

Nuclear Spin Quenching of the $^2S_{1/2} \rightarrow ^2F_{7/2}$ Electric Octupole Transition in $^{173}\text{Yb}^+$

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We report the coherent excitation of the highly forbidden $^2S_{1/2} \rightarrow ^2F_{7/2}$ clock transition in the odd isotope $^{173}\text{Yb}^+$ with nuclear spin $I = 5/2$, and reveal the hyperfine-state-dependent, nuclear-spin-induced quenching of this transition. The inferred lifetime of the $F_e = 4$ hyperfine state is one order of magnitude shorter than the unperturbed $^2F_{7/2}$ clock state of $^{171}\text{Yb}^+$. This reduced lifetime lowers the required optical power for coherent excitation of the clock transition, thereby reducing the ac Stark shift caused by the clock laser. Using a three-ion Coulomb crystal, we experimentally demonstrate an approximately twentyfold suppression of the ac Stark shift, a critical improvement for the scalability of future multi-ion Yb^+ clocks. Furthermore, we report the $|^2S_{1/2}, F_g = 3\rangle \rightarrow |^2F_{7/2}, F_e = 6\rangle$ unquenched reference transition frequency as 642.11917656354(43) THz, along with the measured hyperfine splitting and calculated quadratic Zeeman sensitivities of the $^2F_{7/2}$ clock state. Our results pave the way toward multi-ion optical clocks and quantum computers based on $^{173}\text{Yb}^+$.

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Trapped ion systems have widely been used to advance fundamental physics and develop high-precision optical clocks [1–21]. In particular, the forbidden electric octupole (E3) transition in the Yb^+ ion, with a measured lifetime of 1.6(1) yr [20,22], offers both exquisite clock accuracy [1–6] and high sensitivity to new physics beyond the standard model, such as temporal variation of the fine-structure constant [21,23,24], violations of local Lorentz invariance [15,16,25,26], and searches for fifth forces [17–19,27]. Precision spectroscopy on Yb^+ isotopes also opens a new window to nuclear physics [28,29] and the study of deformations of neutron-rich nuclei [17,18,30].

The sensitivity of single-ion precision spectroscopy is fundamentally limited by the quantum projection noise, which scales inversely with $\sqrt{N} \cdot \tau$, where N and τ are the ion number and the measurement time, respectively. Thus, increasing the number of ions in clocks and precision spectroscopy can extend the reach of searches for new physics. In the case of the Yb^+ E3 transition, however, scalability and multi-ion operation face challenges due to the strong ac Stark shift caused by the clock laser (hereafter

referred to as the “ac Stark shift”). While advanced Ramsey techniques can compensate for the ac Stark shift during clock interrogation, achieving sufficient excitation rates across multiple ions remains challenging and requires laser beam profile inhomogeneities of less than 2% [31].

In 2016, Dzuba and Flambaum predicted a strong hyperfine-induced electric dipole (HFE1) contribution in $^{173}\text{Yb}^+$ [32] due to the large electric quadrupole moment of its deformed nucleus [18]. Among stable Yb isotopes, this unique property of $^{173}\text{Yb}^+$ could effectively enhance the extremely weak $^2S_{1/2} \rightarrow ^2F_{7/2}$ transition, which otherwise is a pure E3 transition, thereby reducing the required laser power, and thus the ac Stark shift. The ^{173}Yb isotope, which has also recently been proposed for studying the nuclear magnetic quadrupole moment and parity violation [33], is still largely unexplored. In addition, precise measurement of the $^{173}\text{Yb}^+$ hyperfine (HF) structure is expected to resolve higher-order nuclear moments [34,35], offering insights into the nuclear structure of heavy atoms. Such investigations can help to refine theoretical models and address the discrepancy between two reported values of the magnetic octupole moment [36,37]. Overall, the reduced ac Stark shift and the narrow linewidth make $^{173}\text{Yb}^+$ an intriguing candidate for multi-ion clock spectroscopy and nuclear structure studies [30].

Beyond its role in clock spectroscopy, $^{173}\text{Yb}^+$ is promising for scalable quantum computing architectures. In addition, Yb^+ has been proposed for a quantum computing protocol interrogating multiple qubit types within a single

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atomic species [38,39]. The long-lived $^2F_{7/2}$ state, with its rich HF structure, also supports polyqubit construction [40], the implementation of advanced quantum error correction techniques, and offers a new shelving transition for high detection fidelity [41,42].

In this Letter, we determine the absolute frequency of the $4f^{14}6s|^2S_{1/2}, F_g = 3\rangle \rightarrow 4f^{13}6s|^2F_{7/2}, F_e = 6\rangle$ clock transition, the HF splittings and the quadratic Zeeman sensitivities of the $^2F_{7/2}$ clock states, and quantify the enhancement of decay rates in the $|^2F_{7/2}, F_e = 2, 4\rangle$ hyperfine states due to nuclear spin quenching.

