

Measurement of the Differential Static Scalar Polarizability of the $^{88}\text{Sr}^+$ Clock Transition

T. Lindvall^{✉,*}, K. J. Hanhijärvi[✉], T. Fordell[✉], and A. E. Wallin[✉]

VTT Technical Research Centre of Finland Ltd, National Metrology Institute VTT MIKES, P.O. Box 1000, FI-02044 VTT, Finland



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We report on a precision measurement of the differential static scalar polarizability $\Delta\alpha_0$ of the $^2S_{1/2} \rightarrow ^2D_{5/2}$ optical clock transition in the $^{88}\text{Sr}^+$ ion. The polarizability was determined from the “magic” ion-trap drive frequency where the micromotion-induced second-order Doppler and quadratic Stark shifts cancel, using a single clock in an interleaved scheme by switching between minimized and large micromotion. By measuring at different Mathieu q_z parameters, $\Delta\alpha_0$ can be obtained without prior knowledge of the angle between the rf electric field and the trap axis, which would otherwise dominate the systematic uncertainty. For validation, measurements were carried out at different micromotion levels and by displacing the ion in opposite directions. The results show excellent consistency and our value, $\Delta\alpha_0 = -4.8314(20) \times 10^{-40} \text{ Jm}^2/\text{V}^2 = -29.303(12) \text{ au}$, reduces the uncertainty by a factor of 3.5 compared to a previous measurement, while showing a discrepancy of 5σ . Our measurement reduces the polarizability-related uncertainty of the $^{88}\text{Sr}^+$ clock to 2.2×10^{-19} for a blackbody radiation temperature of 295 K—a significant step towards total uncertainties $< 1 \times 10^{-18}$. Using existing polarizability transfer schemes, the result can reduce the uncertainty also for other ion species.

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Optical atomic clocks [1] with fractional frequency uncertainties around 1×10^{-18} are sensitive probes for new physics [2], enable applications such as relativistic geodesy [3], and are expected to lead to a redefinition of the second in the International System of Units (SI) [4]. In many optical clocks, the frequency shift due to the ambient blackbody radiation (BBR) is the dominating source of uncertainty. The atomic response to BBR is determined by the differential static scalar polarizability (DSSP) $\Delta\alpha_0$ of the clock transition, with a small dynamic correction from the variation in polarizability over the BBR spectrum.

For ion clocks, theoretical DSSP values have relative uncertainties around 1%–4% for monovalent ions and worse for more complex ones [5]. Considerable efforts have therefore been put into experimental assessment of DSSPs. Comparing room-temperature and cryogenic clocks is a very fundamental method, but has so far achieved only a 2.6% uncertainty [6]. Measuring the ac Stark shift induced by (near)-infrared laser light has been used for different ion species [7–11], but the uncertainty is limited to $\gtrsim 2\%$ due to the need to determine the absolute light intensity at the ion. This can be avoided by instead measuring magic wavelengths, where the differential

dynamic polarizability is zero, but extrapolation to dc limits the uncertainty to $\gtrsim 0.5\%$ [12–14].

For ions whose clock transition has a negative DSSP, the micromotion-induced second-order Doppler (time dilation) and quadratic Stark shifts cancel at a particular “magic” trap drive frequency. This has been used to measure the DSSP in $^{88}\text{Sr}^+$ [15] and $^{40}\text{Ca}^+$ [16] with uncertainties of 1.5×10^{-3} and 2.9×10^{-4} , respectively. This makes it by far the most accurate method to determine DSSP values and different methods to transfer the accuracy to ions with positive DSSP have therefore been proposed and demonstrated [12,17,18].

We have measured the differential polarizability $\Delta\alpha_0$ of the $^{88}\text{Sr}^+ ^2S_{1/2} \rightarrow ^2D_{5/2}$ clock transition, a secondary representation of the second, using a single ion in an interleaved measurement, switching between minimized and large applied excess micromotion (EMM). We will refer to these two servo locks as the reference and high-EMM clocks, respectively. This method has the benefit that most frequency shifts, except for the micromotion shifts under study, are common mode to the two clocks. We show that by measuring the magic frequency for different Mathieu q_z parameters, the DSSP can be obtained without knowledge of the angle between the rf electric field and the trap axis. Our result reduces the uncertainty by a factor of 3.5 compared to [15], while showing a discrepancy of 5σ , corresponding to a fractional frequency correction of -4×10^{-18} at room temperature.

