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115 In $^+\text{-}^{172}$ Yb $^+$ Coulomb Crystal Clock with 2.5×10^{-18} Systematic Uncertainty

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(Received 30 April 2024; revised 25 October 2024; accepted 19 November 2024; published 16 January 2025)

We present a scalable mixed-species Coulomb crystal clock based on the ${}^{1}S_{0} \leftrightarrow {}^{3}P_{0}$ transition in ${}^{115}\text{In}^{+}$. ${}^{172}\text{Yb}^{+}$ ions are cotrapped and used for sympathetic cooling. Reproducible interrogation conditions for mixed-species Coulomb crystals are ensured by a conditional preparation sequence with permutation control. We demonstrate clock operation with a 1In^{+} - 3Yb^{+} crystal, achieving a relative systematic uncertainty of 2.5×10^{-18} and a relative frequency instability of $1.6 \times 10^{-15}/\sqrt{\tau/1 \text{ s}}$. We report on absolute frequency measurements with an uncertainty of 1.3×10^{-16} and optical frequency comparisons with clocks based on ${}^{171}\text{Yb}^{+}$ (E3) and ${}^{87}\text{Sr}$. With a fractional uncertainty of 4.4×10^{-18} , the former is—to our knowledge—the most accurate frequency ratio value reported to date. For the ${}^{115}\text{In}^{+}/{}^{87}\text{Sr}$ ratio, we improve upon the best previous measurement by more than an order of magnitude. We also demonstrate operation with four ${}^{115}\text{In}^{+}$ clock ions, which reduces the instability to $9.2 \times 10^{-16}/\sqrt{\tau/1 \text{ s}}$.

DOI: 10.1103/PhysRevLett.134.023201

Optical clocks with 10^{-18} level fractional frequency uncertainties [1] enable precise tests of fundamental physics [2], new applications such as chronometric leveling [3– 5], and are the prerequisite for a future redefinition of the Systeme International unit of time [6]. In order to validate their performance, repeated comparisons between independent systems, operated by separate laboratories and employing different species, are necessary at this level [7–9]. To this day, frequency ratios of different optical transitions have been measured with uncertainties as low as 5.9×10^{-18} [8]. A very promising species for low systematic uncertainties is ¹¹⁵In⁺ [10–14]. In 2012, In⁺ was proposed as a favorable candidate for a multi-ion clock [15], addressing a fundamental problem in trapped-ion optical clocks: As their systematic uncertainties are reduced, measurements are increasingly limited by the statistical uncertainty due to the quantum projection noise (QPN) of a single particle [16,17]. Multi-ion clocks will open up the path for ion clock measurements with 10^{-19}

level overall uncertainties [18] or can relax local oscillator stability requirements [19].

In this Letter, we demonstrate the operation of an $^{115}In^+$ clock which is based on linear Coulomb crystals (CCs) and can be operated with a variable number of clock ions. We first evaluate our new clock setup with a CC containing a single In⁺ ion, obtaining a fractional systematic uncertainty of 2.5×10^{-18} . In comparisons with two other optical clocks, we observe averaging behavior compatible with white frequency noise down to this level. We report measurements of the frequency ratios with respect to ⁸⁷Sr and the electric octupole (E3) transition in 171 Yb⁺, with respective relative uncertainties of 4.2×10^{-17} —more than one order of magnitude lower than previously reported [14]—and 4.4×10^{-18} . In addition, we determine the absolute frequency of the ${}^{1}S_{0} \leftrightarrow {}^{3}P_{0}$ transition in ${}^{115}\text{In}^{+}$ with an uncertainty limited by the realization of the Systeme International (SI) second, which constitutes a further requirement for the optical redefinition of the SI second [6]. Finally, we demonstrate scalability by operation with up to four clock ions.

In our setup, ¹¹⁵In⁺ ions are cotrapped with ¹⁷²Yb⁺ ions, which provide sympathetic cooling on a strong, dipoleallowed transition ($\Gamma \approx 2\pi \times 20$ MHz). Their positions within the crystal determine the cooling rates of its motional degrees of freedom [20,21]. Since background gas collisions can enable swapping of ion positions, we have developed a mechanism to reproducibly restore crystals to a target permutation, which we identify by a

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FIG. 1. Coulomb crystal clock cycle sequence: (a) time sequence, (b) decision chart. The crystal is initialized in the target permutation with conditional reordering, cooling and molecule dissociation steps. Rabi interrogation is carried out after sympathetic cooling on the Yb⁺ ions and followed by state detection via electron shelving on the In⁺ detection transition.

binary string in this Letter $(0 \cong \ln^+, 1 \cong Yb^+)$. For example, with the $1\ln^+-3Yb^+$ composition used below, we operate the clock with the permutation 1011 (or its mirror image), for which the motional modes with predominant \ln^+ ion motion have four times higher sympathetic cooling rates than those of the permutation 0111. This ensures reproducible and low kinetic energies with a fixed duration cooling pulse.