In our setup, we initially confine a single Yb^+ ion in a radio frequency (rf) Paul trap [43] with motional trapping frequencies of $\omega_x \approx 2\pi \times 650$ kHz, $\omega_y \approx 2\pi \times 612$ kHz, and $\omega_z \approx 2\pi \times 212$ kHz. Three pairs of coils on the vacuum chamber are used to compensate for the background magnetic field and to generate a defined quantization field of $B = 0.2$ mT. Doppler cooling to temperatures below 1 mK and state detection are accomplished by 370 nm laser beams assisted by repumper lasers at 935 and 760 nm (Fig. 1). To address the HF states, we use a 10.49 GHz resonant electro-optic modulator at 370 nm and a broadband electro-optic modulator at 935 nm. As we resonantly excite the $|^2P_{1/2}, F = 3\rangle$ state, and the transition from $|^2S_{1/2}, F_g = 3, m = 0\rangle$ to $|^2P_{1/2}, F = 3, m = 0\rangle$ is forbidden according to angular momentum selection rules, a π -polarized 370 nm beam is used to efficiently pump the atomic population to the $|^2S_{1/2}, F_g = 3, m = 0\rangle$ state. Optionally, the atomic population can be transferred to $|^2S_{1/2}, F_g = 2, m = 0\rangle$ by a successive rf pulse at 10.49 GHz, resonant with the $^2S_{1/2}$ state HF splitting. State preparation

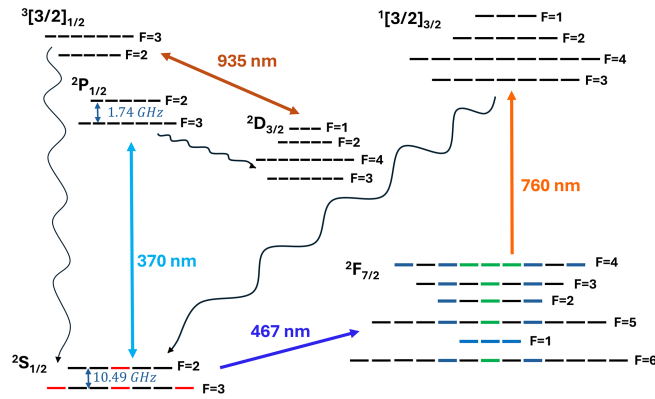


FIG. 1. Level scheme of the $^{173}\text{Yb}^+$ ion. The 370 nm dipole transition is used for Doppler cooling and state detection. The 935 nm laser prevents population trapping in the $^2D_{3/2}$ state. A 467 nm laser excites the highly forbidden electric octupole transition. Laser light at 760 nm repumps the electron from the $^2F_{7/2}$ state. For spectroscopic measurements, the electron population can be prepared in the $|F_g = 3, m_g = 0, \pm 3\rangle$ or $|F_g = 2, m_g = 0\rangle$ (red) of the $^2S_{1/2}$ state. The interrogated $^2F_{7/2}$ HF states are marked in green (coherent excitation) or blue (rapid adiabatic passage).

into the $|^2S_{1/2}, F_g = 3, m = \pm 3\rangle$ stretched states can be achieved by utilizing σ -polarized 370 nm light. The highly forbidden clock transitions are probed with an ultrastable laser at 467 nm, which obtains its stability from a cryogenic silicon cavity [44]. The clock laser, with 6 mW power, is focused to a 30 μm waist. It propagates orthogonally to the plane defined by the trap and quantization axes, with linear polarization parallel to the quantization axis. Laser light at 760 nm repumps the electron back to the ground state after excitation. Further details of the experimental setup can be found in the End Matter (EM).

To search for the $^2S_{1/2} \rightarrow ^2F_{7/2}$ clock transitions in $^{173}\text{Yb}^+$, we predict its unperturbed frequency (without hyperfine interaction and systematic shifts) using a King plot [18,45,46], which combines the transition frequencies of the five even isotopes [18] with the measured $^2S_{1/2} \rightarrow ^2D_{5/2}$ quadrupole transitions for $^{173}\text{Yb}^+$, yielding $\nu_{173} = 642.118\,148(243)$ THz. To determine the HF structure, we extrapolate the A coefficient of $^{173}\text{Yb}^+$ to $A_{173} = -249.3(3)$ MHz from the known A coefficient of $^{171}\text{Yb}^+$ using the isotopes' nuclear dipole moments $\mu_{171} = 0.4923(4)\mu_N$ and $\mu_{173} = -0.6780(6)\mu_N$ [47], while the B coefficient is taken from theoretical predictions, $B = -4700 \pm 500$ MHz [30,32].

To increase the scanning speed, we use the rapid adiabatic passage (RAP) technique [48]. A RAP sweep across resonance enables robust state excitation even with fast decoherence, as detailed in Supplementary Material (SM) [49]. With this method, we first identified 15 previously unobserved transition frequencies with a few kHz accuracy. The F and m quantum numbers of the HF states are inferred from their Zeeman sensitivities.