The international metrology community has prepared a roadmap with mandatory criteria that must be fulfilled

*Contact author: thomas.lindvall@vtt.fi

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before a redefinition of the SI second can take place [4]. Criterion I.1 on uncertainty budgets requires multiple optical clocks with systematic uncertainties $\lesssim 2 \times 10^{-18}$, while criterion I.2 on validation via frequency ratios sets a target uncertainty $\lesssim 5 \times 10^{-18}$ for clock comparisons. The -4×10^{-18} correction corresponding to the observed polarizability discrepancy is very significant compared to these target uncertainties.

Since most optical clocks are operated under similar conditions, same-transition clock comparisons are generally not able to detect errors in common atomic parameters, and independent measurements of the parameters are therefore crucial. This is true in particular for the BBR shift and DSSP, as most clocks operate close to room temperature. Our result will thus benefit the redefinition work, the wide range of $^{88}\text{Sr}^+$ clocks being developed [19–23], and, via polarizability transfer, clocks based on ions with positive DSSP such as $^{171}\text{Yb}^+$. In addition to $^{88}\text{Sr}^+$ and $^{40}\text{Ca}^+$, our method can be applied to $^{138}\text{Ba}^+$ [24], $^{226}\text{Ra}^+$ [25], and the secondary transition in $^{176}\text{Lu}^+$ [26].

Experimental setup—The end-cap ion trap [27] and laser-beam geometry is shown in Fig. 1 (for details on the laser setup and clock interrogation, see End Matter). EMM is measured and minimized using photon correlation [28] with three noncoplanar 422-nm cooling laser beams (labeled A–C in Fig. 1). After minimization, the rms rf electric field is $E_{\text{rms}} \lesssim 10$ V/m.

To apply a large, well-defined EMM, we iteratively determined a combination of dc bias voltages that minimizes the photon-correlation contrast for the cooling and probe (B) and state-preparation (A) beams, while causing a large contrast for the third beam (C). Since the relation between the applied dc electric field and the resulting

rf field depends on the Mathieu q_i and a_i parameters ($i = x, y, z$ labels the trap axes), proportional to the rf and dc electric field gradients, respectively, a slightly different voltage combination had to be found for each set of parameters. The dc fields applied for the high-EMM clock were 0.47–1.1 kV/m, resulting in rms rf fields $E_{\text{rms}} = 4.3 \dots 6.8$ kV/m. The rf field is orthogonal to the cooling and probe beam (B), but since the q_i parameters have opposite signs in the axial and radial directions, the ion displacement is nearly along the beam (at a 6–7° angle), see Fig. 1. Thus even the maximum ion displacement, 8 μm , causes no significant change in the cooling and probe beam intensity at the ion. The measurements utilized the full tuning range, 14.22–14.61 MHz, of the helical resonator, see End Matter for details.

Theory—The scalar shift caused by EMM is the sum of the second-order Doppler shift, $\Delta\nu_{\text{D2}} = -(\nu_0/2)\langle v^2 \rangle/c^2$, and the quadratic Stark shift, $\Delta\nu_{\text{QS}} = -\Delta\alpha_0/(2h)\langle E^2 \rangle$. Here, ν_0 is the transition frequency, c is the speed of light, and h is the Planck constant. From the solution of the Mathieu equation for EMM [29], the spectrum of the ion's velocity v relates to that of the rf electric field E by $\langle v^2(n\Omega) \rangle = (e/m\Omega)^2 \langle E^2(n\Omega) \rangle / n^2$, where Ω is the trap drive frequency, e is the elementary charge, m is the ion mass, and $n = 1, 2, 3, \dots$. This gives the total scalar shift

$$\Delta\nu_s = -\frac{\nu_0}{2} \sum_{i=x,y,z} \sum_{n=1}^{\infty} \left[\frac{\Delta\alpha_0}{h\nu_0} + \frac{1}{n^2} \left(\frac{e}{mc\Omega} \right)^2 \right] \langle E_i^2(n\Omega) \rangle. \quad (1)$$