The CC is trapped in a scalable 3D chip ion trap [18]. The secular frequencies for ¹⁷²Yb⁺ center-of-mass (COM) motion are $(\omega_{rad1}, \omega_{rad2}, \omega_{ax})/2\pi \approx (822 \text{ kHz}, 794 \text{ kHz}, 275 \text{ kHz})$. A bichromatic imaging system allows site-resolved fluorescence detection of both species on an EMCCD camera via the ${}^{2}S_{1/2} \leftrightarrow {}^{2}P_{1/2}$ transition at 369.5 nm for the 172 Yb⁺ ions and the $|{}^{1}S_{0}, F = 9/2, m_{F} = \pm 9/2\rangle \leftrightarrow |{}^{3}P_{1}, F = 11/2, m_{F} = \pm 11/2\rangle$ transitions at 230.6 nm for the 115 In⁺ ions [22]. The light used to interrogate the clock transition is derived from a high-finesse-cavity-stabilized 946 nm Nd:YAG laser [23] which is transfer-locked to an ultrastable cryogenic silicon resonator [24]. Further details of the experimental setup can be found in Ref. [25].

The clock interrogation loop used to stabilize the laser frequency to the atomic reference line is shown in Fig. 1. Each iteration consists of four stages: preparation, cooling, interrogation, and detection. In the preparation stage, the system determines the position of the ytterbium ions in the crystal via 369.5 nm fluorescence, reorders the ions if necessary, and optically pumps the indium ions into either of the $|{}^{1}S_{0}, m_{F} = \pm 9/2\rangle$ states. A 230.6 nm fluorescence measurement verifies the initialization of all clock ions. If too few ions of either species are detected, additional cooling laser frequency sweeps to recrystallize and illumination with about 10 μ W at 230.6 nm focused to 90 μ m to dissociate molecular ions—likely YbOH⁺ formed in background gas collisions-are applied ["rescue" branch in Fig. 1(b)]. After successful preparation, the crystal is Doppler cooled via the Yb⁺ ions. A rectangular pulse of 150 ms duration probes the clock transition. Finally, a fluorescence measurement determines the indium ions' states. A measurement is discarded if the crystal is found in a different permutation in the next iteration. The entire clock sequence is thus ready for operation with a linear CC of multiple clock and cooling ions. We demonstrate this concept by using a $1 \text{In}^+ - 3 \text{Yb}^+$ CC for the measurements presented in the following sections.

We apply a bias magnetic field of about 106 µT, which is more than an order of magnitude higher than typical magnetic field fluctuations in our setup, along the axial trap direction and interrogate the two Zeeman transitions $|{}^{1}S_{0}, m_{F} = \pm 9/2\rangle \Leftrightarrow |{}^{3}P_{0}, m_{F} = \pm 9/2\rangle$ (split by approximately 4280 Hz) using π -polarized light at 236.5 nm. Averaging the two components provides a first-order Zeeman shift insensitive frequency measurement and *in situ* magnetic field determination. We observe a Fourier limited linewidth and a contrast of about 60%, which is consistent with the excited state lifetime of 195(8) ms [11] and residual thermal motion after sympathetic cooling.

The systematic shifts of the clock operating with a $1 \text{In}^+-3 \text{Yb}^+$ CC in permutation 1011 and their uncertainties are summarized in Table I. The overall fractional systematic uncertainty is evaluated to be $u_B^{\text{In}^+} = 2.5 \times 10^{-18}$. In the following, the individual contributions are discussed in order of relevance. Additional details for all contributions can be found in Supplemental Material [26].