To determine the absolute frequencies of the clock transitions at the kHz level, we use the E3 transition in $^{172}\text{Yb}^+$ as a reference (the level scheme can be found in Fig. 4 in SM [49]), whose absolute frequency is known with an uncertainty of 11 Hz, with $\nu_{172} = 642\,116\,785\,150\,892.3(11.1)$ Hz [48]. In this Letter, we measured a magnetic field drift of 0.07 μT over one day, corresponding to a measurement uncertainty of $\sigma_{\text{total}}^{(172)} = 432$ Hz. This represents the main contribution to the systematic uncertainty budget, shown in SM [49].

For the absolute frequency measurements, we prepare the atomic population in the $|^2S_{1/2}, F_g = 3, m_g = 0\rangle$ ground state and coherently excite transitions to the $|^2F_{7/2}, F_e = 2, 4, 6, m_e = 0\rangle$ states. Details of the experimental sequence are provided in SM [49]. A summary of the measured transition frequencies and hyperfine splittings is provided in Table I, and the leading systematic shifts of the absolute frequencies are listed in SM [49]. To determine additional HF splittings and reduce systematic uncertainty, we also prepare the population in the $|^2S_{1/2}, F_g = 2, m_g = 0\rangle$ or $|^2S_{1/2}, F_g = 3, m_g = \pm 3\rangle$ states, enabling interrogation of further transitions. A detailed overview of the measurement

TABLE I. Measured HF splittings of $^2F_{7/2}$ state referenced to its $F_e = 6$ substate ($W_{F_e} - W_6$). The $F_e = 2, 3, 5, 6$ states are interrogated by coherent excitation of the first-order magnetic insensitive $\Delta m_F = 0$ transitions. By exciting the magnetic sensitive transitions using the RAP technique, the uncertainties due to quadratic Zeeman shift of the $F_e = 1, 4$ states are reduced. In addition, we report the absolute $^{173}\text{Yb}^+$ transition frequencies $\nu^{(F_e)}$ from the $|^2S_{1/2}, F_g = 3\rangle$ ground state to the $|^2F_{7/2}, F_e = 2, 6\rangle$ excited states from an independent measurement compared to the measured $^{172}\text{Yb}^+$ clock transition. The statistical (u_A) and systematic (u_B) uncertainties are shown in brackets, respectively.

F_e	$W_{F_e} - W_6$ (Hz)	$\nu^{(F_e)} - 642.1$ THz (Hz)
1	3 659 258 477 (71) (503)	
2	4 487 804 282 (2) (67)	23 664 367 825 (16) (435)
3	5 232 926 857 (4) (5290)	
4	5 297 389 527 (4) (5430)	
5	3 884 874 642 (2) (175)	
6	0	19 176 563 542 (16) (432)

results can be found in SM [49]. The derivation of the HF coefficients from the measured HF splittings is beyond the scope of this Letter and will be discussed in a separate paper [54], which involves novel atomic structure theory.

Because of the large nuclear electric quadrupole moment of ^{173}Yb , the HF states of the $^2F_{7/2}$ clock state and $^2S_{1/2}$ ground state mix with intermediate states with the same hyperfine F number, which creates additional electric dipole-allowed decay (HFE1, details in SM [49]) channels to the ground state. In particular, the mixing between the $^2F_{7/2}$ and $^2P_{3/2}$ states is expected to contribute most significantly to the HFE1 channel due to their small energy separation. The decay rate of an excited HF state, $\mathcal{R}^{(F_e)}$, given by the sum of the E3 and HFE1 transition rates $[\mathcal{R}_{\text{E3}}^{(F_e \rightarrow F_g)} \text{ and } \mathcal{R}_{\text{HFE1}}^{(F_e \rightarrow F_g)}]$ as detailed in EM and SM [49], can thus be significantly enhanced. To quantify this enhancement, we measure the laser-induced oscillation frequencies [Rabi frequencies $\Omega^{(F_g \rightarrow F_e)}$] between the ground and excited states for several hyperfine levels.

In a trapped-ion system, accurately determining the Rabi frequency from experimental data requires accounting for decoherence effects such as thermal dephasing, magnetic field fluctuations, and laser phase noise. As detailed in SM [49], thermal dephasing is modeled using the Jaynes-Cummings model to describe the excitation probability. It arises from the thermal distribution over motional quantum states, characterized by the mean phonon number $\bar{n}$, which leads to different Rabi frequencies and results in damping of the observed Rabi oscillations. Magnetic fluctuations and laser phase noise decoherence are modeled with an added exponential decay term involving the decoherence time t_{decoh} . The Rabi frequency is then extracted by fitting the experimental data using this combined model, as described in SM [49].