To lowest order in n , Eq. (1) has a zero crossing at $\Omega_0^0 = e/(mc)\sqrt{-h\nu_0/\Delta\alpha_0}$ for an ion with $\Delta\alpha_0 < 0$. In previous work [15,16], terms up to $n = 2$ were included, but for the highest q_z values used here, the third-order term is not negligible and is thus included. Then, from the Mathieu solution [29], the ratios of the field intensities at 2Ω and 3Ω relative to that at Ω are given by

$$\rho_{2,i} = \frac{\langle E_i^2(2\Omega) \rangle}{\langle E_i^2(\Omega) \rangle} = \left(\frac{4q_i}{a_i - 16 - q_i^2/(a_i - 36)} \right)^2, \quad (2a)$$

$$\rho_{3,i} = \frac{\langle E_i^2(3\Omega) \rangle}{\langle E_i^2(\Omega) \rangle} = \left(\frac{9q_i^2}{(a_i - 36)(a_i - 16) - q_i^2} \right)^2. \quad (2b)$$

The first harmonic of the rf field, $\mathbf{E}(\Omega)$, has a well-defined direction $\mathbf{u}_\Omega = \mathbf{E}(\Omega)/|\mathbf{E}(\Omega)|$ orthogonal to the beams A and B, see Fig. 1, as we minimize their photon-correlation contrasts at Ω . Noting that $\langle E_i^2(\Omega) \rangle = u_{\Omega,i}^2 \langle E^2(\Omega) \rangle$ and defining the mean ratios $\bar{\rho}_j = \sum_i u_{\Omega,i}^2 \rho_{j,i}$, we can rewrite Eq. (1) as

$$\Delta\nu_s \approx -\frac{\Delta\alpha_0}{2h} \left[1 - \left(\frac{\Omega_0^0}{\Omega} \right)^2 \frac{1 + \bar{\rho}_2/4 + \bar{\rho}_3/9}{1 + \bar{\rho}_2 + \bar{\rho}_3} \right] \langle E^2 \rangle, \quad (3)$$

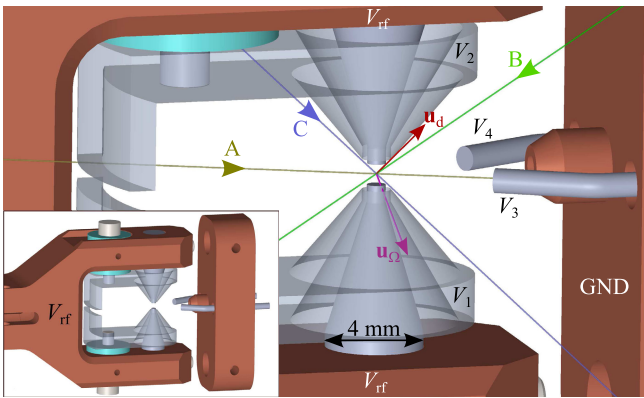


FIG. 1. Trap assembly (inset) and laser-beam geometry. B is the main cooling and probe beam, while A is the state-preparation beam. All beams A–C are used for micromotion measurements. For high excess micromotion, a combination of dc bias voltages V_1 – V_4 is applied that causes micromotion along $\mathbf{u}_\Omega$, orthogonal to the A and B beams, while displacing the ion along $\mathbf{u}_d$, at a 6–7° angle to the B beam. V_{rf} , trap rf voltage; GND, ground.

where $\langle E^2 \rangle \approx (1 + \bar{\rho}_2 + \bar{\rho}_3) \langle E^2(\Omega) \rangle$ is the total mean-square rf field. For a constant applied dc field, $\langle E^2 \rangle$ depends on the Mathieu parameters but not on Ω .

In [27], the rf gradient of our trap was found to be radially symmetric within experimental uncertainty. As the a_i parameters are very small, $|a_i| \ll 1$, we can initially neglect them and finally evaluate the small correction they cause. This justifies neglecting the radial asymmetry and writing $\bar{\rho}_j = \rho_{j,z} \cos^2 \theta_\Omega + \rho_{j,r} \sin^2 \theta_\Omega$, where θ_Ω is the angle between $\mathbf{u}_\Omega$ and the trap z axis (vertical in Fig. 1) [32].