Thermal time dilation (TD) results from the residual motion of the clock ion after sympathetic cooling and the corresponding intrinsic micromotion [18]. Because of the low heating rates in our trap (< 1 s⁻¹ for the radial Yb⁺ COM modes) [25], we assume constant crystal temperatures throughout the clock interrogation. The radial temperatures are determined from thermal dephasing of Rabi oscillations and used to calibrate our sympathetic cooling model, from which we derive the axial temperatures. This yields kinetic energy bounds of $k_B \times (T_{\text{rad1}} + T_{\text{rad2}}) = k_B \times 2.3(14)$ mK for the radial modes with the lowest cooling rates and $k_B \times (T_{\text{rad1}} + T_{\text{rad2}}) = k_B \times 0.9(1)$ mK each for all other radial

TABLE I. Fractional frequency shifts and associated uncertainties for the ¹¹⁵In⁺ Coulomb crystal clock operated in the 1In⁺-3Yb⁺ composition. Further contributions with uncertainties below 2×10^{-19} are omitted [26].

Effect	Shift (10 ⁻¹⁸)	Uncertainty (10 ⁻¹⁸)
Thermal time dilation	-2.7	1.6
Blackbody radiation	-13.4	1.4
Quadratic Zeeman	-35.9	1.1
Servo error	-2.6	0.5
AOM chirp	-0.5	0.5
Background gas collisions	0	0.4
Time dilation (EMM)	-0.8	0.1
Electric quadrupole	-0.14	0.03
Total	-56.0	2.5

modes. Together with the axial temperatures, which are estimated as $T_{\rm ax} = 1.7(10)$ mK for all modes, we obtain a total shift of $\Delta \nu_{\rm TD}/\nu_0 = -2.7(16) \times 10^{-18}$. The temperature uncertainty is currently limited by the uncertainty of the angle between the clock laser *k* vector and radial mode principal axes $\theta_1 = 40(13)^\circ = 90^\circ - \theta_2$. This angle will be determined more accurately in future sideband spectroscopy measurements. Directly cooling the clock ions on the ${}^1S_0 \leftrightarrow {}^3P_1$ intercombination line ($\Gamma = 2\pi \times 360$ kHz) will yield overall thermal TD shifts $< 2 \times 10^{-19}$ [37,38].

The blackbody radiation (BBR) shift is calculated based on the static differential polarizability $\Delta \alpha_{\text{stat}} =$ $3.3(3) \times 10^{-41} \text{ J/(V/m)}^2$ [39] of the clock transition. At room temperature, the shift uncertainty is dominated by the uncertainty of this value. We therefore assume the temperature uncertainty of the ion's environment to be half of the entire span of temperatures observed during the clock uptime from sensors on the trap [40] and chamber, with T = 299(1) K, as this translates to a negligible 2×10^{-19} uncertainty contribution to the total shift of $\Delta \nu_{\text{BBR}}/\nu_0 =$ $-13.4(14) \times 10^{-18}$. The temperature contribution could be further reduced with a time-resolved instead of constant correction.

The quadratic Zeeman shift is calculated as $\Delta \nu = \beta \langle B^2 \rangle$ with the coefficient $\beta = -4.05$ Hz/mT² [15,41]. The timeresolved magnetic field data, determined from the observed Zeeman splitting, are used to postcorrect the measured frequencies. The uncertainty in this magnetic field data is dominated by the knowledge of the excited state g factor $g({}^{3}P_{0}) = -9.87(5) \times 10^{-4}$ [11]. The average shift is determined to be $\Delta \nu_{Z2}/\nu_{0} = -35.9(11) \times 10^{-18}$. The uncertainty can be reduced with a more precise measurement of $g({}^{3}P_{0})$. For the ac contribution due to trap rf currents, simulations indicate $B_{\rm rms}^2 = 1.3 \times 10^{-12}$ T² [42], which is comparable to experimental observations in other ion traps [43,44]. Even the highest reported value to date of $B_{\rm rms}^2 =$ 2.17×10^{-11} T² [45] amounts only to a fractional frequency shift of -7×10^{-20} in ¹¹⁵In⁺.

The transfer lock reduces the linear drift rate of the clock laser to that of the Si cavity of ca. $-120 \ \mu\text{Hz} \,\text{s}^{-1}$. Because of an imperfect compensation algorithm, this residual drift resulted in a servo offset of $\Delta \nu_{\text{servo}}/\nu_0 = -2.6(5) \times 10^{-18}$, as determined in postprocessing using the logged error signal [26].

Thermal effects in the clock laser acousto-optic modulator (AOM) can cause cycle-synchronous phase chirps which result in a systematic frequency offset. From interferometric phase measurements [46] at different drive powers, we infer a fractional shift of $-5(5) \times 10^{-19}$.

