangle - \frac{1}{4} \alpha_2(\omega) \frac{\hbar^2}{\langle J^2 \rangle - J(J+1)} (3 \cos^2 \theta - 1) \langle E^2 \rangle,$$

(1)

where $M_J$ denotes the applicable eigenstate of $D_{5/2}$, $\alpha_2(\omega)$ is the dynamic tensor polarizability of $D_{5/2}$, and $\theta$ is the angle between the 653-nm laser polarization and the quantization axis. The tensor component can be eliminated by determining the average ac Stark shift:

$$\delta_0(\omega) = \frac{1}{4} \left[ \delta_1(1/2) + \delta_1(3/2) + \delta_1(5/2) \right]$$

$$= - \frac{1}{2} \Delta \alpha_0(\omega) \langle E^2 \rangle.$$

(2)

(3)

Hence, with the laser intensity fixed, $\delta_0(\omega)$ is directly proportional to $\Delta \alpha_0(\omega)$, which is approximately linear in a neighbourhood of $\omega_0$. Consequently, $\omega_0$ can be determined from a linear fit to measurements of $\delta_0(\omega)$ as a function of $\omega$ with an accuracy limited by the projection noise of the measurements, and a small nonlinearity in $\Delta \alpha_0(\omega)$, which can be estimated from theory.

Although the laser power is actively stabilized outside the experiment chamber, étalon effects and uncalibrated frequency response of the detector can give rise to a frequency dependence of the resulting laser intensity at the ion. This would alter the frequency dependence in Eq. (3) and potentially influence the accuracy of interpolation to find the zero crossing. Pointing stability of the laser can have a similar effect if it causes the intensity to drift on a timescale comparable to the measurement time at multiple frequencies. To compensate these effects, one can make use of the weighted average

$$\delta_2(\omega) = \frac{25}{47} \delta_1(5/2) - \frac{5}{2} \delta_1(3/2) - \frac{1}{2} \delta_1(1/2)$$

$$= - \frac{1}{4} \alpha_2(\omega) (3 \cos^2 \theta - 1) \langle E^2 \rangle.$$

(4)

(5)
cooling and repumping light at 493, 614, and 650 nm are used, and the repumping transition at 614 nm facilitates efficient coupling to the lower clock state. When including the 614-nm beam to facilitate repumping from the upper clock state. When including the 614-nm beam, the 653-nm laser is provided by a tunable extended-cavity-diode laser (ECDL) together with an injection-locked slave laser to boost the power. The laser is locked to a wavemeter with 10-MHz accuracy (high finesse WS88-10), which is routinely calibrated to a caesium-locked 852-nm diode laser. Control of the rf power driving a switching AOM is used to stabilize the beam power, which is sampled by a beam pick-off before the vacuum chamber.

The 1762-nm clock laser is an ECDL, which is phase locked to an optical frequency comb (OFC). The short term (< 10 s) stability of the OFC is derived from a ≳ 1-Hz linewidth laser at 848 nm which is referenced to a 10-cm-long ultralow expansion cavity with finesse of ≳ 4 × 105. For longer times (≳ 10 s), the OFC is steered to an active hydrogen maser reference. The 1762-nm laser is switched with an AOM, but frequency shifting is achieved via a wide-band electro-optic modulator (EOM) with the lower sideband driving the transition of interest. Control of the rf power driving the EOM enables equal π times for each clock transition.

A typical experiment consists of four steps: 1 ms of Doppler cooling, optical pumping for 50 μs to either |S1/2, m = ±1/2⟩ ≡ |S, ±⟩, clock interrogation (Rabi) to |D3/2, ±Mf⟩ for 1 ms, and finally detection for 1 ms. The initial Doppler cooling step includes the 614-nm beam to facilitate repumping from the upper clock state. When including the 653-nm beam to Stark shift the transition, the beam is switched on 200 μs before the clock laser, to ensure the beam power has stabilized.

The clock transition is realized by steering the clock laser to the average of the Zeeman pair |S1/2, ±1/2⟩ ↔ |D3/2, ±Mf⟩ to cancel linear Zeeman shifts of both levels. Using an interleaved servo in which the optical transition is alternately interrogated both with and without the 653-nm laser, the shift is determined from the difference frequency in the two configurations. In a single clock cycle, all three Zeeman pairs are measured: each of the six transitions |S1/2, ±1/2⟩ − |D3/2, ±Mf⟩ for Mf = 1/2, 3/2, and 5/2 is interrogated on
both sides of each transition, both with and without the 653-nm Stark shift beam. All 24 measurements are repeated 
N = 50 times. From these measurements error signals are
derived to track the center and splitting for each Zeeman pair |S_{1/2}, ±1/2⟩ − |D_{3/2}, ±M_J⟩, both with and without the
Stark shift. The ratio δ_0(ω)/δ_2(ω) is inferred at each step from the
appropriate frequency differences. The servo is first run
for several clock cycles, to ensure each shifted and unshifted
the appropriate frequency differences.

To estimate the bias, we use theory to calculate
δ_0(ω)/δ_2(ω) at the frequencies used in the measurements,
fit the resulting points to a straight line, and compare the
interpolated zero crossing to that calculated from theory.
This procedure indicates that a straight line fit would underestimate
the zero crossing by ≈2.2 GHz. This does not change signif-
ically if one offsets the frequencies used in the calculation
by the small difference between the experimentally measured and
calculated zero crossing. Consequently we apply 2.2 GHz
as a correction, but we add, in quadrature, the full size of the
correction to the uncertainty to obtain the final value of
ω_0 = 2π × 459.1614(28) THz. Note that, near to ω_0, the 614-nm transition has a significant contribution to the curvature of
both Δa_0(ω) and a_2(ω), but these contributions tend to cancel
in the ratio. This explains the excellent linearity in Fig. 2 and
the correspondingly small bias correction.