Defining the zero crossing of Eq. (3) as

$$\Omega_0 = \Omega_0(\bar{\rho}_2, \bar{\rho}_3) = \Omega_0^0 \sqrt{\frac{1 + \bar{\rho}_2/4 + \bar{\rho}_3/9}{1 + \bar{\rho}_2 + \bar{\rho}_3}}, \quad (4)$$

the expression in square brackets in (3) becomes $1 - (\Omega_0/\Omega)^2$. This function is not well suited for fitting to data over a narrow frequency range, so we use a second-order series expansion in δ/Ω_0 , where $\delta = \Omega - \Omega_0$ is the detuning, which yields

$$\Delta\nu_s \approx k \left[\frac{\delta}{\Omega_0} - \frac{3}{2} \left(\frac{\delta}{\Omega_0} \right)^2 \right], \quad (5)$$

where $k = (-\Delta\alpha_0/h) \langle E^2 \rangle$. For our parameters, the maximum value of the second-order term is of the same order as the statistical uncertainties, while the third-order term is negligible. Since we keep q_z constant when changing Ω (and $|a_i| \ll 1$), $\langle E^2 \rangle$ is constant and Eq. (5) has only two fit parameters, the zero crossing Ω_0 and the slope at the zero crossing, k . By contrast, in [15,16] the ion secular frequencies were kept constant, which causes $\langle E^2 \rangle$ to have a complicated dependence on Ω and necessitates using a physically less meaningful fit function with three free parameters.

Differential frequency shifts—Although most frequency shifts are common mode to the two clocks, we must carefully evaluate those that may differ. A large first-order Doppler shift would occur if the ion were probed during the displacement of its equilibrium position following the switching of the voltages. This was studied, see End Matter for details, and we estimate that a dwell time of 300 ms between the switching and probing gives negligible shifts of $<30 \mu\text{Hz}$ for all measurements, which we include as uncertainties.

Since the trap has only two bias electrodes in the radial plane, changing the electric field at the position of the ion also changes its gradient [27], causing a small change in the electric quadrupole shift (EQS). This does not affect the scalar shift studied here due to the efficient EQS cancellation scheme [27,33], but it prevents us from determining the tensor polarizability, as the EQS and the tensor Stark shift, both tensor shifts, cannot be reliably distinguished.

We measured the ion temperature and heating rate without and with applied EMM for different q_z and Ω using carrier Rabi flopping. In most cases, the difference in estimated mean ion temperature was small, $\lesssim 0.3 \text{ mK}$. However, for a few secular frequencies we observed an increased heating rate. These resonances were narrower than the small change in secular frequencies due to the change in dc gradient with applied EMM and lead to a temperature difference of $\approx 2 \text{ mK}$ magnitude for two measurements (out of 24). These were corrected for, and a conservative uncertainty of 1 mHz (1.4 mK) was assigned to the differential thermal second-order Doppler shift of all measurements.

Measurements—Clock interrogation times of 124 or 137 ms were used. The frequency difference between the high-EMM and reference clocks averaged down as $\approx 7 \times 10^{-15} \tau^{-1/2}$, resulting in a statistical uncertainty around 10 mHz after one day of averaging for each combination of parameters (rf frequency, q_z , and ion displacement). The secular frequencies were measured before and after each measurement to determine the Mathieu parameters, see End Matter for details.

The maximum ion displacement was ultimately limited by the range of the bias voltage supply, but also by the finite field of view of the fluorescence detection. The measurements at $q_z = 0.34 \dots 0.46$ were carried out with small a_i parameters of $(-0.0014, 0.0004, 0.0010)$ used to minimize the EQS [27]. For even lower q_z , we were not able to achieve high EMM shifts without a significantly increased risk of ion loss when switching the voltages. For the highest q_z values of 0.60 and 0.71, the trap was operated with increased a_i parameters of $(0.0047, 0.0069, -0.0117)$ in order to achieve a sufficient ion displacement.

Analysis and results—Figure 2 shows an example of measured EMM-induced shifts fitted with Eq. (5). For the

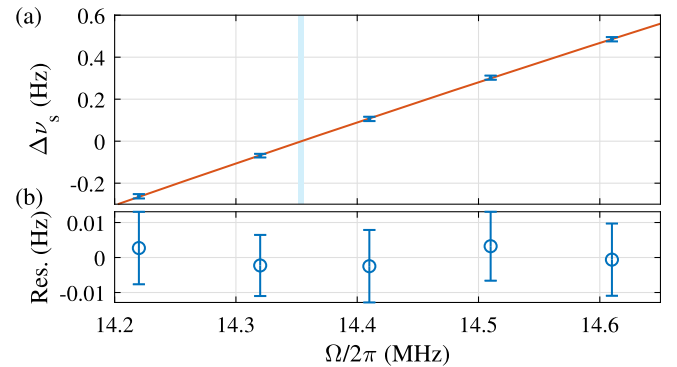


FIG. 2. (a) Measured micromotion-induced scalar frequency shifts with statistical uncertainties (error bars) as function of the rf drive frequency for $q_z = 0.37$ and an applied dc field of 0.65 kV/m. The line is a fit of Eq. (5) and the shaded area indicates the uncertainty of the zero crossing frequency ($\pm 1\sigma$). (b) Residuals of fit in (a).