Site-resolved detection in mixed-species operation allows us to detect all background gas collisions which result in CC permutation changes. In the $1In^+-3Yb^+$ composition, this corresponds to 75% of all events. Collisions below the ion-swapping energy barrier constitute a negligible fraction of events [26]. Thus, from a measured collision rate per ion of $\Gamma_{\text{meas}}^{\text{ion}} = 0.0029(3) \text{ s}^{-1}$ [25], we can estimate the rate of undetected collisions for the four-ion crystal as $\Gamma_{\text{undet}}^{\text{CC}} = 4 \times 0.25 \times \Gamma_{\text{meas}}^{\text{ion}} =$ $0.0029(3) \text{ s}^{-1}$. The treatment of Ref. [47] for the collision-induced phase shifts yields a fractional frequency shift of $\Delta \nu_{\text{coll}}/\nu_0 = 0(4) \times 10^{-19}$. TD shifts due to transferred kinetic energy which would be relevant at this level are suppressed by their detrimental effect on state detection [26,48].

Excess micromotion (EMM) leads to additional TD and Stark shifts [49], the inhomogeneity of which is below 1×10^{-19} across the crystal [25,50]. We compensate radial stray fields via photon-correlation measurements using the Yb⁺ ions [51] at least once every eight hours. The timeaveraged residual shift is calculated in postprocessing by linearly interpolating electric stray fields between 0 V/m and the value observed in the subsequent compensation measurement. Axial EMM contributes $\Delta \nu_{\rm EMM}^{(\rm ax)}/\nu_0 = -8(1) \times 10^{-19}$ and determines the overall shift.

The electric quadrupole shift is calculated via the Hamiltonian $H_{E2} = \nabla E^{(2)} \Theta^{(2)}$ [52] with the *E*-field gradient $\nabla E^{(2)}$ accounting for the trap electric field and Coulomb interaction within the ion crystal [18] and the electric quadrupole moment $\Theta = -1.6(3) \times 10^{-5} ea_B^2$ [53], yielding $\Delta \nu_{E2}/\nu_0 = -1.4(3) \times 10^{-19}$.

Further shifts, such as the clock laser ac Stark and firstorder Doppler shifts have been estimated to contribute less than 2×10^{-19} to the overall uncertainty [26].

We have compared the ¹¹⁵In⁺ clock to an ¹⁷¹Yb⁺ (*E*3) single-ion clock [54] and ⁸⁷Sr lattice clock [56] via an optical frequency comb. In both cases, the measurement instability reaches a few parts in 10¹⁸ for total measurement times of about one week (see Fig. 2). The instability of $1.6 \times 10^{-15} \sqrt{\tau/1 \text{ s}}$ of the In⁺/Sr comparison reflects that



FIG. 2. Instabilities (ADEV) of the ¹¹⁵In⁺/⁸⁷Sr (blue circles) and ¹¹⁵In⁺/¹⁷¹Yb⁺ (*E*3) (red triangles) frequency ratio measurements. The fitted instabilities are $\sigma_y^{\rm fit,In^+/Sr} = 1.6 \times 10^{-15}/\sqrt{\tau/1 \text{ s}}$ (solid blue line) and $\sigma_y^{\rm fit,In^+/Yb^+} = 1.9 \times 10^{-15}/\sqrt{\tau/1 \text{ s}}$ (dashed red line). The shaded areas represent 95% confidence intervals for white frequency noise datasets of the respective durations, as determined in Monte Carlo simulations.

TABLE II. Frequency ratios with combined statistical (u_A) , systematic $(u_{B1,2})$, where 1 refers to the In⁺ clock), and total (u_C) uncertainties (in units of 10^{-18}), and inferred absolute frequencies for the ${}^{1}S_{0} \leftrightarrow {}^{3}P_{0}$ transition in ${}^{115}\text{In}^{+}$. The absolute frequencies inferred from the optical ratios use results reported in Ref. [57] (Sr) and Ref. [58] (Yb⁺). All values are corrected for the relativistic redshifts (RRSs) due to clock height differences [26].