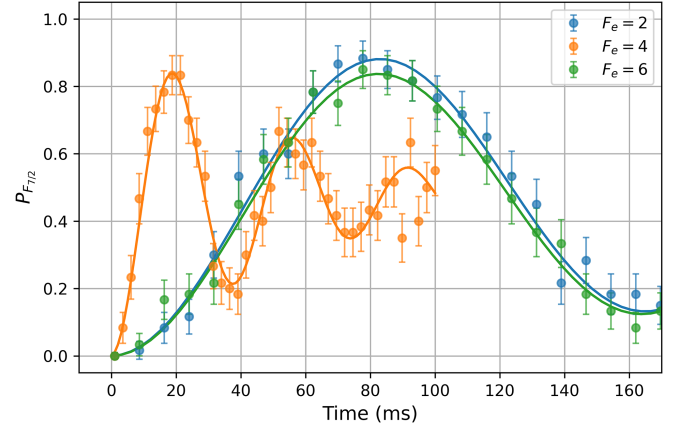


FIG. 2. Experimentally measured Rabi oscillations for clock transitions from $|^2S_{1/2}, F_g = 3, m_g = 0\rangle$ ground state to $^2F_{7/2}$ $F_e = 2$ (blue), $F_e = 4$ (orange), and $F_e = 6$ (green) HF states. The experimental data (circle) is fitted with the Jaynes-Cummings model (solid line) with $\bar{n} = 17.6(1.4)$, leading to $\Omega^{(3 \rightarrow 2)} = 2\pi \times 6.74(5)$ Hz, $\Omega^{(3 \rightarrow 4)} = 2\pi \times 30.12(36)$ Hz, and $\Omega^{(3 \rightarrow 6)} = 2\pi \times 6.78(5)$ Hz. The error bar shows the quantum projection noise. The quenched transition ($F_e = 4$) can be driven at a much higher Ω with the same optical intensity.

The measurement was performed at a well-controlled ion temperature, achieved by carefully adjusting the cooling beam power and by maintaining stable environmental conditions, enabling reliable extraction of both the decoherence time τ_{decoh} and the mean phonon number $\bar{n}$. We coherently drive the first-order magnetically insensitive transitions using a 467 nm clock laser with polarization parallel to the magnetic field (see EM). The $|^2S_{1/2}, F_g = 3\rangle \rightarrow |^2F_{7/2}, F_e\rangle$ transitions were interrogated in the order $F_e = 4 \rightarrow 2 \rightarrow 6 \rightarrow 4$. An ion temperature of $0.56(4)$ mK, close to the Doppler limit and corresponding to a mean phonon number $\bar{n} = 17.6(1.4)$, was confirmed by fitting the Rabi oscillations of the transitions to $F_e = 2$ and $F_e = 6$ using the Jaynes-Cummings model. Representative Rabi oscillations for the interrogated transitions are shown in Fig. 2. From these measurements, we extracted decoherence times of $\tau_{\text{decoh}} = 1777(865)$, $108(22)$, and $2370(1425)$ ms for the transitions to the $F_e = 2, 4, 6$ HF states, respectively. The observed decoherence is most likely caused by residual magnetic field fluctuations and the differing quadratic Zeeman sensitivities of the HF states (see SM [49]). The uncertainty in the absolute laser intensity cancels in the relative measurement; only intensity fluctuations due to beam pointing at the timescale between two measurements contribute to the final uncertainty. The extracted values of $\Omega^{(3 \rightarrow 4)}$ from the first and last measurements, $17.2(2)$ and $17.3(2)$ ms, respectively, indicate that laser intensity variation due to beam pointing contributes less than 0.8% to the overall uncertainty. The validity of the model is proved by a repeated measurement at an increased ion temperature, which shows consistent results, as detailed in SM [49].

TABLE II. Comparison of experimental and theoretical quantities for $F_e = 2, 4$. Measured Rabi frequency ratios $\Omega^{(3 \rightarrow F_e)}/\Omega^{(3 \rightarrow 6)}$ are used to derive effective E1 rates $\mathcal{R}_{\text{E1,eff}}^{(F_e \rightarrow 3)}$ via Eqs. (B2) and (B4), which are compared to E3 rates $\mathcal{R}_{\text{E3}}^{(F_e \rightarrow 3)}$ from the measured lifetime [20,22], and to theoretical HFE1 rates $\mathcal{R}_{\text{HFE1}}^{(F_e \rightarrow 3)}$ from Ref. [32]. Uncertainties include Rabi frequency measurements and the E3 lifetime [20,22]. Rates are in $10^{-8}/\text{s}$.

F_e	$\Omega^{(3 \rightarrow F_e)}/\Omega^{(3 \rightarrow 6)}$	$\mathcal{R}_{\text{E1,eff}}^{(F_e \rightarrow 3)}$	$\mathcal{R}_{\text{E3}}^{(F_e \rightarrow 3)}$	$\mathcal{R}_{\text{HFE1}}^{(F_e \rightarrow 3)}$
2	0.987(35)	1.73(16)	0.29(1)	51
4	4.22(20)	23.7(3.0)	0.96(5)	650

After measuring the Rabi frequencies, we relate them to the corresponding transition rates. The $|^2S_{1/2}, F_g = 3\rangle \rightarrow |^2F_{7/2}, F_e = 6\rangle$ transition is an unquenched pure E3 transition and serves as a reference. Its lifetime is taken as 1.6(1) yr, as previously measured in $^{171}\text{Yb}^+$ [20,22]. From this lifetime, we extract the transition rate $\mathcal{R}_{\text{E3}}^{(F_e \rightarrow 3)}$ for each F_e using angular momentum reduction (see EM). Results are listed in Table II.