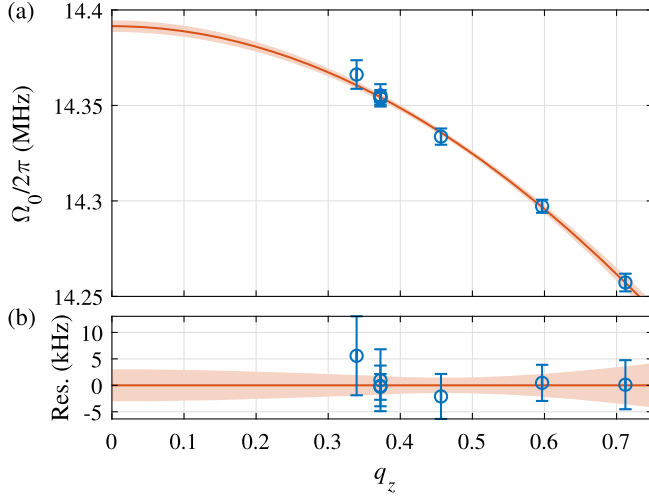


FIG. 3. (a) Measured zero crossing frequencies with statistical uncertainties (error bars) as function of q_z . Each point is the result of a fit like in Fig. 2. The uncertainties of the q_z values are $\lesssim 3 \times 10^{-4}$ and the corresponding error bars are not visible. The line is a fit of Eq. (4) and the shaded area the corresponding uncertainty ($\pm 1\sigma$). (b) Residuals of fit in (a).

remaining six measurements, 3–4 points were measured. In total, we collected 23.4 days of data for 24 points.

The measured zero crossing frequency depends on the angle θ_Ω between $\mathbf{u}(\Omega)$ and the trap axis. Based on the laser beam directions, the angle between $\mathbf{u}(\Omega)$ and the vertical *chamber* axis is $32(1)^\circ$, but the direction of the trap axis is not known as accurately. In previous work [15,16], the angle was determined from the tensor shift measured with the magnetic field aligned with the trap axis. In addition to relying on theoretical tensor polarizability values, this method requires estimates of the direction of the trap axis and the accuracy of the magnetic field direction. It is also unclear whether the EQS was taken into account, and we noted that even a small change in it prevented us from determining the tensor polarizability.

In our case, the angle θ_Ω would dominate the systematic uncertainty. Instead of evaluating its value and uncertainty using multiple assumptions, we measure Ω_0 over a wide range of q_z parameters, which allows determining the DSSP without knowledge of the angle. Neglecting the small a_i parameters, the zero crossing, Eq. (4), depends only on Ω_0^0 , θ_Ω and q_z , and the first two parameters can be obtained from a fit of Ω_0 as a function of q_z , see Fig. 3. This gives $\Omega_0^0/2\pi = 14.3915(30)$ MHz and $\theta_\Omega = 31.2(3.1)^\circ$, in agreement with the nominal angle given above.

To demonstrate the robustness of the q_z fitting method, we repeat the fitting using (i) the second-order Mathieu solution, which yields $\Omega_0^0/2\pi = 14.3916(30)$ MHz and $\theta_\Omega = 30.9(3.1)^\circ$, and (ii) a series expansion to order q_z^2 , which yields $\Omega_0^0/2\pi = 14.3909(30)$ MHz and $\theta_\Omega = 32.5(2.9)^\circ$ [29]. This shows that the angle θ_Ω adapts to errors in the fit function, while the effect on Ω_0^0 , and thus

TABLE I. $\Delta\alpha_0$ uncertainty budget.

Contribution	Uncertainty ($10^{-43} \text{ Jm}^2/\text{V}^2$)
Statistical or fitting	2.0
q_z parameter	0.0072
Ion temperature	0.22
First-order Doppler (dwell time)	0.0046
Reference-clock residual EMM	0.0003
a_i parameters	0.053
Trap anharmonicity	0.0020
Total	2.0

$\Delta\alpha_0$, is relatively small even for significant errors or approximations.