Clocks	u_A	u_{B1}	u_{B2}	<i>u</i> _{RRS}	u_C	Ratio	Absolute frequency (Hz)
$\frac{115}{115}$ In ⁺ / ¹⁷¹ Yb ⁺ (E3)	2.4	2.5	2.7	0.5	4.4	1.973 773 591 557 215 789(9)	1 267 402 452 901 038.87(16)
$^{115}\text{In}^{+}/^{87}\text{Sr}$	2.4	2.5	42	0.5	42	2.952 748 749 874 860 78(13)	1 267 402 452 901 038.96(21)
$^{115}\text{In}^{+}/^{133}\text{Cs}$	170 ^a	15 ^b	170	0.7	240		1 267 402 452 901 039.05(30)
CCTF2021 [59]					4300		1 267 402 452 901 041.3(54)

^aThe combined averaging time of 6.2×10^5 s is extended to 1.0×10^6 s by using a hydrogen maser as a flywheel for gaps in the combined In⁺/Cs uptime (cf. [57,58]). u_A contains an extrapolation uncertainty contribution of $u_{\text{ext}} = 9 \times 10^{-17}$. ^bAdditional intervals with u_{B,In^+} up to 1.5×10^{-17} due to increased EMM are included in the In⁺/Cs comparison.

of the In⁺ CC clock, since the contribution from the Sr clock is only about $2 \times 10^{-16} / \sqrt{\tau/1 \text{ s}}$ [57]. This is consistent with the instability of the In⁺/Yb⁺ comparison, given the 171 Yb⁺ (E3) clock instability of $1.0 \times$ $10^{-15}/\sqrt{\tau/1 \text{ s}}$ [55].

For both datasets, we observe $\chi^2_{\rm red} \approx 1.3$ [26] and averaging behaviors compatible with white frequency noise, which correspond to a statistical uncertainty of $u_A = 2.4 \times 10^{-18}$ (in both cases) at the respective data-set duration of 4.6×10^5 s (In⁺/Sr) and 6.0×10^5 s (In^+/Yb^+) . After correcting for systematic frequency shifts and relativistic shifts from the height difference between the clocks [26], we find an optical frequency ratio of $\nu_0^{\text{In}^+}/\nu_0^{\text{Yb}^+} = 1.973\,773\,591\,557\,215\,789(9).$ We find a frequency ratio

 $\nu_0^{{\rm In}^+}/\nu_0^{{
m Sr}} =$ 2.952 748 749 874 860 78(13), where we include a provisional frequency correction estimation which accounts for long-term variations of the Sr clock frequency and correspondingly increases the uncertainty. These variations became apparent in a repeated comparison of the three clocks in 2024, which shows deviations in $\nu_0^{\text{In}^+}/\nu_0^{\text{Sr}}$ and $\nu_0^{\text{Yb}^+}/\nu_0^{\text{Sr}}$ at the mid-10⁻¹⁷ level and reproduces the above value for $\nu_0^{\text{In}^+}/\nu_0^{\text{Yb}^+}$ within a combined relative uncertainty of 9.7×10^{-18} . Details on the repeated measurement and frequency correction are given in Supplemental Material [26]. We include the value $\nu_0^{\text{In}^+}/\nu_0^{\text{Sr}}$ for comparison with the previously most accurate measurement of the In⁺ frequency via this ratio [14], with which we find agreement within 1.1σ . Table II summarizes the measured frequency ratios together with the individual uncertainty contributions.

The absolute frequency of the unperturbed ${}^{1}S_{0} \leftrightarrow {}^{3}P_{0}$ transition is determined by comparison to Physikalisch-Technische Bundesanstalt's primary Cs fountain clock CSF2 [60] via a hydrogen maser, analogous to Refs. [57,58]. The optical frequency ratio measurements of Table II provide an indirect method to determine the absolute frequency with reduced statistical uncertainty via previous absolute frequency measurements of 171 Yb⁺ (E3) [58] and ⁸⁷Sr [57]. The former yields a frequency uncertainty of $u_C^{\ln^+//Cs} = 1.3 \times 10^{-16}$, limited by the determination of the Yb⁺ absolute frequency (see Table II). However, it is important to note that these results should not be considered as independent (see Ref. [26] for information regarding correlations). The inferred absolute frequencies are shown in Fig. 3. They agree well with the recommended frequency value [59] as well as previously reported values [12–14].