If the $F_g = 3 \rightarrow F_e = 2, 4$ transitions had only E3 contributions, these transition rates $\mathcal{R}_{\text{E3}}^{(F_e \rightarrow 3)}$ can be used to calculate their expected Rabi frequencies $\Omega_{\text{E3}}^{(3 \rightarrow F_e)}$ (see EM), yielding predicted ratios of $\Omega_{\text{E3}}^{(3 \rightarrow F_e)}/\Omega^{(3 \rightarrow 6)} = 0.25$ for $F_e = 2$ and 0.51 for $F_e = 4$.

The experimentally measured ratios $\Omega^{(3 \rightarrow F_e)}/\Omega^{(3 \rightarrow 6)}$, listed in Table II, are significantly larger, confirming that the HFE1 decay channel is dominating in these transitions, as discussed in SM [49]. Hence we assume that the E3 contribution is small compared to the HFE1 contribution for $F_e = 2, 4$, and treat these as effectively pure E1 transitions, as detailed in SM [49]. Under this approximation, the effective E1 rates $\mathcal{R}_{\text{E1,eff}}^{(F_e \rightarrow 3)}$ are derived from the measured ratio between Rabi frequencies $\Omega^{(3 \rightarrow F_e)}/\Omega^{(3 \rightarrow 6)}$ by combining Eqs. (B2)–(B5) (see EM). The inferred effective rate for the $F_g = 3 \rightarrow F_e = 2$ transition is approximately 6 times larger than the E3 transition rate, and about 25 times larger for $F_e = 4$, indicating a strong enhancement of the $F_e = 2$ and $F_e = 4$ decay rates due to the HFE1 contribution. As discussed in SM [49], this enhancement leads to the intriguing effect of the excited HF states having individual lifetimes, while identical lifetimes are expected for unperturbed hyperfine states. By neglecting the E3 contribution, the inferred lifetime of the $F_e = 4$ state is about 12 times shorter than the unquenched state. Since $F_e = 4 \rightarrow F_g = 2$ is not quenched, the lifetime of this state is approximately $1/\mathcal{R}_{\text{eff,E1}}^{(4 \rightarrow 3)} = 49(21)$ d, scaled from the 1.6 (1)-yr lifetime of $^{171}\text{Yb}^+$ [20,22]; see Eq. (16) in SM [49].

Additionally, we compare the effective rates with the theoretically estimated HFE1 rates $\mathcal{R}_{\text{HFE1}}^{(F_e \rightarrow 3)}$ from [32], as summarized in Table II. The experimentally inferred

effective transition rates differ from the theoretical values by a factor of about 28. Checks were also performed as summarized in EM. The error introduced by approximating $\mathcal{R}_{\text{E1,eff}}^{(F_e \rightarrow 3)}$ as $\mathcal{R}_{\text{HFE1}}^{(F_e \rightarrow 3)}$ is, in our case and based on Table II, at most 50% (see EM). A potential change in the inferred lifetime may arise from the taken Refs. [20,22], i.e., the experimental lifetime of the $^2F_{7/2}$ clock state of $^{171}\text{Yb}^+$, which is currently under reevaluation [22].

To demonstrate the advantage of the quenched transition ($F_g = 3 \rightarrow F_e = 4$), we trap a three-ion Coulomb crystal and compare spectra to the reference transition ($F_g = 3 \rightarrow F_e = 6$). Both transitions are driven by a Gaussian beam, with identical pulse durations but different probe beam powers chosen to achieve the same Rabi frequency. We observe that for the same Rabi frequency, the quenched transition requires significantly less optical intensity than the reference transition, resulting in smaller ac Stark shifts on the transition frequency. As visualized in Fig. 3(a), multi-ion operation is feasible with the quenched transition, despite the large intensity gradient of the Gaussian beam. In contrast, simultaneous excitation of multiple ions is not possible with the reference transition [Fig. 3(b)] due to the larger differential ac Stark shift. In this measurement, the extracted reduction factor of the ac Stark shift is 22(5), in good agreement with the value extrapolated from the measured Rabi frequency ratio, 18(2).

As a candidate for future optical clocks, the quadratic Zeeman shift may become a limiting factor for accuracy. Therefore, based on the measured hyperfine splittings of the $^2F_{7/2}$ state in Table I, we calculate the second-order