From the measured residual photon-correlation contrasts with high EMM, we estimate that the applied micromotion is orthogonal to the A and B beams with an uncertainty $\lesssim 0.9^\circ$. However, any variations in the direction are accounted for by the fit uncertainty. The systematic uncertainty contributions from q_z , the first-order Doppler shift, and the ion temperature difference were evaluated using a Monte Carlo simulation. The effect of reference-clock residual EMM was found to be negligible, see End Matter for details. To take into account the so far neglected a_i parameters, we correct the measured zero-crossing frequencies using the square root in Eq. (4) evaluated with and without a_i parameters [34]. Fitting the third-order solution gives $\Omega_0^0/2\pi = 14.3916(30)$ MHz and $\theta_\Omega = 31.1(3.1)^\circ$. The uncertainty contribution from the a_i parameters is conservatively taken as the difference in $\Delta\alpha_0$ between included and neglected a_i .

We also considered the effect of anharmonic terms in the trap potential and found that the q_z fitting reduces the corresponding DSSP error by one order compared to a measurement at a single q_z value [29]. This completes the uncertainty budget, summarized in Table I, and gives the final DSSP value $\Delta\alpha_0 = -4.8314(20) \times 10^{-40} \text{ Jm}^2/\text{V}^2 = -29.303(12)$ au.

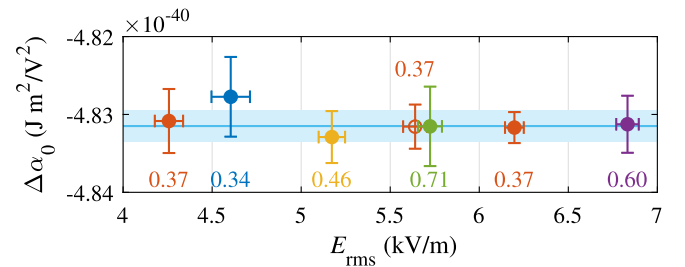


FIG. 4. $\Delta\alpha_0$ values from the individual measurements as function of the rms rf field with q_z values below or above. For the open circle, the ion was displaced in the opposite direction. The line and shaded area are the $\Delta\alpha_0$ value and uncertainty from the q_z fitting.

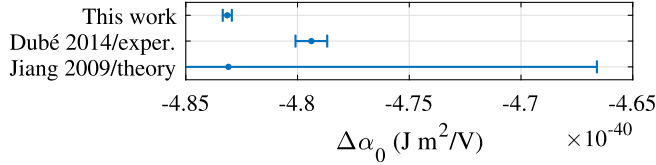


FIG. 5. $\Delta\alpha_0$ reported in this Letter compared to previous experimental [15] and theoretical [36] values.

For $q_z = 0.37$, we measured the zero crossing with three different ion displacements, one of them in the opposite direction ($-\mathbf{u}_d$ in Fig. 1). As seen from Fig. 3, these agree well. With the angle θ_Ω known, we can also solve the DSSP separately from each measurement. These results are shown in Fig. 4 as a function of E_{rms} , solved from the slope k in Eq. (5). Again, no trends with E_{rms} , q_z , or the direction of displacement are seen. For simplicity, we have here neglected the correlation between the fitted θ_Ω and the individual measurements.

Discussion—As seen from Fig. 5, our $\Delta\alpha_0$ value shows a discrepancy of 5σ with the experimental result in [15]. The reason for this is unknown. For example, an error in the angle between the rf field and the trap axis in [15] could only explain about half of the difference. However, we note that the high-EMM clock used in [15] was not very well characterized [35]. With only a single laser beam, there was limited knowledge about the ion temperature and EMM. As the EMM could not be minimized, there was also no reference measurement where the two clocks agreed, but the measurement relied solely on the estimated systematic shifts. With measurements at a single EMM level, any undiagnosed frequency offset between the two clocks would cause an offset in the DSSP value. By contrast, since we get consistent results at different EMM levels, a bias in our DSSP value could only be caused by an unknown shift proportional to $\langle E^2 \rangle$. Our value is close to the theoretical DSSP value [36] but has an 80 times smaller uncertainty and can therefore provide a benchmark for future atomic structure calculations.

Our $\Delta\alpha_0$ value reduces the polarizability-related uncertainty of the $^{88}\text{Sr}^+$ clock to 2.2×10^{-19} for a BBR temperature of 295 K. Reducing the BBR-field-related uncertainty to the same level would require a BBR temperature uncertainty of 31 mK. Still, since our uncertainty is dominated by statistics, the q_z fitting method has potential to further reduce the $\Delta\alpha_0$ uncertainty to a level limited by the repeatability of the applied micromotion and the remaining systematics, see End Matter for details.