The CC clock is implemented with the intent of scaling up the number of clock ions. We demonstrate this capability in instability measurements with up to four clock ions against the 171 Yb⁺ (E3) clock, as shown in Fig. 4. The chosen compositions, 2In+-4Yb+ and 4In+-8Yb+, are operated in the respective permutations 101011 and 110101010111. We observe reduced instabilities with increasing clock ion number down to $9.2(4) \times 10^{-16}$ at 1 s for four clock ions. However, state preparation currently relies on the spontaneous decay of the ${}^{3}P_{0}$ level with a



FIG. 3. Measurement history for the unperturbed frequency of the ${}^{1}S_{0} \leftrightarrow {}^{3}P_{0}$ transition in ${}^{115}\text{In}^{+}$ [12–14,61]. The blue line and shaded area correspond to the Consultative Committee for Time and Frequency 2021 recommendation of $f_0 =$ 1 267 402 452 901 041.3(54) Hz [59].



FIG. 4. Multi-ion clock frequency instabilities at 1 s with up to 4 In⁺ clock ions. Red triangles depict the combined instability of the In⁺/Yb⁺ ratio. Pink circles show the instability of the In⁺ clock as extracted from the ratios In⁺/Sr (filled) and In⁺/Yb⁺ (open). The solid green line shows the expected QPN-limited scaling with clock ion number, taking into account the additional dead time for state preparation. The addition of a repumper laser for depletion of the ³P₀ level would lead to instabilities indicated by the solid black line.

lifetime of 195(8) ms [11] after successful excitation attempts. The associated dead time increases with ion number and reduces the instability advantage of the increased signal. This will be mitigated in the future by a quench laser at 481.6 nm which returns the ${}^{3}P_{0}$ population to the ground state via ${}^{1}P_{1}$.

In summary, we have demonstrated a Coulomb crystal clock based on $^{115}In^{+}-^{172}Yb^{+}$ ion chains with a systematic uncertainty of 2.5×10^{-18} and an instability of $1.6 \times$ $10^{-15}/\sqrt{\tau/1 \text{ s}}$ for operation with a $1 \text{In}^+-3 \text{Yb}^+$ CC. We have measured the absolute frequency of the ${}^{1}S_{0} \leftrightarrow {}^{3}P_{0}$ transition in $^{115}\mathrm{In^{+}}$ as well as the optical frequency ratios $^{115}\text{In}^+/^{87}\text{Sr}$ and $^{115}\text{In}^+/^{171}\text{Yb}^+$ (E3) with relative uncertainties of 4.2×10^{-17} and 4.4×10^{-18} . To our knowledge, the latter is the frequency ratio measurement with the lowest uncertainty reported to date. Furthermore, we have shown a reduction of the statistical uncertainty to $9.2 \times$ $10^{-16}/\sqrt{\tau/1 \text{ s}}$ in multi-clock-ion operation. This constitutes a significant step towards multi-ion clock operation with low $10^{-16}/\sqrt{\tau/1 \text{ s}}$ instabilities and 10^{-19} level inaccuracies [18] benefiting future fundamental physics tests and relativistic geodesy.

The presented data will provide information for upcoming adjustments of the Consultative Committee for Time and Frequency recommended frequency value for the ¹¹⁵In⁺ clock transition and contribute to ongoing consistency checks of optical frequency measurements towards a redefinition of the SI unit of time.

Since the major contributions to the uncertainty budget are limited by either atomic constants or our limited knowledge of the radial principal axis orientation θ_1 , we expect a current reproducibility of the In⁺ CC clock of about 6×10^{-19} .

Acknowledgments-We thank Uwe Sterr, Thomas Legero, and Jialiang Yu for providing the ultrastable 1542 nm laser, Stepan Ignatovich and Maksim Okhapkin for their work on the clock laser, Heiner Denker and Ludger Timmen for leveling measurements of our clock systems, Fabian Wolf for support with UV fiber production, and Rattakorn Kaewuam, Piyaphat Phoonthong, and Michael Kazda for their help in the determination of the AOM phase chirp. We acknowledge support by the projects 18SIB05 ROCIT and 20FUN01 TSCAC. These projects have received funding from the EMPIR programme co-financed by the Participating States and from the European Union's Horizon 2020 research and innovation programme. We acknowledge funding by the Deutsche Forschungsgemeinschaft (DFG) under Germany's Excellence Strategy-EXC-2123 OuantumFrontiers-390837967 (RU B06) and through Grant No. CRC 1227 (DQ-mat, projects B02 and B03). This work has been supported by the Max-Planck-RIKEN-PTB-Center for Time, Constants and Fundamental Symmetries.

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