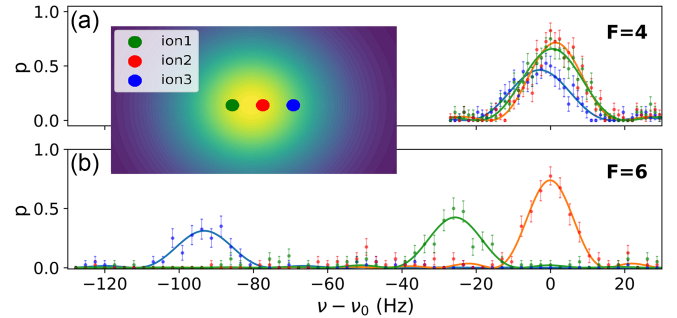


FIG. 3. Experimentally measured clock resonances of a three-ion Coulomb crystal with 10 μm inter-ion spacing. The inset illustrates the laser intensity and ion positions (not to scale). The laser beam radius is $w = 30 \mu\text{m}$, pulse time $\tau = 55$ ms, and $\Omega^{(3 \rightarrow 6)} \approx \Omega^{(3 \rightarrow 4)} = 2\pi \times 9.1(1.2)$ Hz. The relative intensity between the two measurements is $I_{\text{Laser}}^{(3 \rightarrow 6)}/I_{\text{Laser}}^{(3 \rightarrow 4)} = 20.6(1.5)$. Referencing to the transition frequency of ion 2, $\nu_{\text{ion2}}^{(F_g \rightarrow F_e)}$, we observe a differential ac Stark shift of $\nu_{\text{ion1}}^{(3 \rightarrow 4)} = -0.9(4)$ Hz, $\nu_{\text{ion3}}^{(3 \rightarrow 4)} = -4.2(4)$ Hz, $\nu_{\text{ion1}}^{(3 \rightarrow 6)} = -25.8(4)$ Hz, and $\nu_{\text{ion3}}^{(3 \rightarrow 6)} = -93.3(4)$ Hz. Hence for a similarly Fourier-limited linewidth $\Delta\nu \approx 16$ Hz, the transition from $F_g = 3$ to $F_e = 4$ state (a) requires less laser intensity than to the $F_e = 6$ state (b), which results in a smaller ac Stark shift.

Zeeman sensitivities. Among the first-order magnetically insensitive transitions, the $F_g = 3 \rightarrow F_e = 6$ transition exhibits the smallest second-order Zeeman sensitivity of $-33.7 \text{ mHz}/\mu\text{T}^2$. Notably, the small energy gap between the $F_e = 3$ and $F_e = 4$ hyperfine states leads to large second-order Zeeman sensitivities of -7012 and $8060 \text{ mHz}/\mu\text{T}^2$, respectively. These sensitivities, much larger than the value of $-2.03(2) \text{ mHz}/\mu\text{T}^2$ reported for $^{171}\text{Yb}^+$ $F_g = 0 \rightarrow F_e = 3$ transition [55], may limit the interrogation time during clock operation due to magnetic decoherence. Such decoherence can be suppressed by operating the clock at a lower magnetic field while interrogating the transition. The first- and second-order Zeeman sensitivities of all hyperfine and Zeeman substates are summarized in SM [49].

In summary, we measured the hyperfine structure of the $^2F_{7/2}$ state in $^{173}\text{Yb}^+$, from which we calculated its second-order Zeeman sensitivities. The transition frequency of $|^2S_{1/2}, F_g = 3\rangle \rightarrow |^2F_{7/2}, F_e = 6\rangle$ (unquenched) is determined to be $642.119\,176\,563\,54(43) \text{ THz}$. We also calculated the HFE1 transition rates using approximations detailed in SM [49], showing that for the $F_g = 3 \rightarrow F_e = 4$ transition, the inferred HFE1 rate is significantly higher than the E3 rate. This leads to a 12 times shorter lifetime for the $F_e = 4$ hyperfine state, revealing hyperfine-dependent, nuclear-spin-induced quenching of this clock transition. Additionally, we demonstrate simultaneous excitation of multiple ions using a Gaussian beam, highlighting the potential of $^{173}\text{Yb}^+$ for future multi-ion optical clocks.

Compared to the E3 transition of $^{171}\text{Yb}^+$, the $|^2S_{1/2}, F_g = 3\rangle \rightarrow |^2F_{7/2}, F_e = 4\rangle$ transition of $^{173}\text{Yb}^+$ with a lifetime of tens of days, can be driven at a $3.67(17)$ higher Rabi frequency. Therefore, this quenched transition could serve as a interesting shelving transition for future quantum computers. Also, the ac Stark shift at given Rabi frequency is reduced by more than one order of magnitude. This would enable simultaneous interrogation of up to 100 ions using a segmented Paul trap as described in Refs. [43,56] and a flattop beam [31]. The large number of ions would facilitate the implementation of advanced clock protocols, such as zero-dead time clocks [57,58] or cascaded clocks [59,60] for transportable optical clock systems. Such clocks could play a pivotal role in intercontinental clock comparison, contributing to the future redefinition of the Systeme International second [61] and to relativistic geodesy [62–64].

However, the accuracy and the interrogation time of multi-ion optical clocks based on the clock transition $|^2S_{1/2}, F_g = 3\rangle \rightarrow |^2F_{7/2}, F_e = 4\rangle$ are likely to be limited by the strong quadratic Zeeman shift. Employing an HF-averaging technique [65], operation under field-insensitive conditions [66] or cascaded clock protocols combining high stability and high accuracy clock transitions [67] can

improve both the accuracy and the coherence time of such a multi-ion optical clock.

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Data availability—The data that support the findings of this article are openly available [68].