Our $\Delta\alpha_0$ value implies a frequency correction of -4×10^{-18} for room-temperature $^{88}\text{Sr}^+$ clocks that have used the value from [15]. Among published results, this correction is significant compared to the total uncertainty only for the $^{88}\text{Sr}^+ / ^{171}\text{Yb}^+(\text{E}3)$ optical frequency ratio in [22].

Summary—We have measured the differential polarizability $\Delta\alpha_0$ of the $^{88}\text{Sr}^+$ clock transition by utilizing the

magic trap drive frequency where the micromotion shifts cancel. The measurements were carried out using a single ion trap in an interleaved scheme, resulting in common-mode rejection of most other shifts. We have showed that by measuring at different Mathieu q_z values, $\Delta\alpha_0$ can be obtained without knowledge of the angle between the rf field and the trap axis. Our measurements show excellent consistency and the fractional uncertainty of 4.1×10^{-4} , a factor of 3.5 lower than the previous value, reduces the polarizability-related uncertainty of the $^{88}\text{Sr}^+$ clock to 2.2×10^{-19} for a BBR temperature of 295 K. Using existing polarizability transfer schemes, the improved uncertainty can benefit also other ion species.

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Data availability—The data that support the findings of this Letter are openly available [37].

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End Matter

Laser system and interrogation sequence—The ion is Doppler cooled using a commercial external-cavity diode laser at 422 nm, frequency stabilized to a nearby rubidium line [38] using modulation transfer spectroscopy. For repumping at 1092 nm and clock-state clearout at 1033 nm, unpolarized amplified-spontaneous-emission (ASE) light sources [39,40] are used. After cooling, the main cooling beam (B in Fig. 1) is switched off and a separate 422-nm state-preparation beam (A in Fig. 1), propagating along the magnetic field direction and whose polarization can be controlled using a liquid-crystal phase retarder, is briefly switched on with σ^+ or σ^- polarization to transfer the ion into the desired ground-state sublevel. The two beams are controlled using independent double-pass acousto-optic modulators (AOMs), while a common mechanical shutter ensures that the 422-nm light is fully extinguished during clock interrogation. The clock transition is probed using a commercial 1348-nm external-cavity diode laser, which is frequency doubled to 674 nm and stabilized to a 30-cm horizontal ultra-low-expansion (ULE) glass cavity of a design similar to [41]. After probing, the cooling laser and repumper are switched on for state detection. Then a short 1033-nm clearout pulse is applied to return the ion from the clock state to the cooling cycle, followed by a second state detection to verify fluorescence at 422 nm. Ion fluorescence is detected using an objective close to the antireflection-coated fused silica window of the vacuum system. A beam splitter divides the detected photons between a photomultiplier tube (PMT, photon collection efficiency $\approx 1/550$) and an sCMOS camera.

Both the reference and high-EMM clocks probed six $S_{1/2} \rightarrow D_{5/2}$ Zeeman components, $\pm 1/2 \rightarrow \pm 1/2$, $\pm 1/2 \rightarrow \pm 3/2$, and $\pm 1/2 \rightarrow \pm 5/2$, in order to cancel the electric quadrupole shift and other tensor shifts [27,33] in addition to the linear Zeeman shift. Each frequency (red and blue side of six transitions) was probed twice per cycle for both clocks. The order of probing the red and blue sides was alternated in order to avoid noise-induced servo errors

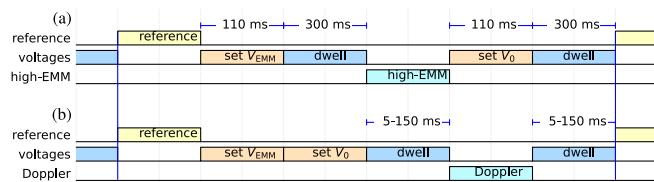


FIG. 6. (a) Interleaved clock sequence for measuring the DSSP. (b) Interleaved clock sequence for measuring the first-order Doppler shift with variable dwell time. The blue vertical lines indicate the start and end of an interleaved clock cycle, with a total duration of 8–9 s (depending on the probe time). V_{EMM} , bias voltages for high EMM; V_0 , bias voltages for minimized EMM.