IV. DISCUSSION

The measured ω_0 is in excellent agreement with the the-
etorical value of 459.10(92) THz given in Ref. [15]. The


delta_0/delta_2

Delta (THz)

FIG. 2. Ratio of scalar to tensor shifts as a function of frequency
difference relative to 459.1 THz. Error bars are smaller than the
points and have been omitted. Measurements at Δf = −0.2 and
−0.6 THz were measured three times giving 16 measurements in total. The dashed line is from a χ^2 fit.

theory used more accurate, experimentally determined values
of ⟨6P_{1/2}||6S_{1/2}⟩ and ⟨6P_{3/2}||6S_{1/2}⟩ from [4], but
⟨6P_{1/2}||5D_{5/2}⟩ and all uv contributions were determined
from atomic structure calculations. Thus, confirmation of the
theoretically estimated zero crossing strongly supports the
accurate theoretical assessment of the uv contributions.

As discussed in Ref. [15], the zero crossing of Δa_0(ω),
455- and 493-nm transition matrix elements, and branching
ratios for 6P_{3/2} decays can be combined to provide an accurate
model of Δa_0(ω) over a wide wavelength range. Accurate
matrix elements have already been reported [4], and we
have recently carried out improved measurements of 6P_{3/2}
branching ratios [16]. Thus the measurement here now allows
us to make an accurate assessment of the room-temperature
blackbody radiation (BBR) shift for the Ba^+ clock transition.
More generally, we give a parametrized model for Δa_0(ω) for
wavelengths above 700 nm.

Applying the formalism in Ref. [15], we approximate
Δa_0(ω) by

Δa_0(ω) = Δa_0^{vis}(ω) − Δa_0^{vis}(ω_0)[1 − (ω/ω_0)^2] /
1 − (ω/ω_0)^2

(6)

where ω_0 = 0.2049(42) a.u. is the position of an effective pole that approximates uv contributions and valence-core corrections [15], and Δa_0^{vis}(ω) is the contribution from the
three transitions at 455, 493, and 614 nm. Explicitly,

Δa_0^{vis}(ω) = 1/ω_{455} + 1/ω_{493} − 1/ω_{614} ×

(7)

where ω_{455}, ω_{493}, and ω_{614} are the frequencies of the
associated transitions expressed in atomic units. The parameters
μ_{455} and μ_{493} are the reduced dipole matrix elements

μ_{455} = ⟨6P_{1/2}||6S_{1/2}⟩ = 4.7017(27) a.u.,

(8)

μ_{493} = ⟨6P_{1/2}||6S_{1/2}⟩ = 3.3251(21) a.u.

(9)

reported in Ref. [4], and

p = 0.763104(65)

(10)

is from recent branching ratio measurements [16]. From
the model we get Δa_0(0) = −73.56(21) a.u., consistent
with −73.1(1.3) a.u. from theory. Similarly, Δa_0(ω_{762}) =
−78.51(21) a.u., where ω_{762} is the clock transition fre-
quency. For both values given, uncertainties from μ_{455}
and μ_{493} are assumed correlated and added absolutely, with the
result added in quadrature with the uncertainty contributions
from p, ω_0, and ω_{762}. A summary of the uncertainty contri-
butions for Δa_0(0) is given in Table I and those for Δa_0(ω_{762})
are very similar.
The BBR shift of a clock transition can be written in the form
\[ h\delta v = -\frac{\Delta \alpha_0(0) E_{\text{BBR}}^2}{2} \left( \frac{T}{T_0} \right)^4 \left[ 1 + \eta \left( \frac{T}{T_0} \right)^2 \right], \] (11)
where \( T_0 = 300 \text{ K}, \ E_{\text{BBR}} = 831.945 \text{ V m}^{-1}, \) and \( \eta \) is a dynamic correction factor [19]. From the model we estimate the fractional BBR shift at 300 K to be 3.724(11) \times 10^{-15} with a dynamic correction factor \( \eta = 1.6 \times 10^{-3}. \) Thus \( \eta \) contributes less than the uncertainty due to the determination of \( \Delta \alpha_0(0). \) We note that all uncertainties quoted are 1σ. As discussed in Ref. [16], we would recommend taking a 2σ uncertainty in the assessment of clock performance, for example.

V. SUMMARY

In summary we have provided an accurate determination of the zero crossing in the dynamic differential scalar polarizability of the \( S_{1/2} - D_{5/2} \) clock transition in \(^{138}\text{Ba}^+\). Measurements utilized a ratio of scalar to tensor components of the differential polarizability so as to eliminate intensity variations in the Stark-shifting laser. The value of 459.1614(28) THz is in excellent agreement with theory. Following the proposal in Ref. [15] we have provided a representation of \( \Delta \alpha_0(\omega) \) that is accurate at the 0.5% level for wavelengths above 700 nm. In principle the model also applies for shorter wavelengths but care would be needed in the vicinity of the zero crossing. The model will allow accurate calibration of polarizabilities for other clock transitions via comparison with shifts induced on the clock transition in \(^{138}\text{Ba}^+\).

ACKNOWLEDGMENTS

This work is supported by the National Research Foundation, Prime Minister’s Office, Singapore and the Ministry of Education, Singapore under the Research Centres of Excellence program. M.S.S acknowledges the sponsorship of ONR Grant No. N00014-17-1-2252.