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End Matter

Experimental geometry—The geometry of the experimental setup for laser spectroscopy on trapped $^{173}\text{Yb}^+$ ions is shown in Fig. 4. The quantization axis is defined by the applied magnetic field at an angle of 26° to the z axis. In addition to the laser beams depicted in Fig. 4, 935 and 760 nm repumping lasers are sent along the trap axis (i.e., the z axis). A linearly polarized 370 nm beam, aligned with the magnetic field direction, assists Doppler cooling. A 411 nm laser beam propagating along the x axis can be used for probing the $|^2S_{1/2}\rangle \rightarrow |^2D_{5/2}\rangle$ transition, with its polarization vector $\vec{e}_{411}$ perpendicular to the magnetic field.

Transition rates, decay rates, and Rabi frequency—In both the main text and SM [49], we use the terms “Rabi frequency,” “transition rate,” and “decay rate.” Here, we clarify their distinctions.

The transition rate $\mathcal{R}^{(F_e \rightarrow F_g)}$ between hyperfine states F_e and F_g is defined as the total rate from the excited state to all allowed Zeeman sublevels of F_g (Fig. 5). The Rabi frequency $\Omega^{(F_g \rightarrow F_e)}$ characterizes the coherent excitation strength of a specific driven transition between individual Zeeman sublevels of F_g and F_e . The decay rate $\mathcal{R}^{(F_e)}$ of an excited state F_e is the sum of all allowed transition rates to ground states

$$\mathcal{R}^{(F_e)} = \sum_{F_g} \mathcal{R}^{(F_e \rightarrow F_g)} = \frac{1}{\tau^{(F_e)}}, \quad (\text{B1})$$

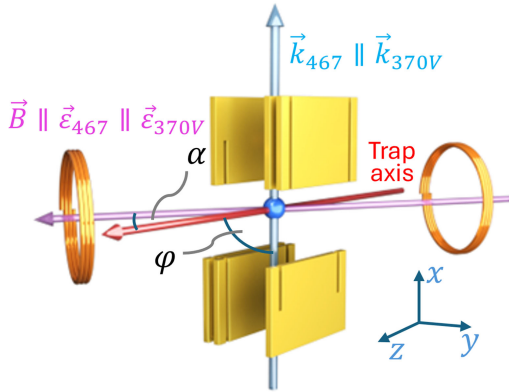


FIG. 4. Experimental setup. A $^{173}\text{Yb}^+$ ion (blue sphere) is trapped in a linear Paul trap. The red arrow indicates the axial direction of the rf trap, which coincides with the z axis. The blue arrow shows the propagation direction of the vertical 370 nm beam (370V) and the 467 nm beam. The polarization of both the 370 V and 467 nm beams is aligned with the magnetic field (pink arrow). The angle between the spectroscopy beam and the trap axis, φ , is 90° , and the angle between the magnetic field and the trap axis, α , is approximately 26° .

and is the inverse of the excited-state lifetime $\tau^{(F_e)}$. The relation between the Rabi frequency and the transition rate is discussed below.

Hyperfine-induced electric dipole (HFE1): We treat the hyperfine-induced electric dipole (HFE1) contribution as an effective E1 process. Since we use π -polarized light, the Rabi frequency can be written as

$$\Omega_{\text{HFE1}}^{(F_g \rightarrow F_e)} = \frac{eE_0}{\hbar} \frac{\langle F_g m_g, 1, 0 | F_e m_e \rangle \langle F_e || \hat{r} || F_g \rangle}{\sqrt{2F_e + 1}}, \quad (\text{B2})$$

where e is the elementary charge, E_0 is the electric field amplitude of the clock laser, $\langle F_g m_g, 1, 0 | F_e m_e \rangle$ and $\langle F_e || \hat{r} || F_g \rangle$ are the Clebsch-Gordan coefficient and the reduced transition matrix element, respectively. The corresponding HFE1 transition rate is [32]

$$\mathcal{R}_{\text{HFE1}}^{(F_e \rightarrow F_g)} = \frac{4}{3} c \alpha k^3 \frac{|\langle F_e || \hat{r} || F_g \rangle|^2}{2F_e + 1}, \quad (\text{B3})$$

where c is the speed of light, α the fine-structure constant, and k is the wave vector of the clock laser. Equations (B2) and (B3) show that the Rabi frequency is proportional to $\sqrt{\mathcal{R}_{\text{HFE1}}^{(F_e \rightarrow F_g)}}$. In the main text, we use measured Rabi frequencies $\Omega^{(3 \rightarrow F_e)}$ for $F_e = 2, 4$ to calculate effective E1 transition rates $\mathcal{R}_{\text{E1,eff}}^{(F_e \rightarrow 3)}$ using Eqs. (B2) and (B3), under the hypothesis that $\mathcal{R}_{\text{HFE1}}^{(F_e \rightarrow F_g)} \approx \mathcal{R}_{\text{E1,eff}}^{(F_e \rightarrow F_g)}$ (see SM [49]).