[42]. The interleaved clock sequence is schematically shown in Fig. 6(a). The frequency difference between the high-EMM and reference clocks was evaluated from the frequency difference of the final probe-laser AOM before the ion.

Trap drive—To allow accurate and repeatable tuning of the resonance frequency between 14.22 and 14.61 MHz, our tunable helical resonator [27] was equipped with a micrometer tuning screw. We use a phase-continuously tunable direct digital synthesizer (DDS) rf source (Sinara Urukul/AD9912) with a power amplifier and rf transformer (for galvanic isolation) to deliver rf to the helical resonator input, which allows changing the frequency without ion loss. We adjust q_z coarsely using a digital step attenuator with 0.5-dB resolution and finely, to keep q_z constant when changing the rf frequency, using the 10-bit full-scale current register of the DDS (relative amplitude resolution 10^{-3}).

First-order Doppler shift—The dc voltage source channels have time constants of ≈ 30 ms, including the low-pass filters at the electric vacuum feedthroughs. We must therefore introduce a dwell time between the voltage switching and the probe pulse to ensure that the first-order Doppler shift due to the displacement of the ion's equilibrium position is at a negligible level. To determine the required dwell time, the clock sequence in Fig. 6(b) was used to measure the residual Doppler shift with different dwell and probe times. For technical reasons, the four voltage channels are switched one at a time, with a total duration of ≈ 110 ms.

For the shortest dwell times, the initial Doppler shift is large enough that the first probe pulse after switching does not excite the ion. Because of the alternating red and blue probing (see above), the servo tracking still works well. Even for a short interrogation time of 36.5 ms, the excitation probability returned to normal already for a dwell time of 50 ms. Here, the measured line center shift was ≈ 500 mHz and changed sign with the ion displacement direction. In the ≥ 50 ms dwell-time regime, a model that assumes that an exponentially decaying instantaneous frequency shift is averaged over each probe pulse gives a good fit. This allows extrapolating to longer dwell and probe times, and we estimate that a dwell time of 300 ms gives negligible shifts of < 30 μHz for all measurements. The agreement between the Ω_0 (Fig. 3) and $\Delta\alpha_0$ values (Fig. 4) measured with opposite ion displacement also confirms that the 300-ms dwell time is sufficient.

Secular frequencies and Mathieu parameters—Secular frequencies were measured using the tickler voltage and photon correlation method [27]. For the highest q_z values of 0.60 and 0.71, the accuracy of the method to

determine Mathieu parameters presented in [27] is not sufficient. The obtained q_z and a_i parameters were therefore iteratively corrected by estimating their error, correcting for it, and estimating the new error. This method is based on the fact that the secular frequencies can be calculated to arbitrary precision from the Mathieu parameters, but not vice versa. After two iterations, the systematic error of the obtained parameters was negligible compared to their stability or repeatability. The latter was estimated as the standard deviation of all values measured for a certain q_z and ion displacement (including all measured rf frequencies).

Reference-clock EMM—The EMM of the reference clock was measured and, if required, minimized before and after each measurement. From these, we interpolate the mean rf electric field during each EMM-shift measurement. An rms field $\lesssim 40$ V/m and a corresponding frequency shift of magnitude $\lesssim 12$ μ Hz were found for all measurements. EMM in the direction of the applied (high) EMM cancels in the differential measurement, while EMM in the orthogonal direction causes a bias in the measured shift. The angle between the estimated reference EMM and the applied EMM

varied between 20° and 70° , so some suppression is expected, but the uncertainty contribution from the reference clock EMM was conservatively estimated by correcting for the reference EMM shifts in full, repeating the fitting, and taking the change in $\Delta\alpha_0$ as the uncertainty. This yielded a negligible contribution as shown in Table I.

Uncertainty reduction—As our q_z fitting method reduces the angle θ_Ω to a fit parameter, it allows further reduction of the total DSSP uncertainty to a level limited by the reproducibility of the applied EMM and the remaining systematics if the statistical uncertainty can be reduced. Using two separate clocks instead of an interleaved measurement would reduce the instability of the frequency difference by a factor of ≈ 1.5 for our parameters, providing a statistical advantage despite the need for separate reference measurements. A two-clock measurement would also allow significantly lower q_z values to be used, which would reduce the fitting uncertainty. Finally, even higher EMM levels would allow determining the zero crossing frequency with lower uncertainty.