Electric octupole (E3): For E3 transitions between $m_g = 0$ and $m_e = 0$ Zeeman substates, excited with π -polarized light the Rabi frequency is

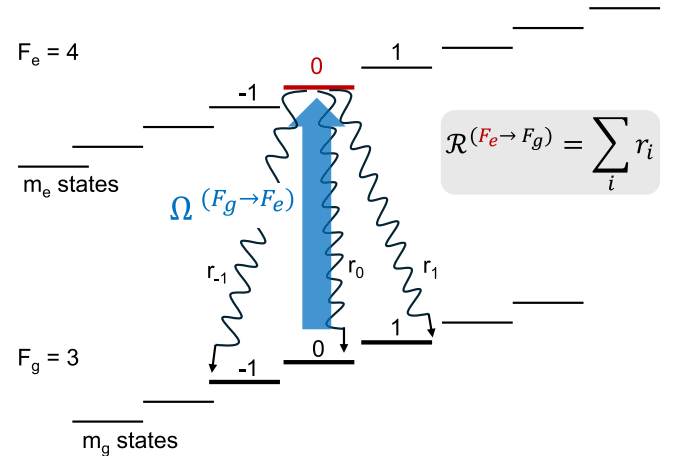


FIG. 5. Illustration of the Rabi frequency and transition rate for the $F_e = 4, m_e = 0 \rightarrow F_g = 3$ case, with an E1 decay channel. Each Zeeman sublevel m_e of the excited state has the same transition rate $\mathcal{R}^{(F_e \rightarrow F_g)}$, obtained by summing over the rates of all allowed decay channels to the ground state (here $r_0 + r_1 + r_{-1}$).

$$\Omega_{E3}^{(F_g \rightarrow F_e)} = \frac{eE_0}{\hbar} \frac{k^2}{30} \frac{\langle F_g, 0, 3, 0 | F_e, 0 \rangle}{\sqrt{2F_e + 1}} \langle F_e \| \hat{O}(r^3) \| F_g \rangle, \quad (\text{B4})$$

with $\hat{O}(r^3)$ the octupole operator. The additional factor k^2 arises from second-order spatial derivative of the laser's electric field. The factor 30 is a geometric factor that depends on $m_e - m_g$, the angles between the polarization vector, the wave vector, and the quantization axis (see Fig. 4 and [69]). The corresponding transition rate is

$$\mathcal{R}_{E3}^{(F_e \rightarrow F_g)} = \frac{8k^4}{4725} c\alpha k^3 \frac{|\langle F_e \| \hat{O}(r^3) \| F_g \rangle|^2}{2F_e + 1}, \quad (\text{B5})$$

where the coefficient $8k^4/4725$ follows from multipole expansion [69], leading to a k^7 dependence combined with the k^3 arising from the state density of the photon modes. In order to relate it to the lifetime, we use angular momentum reduction [70],

$$|\langle F_e \| \hat{O}(r^3) \| F_g \rangle| = \sqrt{(2F_e + 1)(2F_g + 1)} \times \left\{ \begin{matrix} J_e & 3 & J_g \\ F_g & 1 & F_e \end{matrix} \right\} |\langle J_e \| \hat{O}(r^3) \| J_g \rangle|, \quad (\text{B6})$$

and $\left\{ \right\}$ denotes a 6- j Wigner symbol.

Finally, the total E3 decay rate is

$$\mathcal{R}_{E3} = \frac{1}{\tau_{E3}} = \frac{8}{4725} c\alpha k^7 \frac{|\langle J_e \| \hat{O}(r^3) \| J_g \rangle|^2}{2J_e + 1}, \quad (\text{B7})$$

where $\tau_{E3} = 1.6(1)$ yr is the measured lifetime of the unperturbed $F_e = 6$ state [20,22]. We combine Eqs. (B5)–(B7) to derive the transition rate $\mathcal{R}_{E3}^{(F_e \rightarrow 3)}$ for $F_e = 2, 4$, assuming a pure E3 contribution, which is then given in Table II of the main text. We also use Eq. (B4) to evaluate the ratios $\Omega_{E3}^{(3 \rightarrow F_e)}/\Omega^{(3 \rightarrow 6)}$ for $F_e = 2$ and $F_e = 4$.

Additional measurements—Since we observed a large discrepancy between the theoretically predicted HFE1 transition rates $\mathcal{R}_{\text{HFE1}}$ [32] and the experimentally determined effective transition rates $\mathcal{R}_{\text{E1,eff}}$, we performed three additional sets of measurements, summarized in Table III. (1) We compared two sets of unquenched (pure E3) transitions using π -polarized light. The measured Rabi frequency ratios agree with the calculated values, confirming the stability of the laser intensity and spectral purity between the two measurements, as well as validating the derived E3 transition equations presented in EM and SM [49]. (2) By switching the laser polarization $\vec{e}$, we compared the Rabi frequencies of the $\Delta m_F = 0$

TABLE III. Comparison of Rabi frequency ratios. Except the first row, all measurements were performed using the $^{173}\text{Yb}^+$ isotope.

Transition (1)	Transition (2)	$\Omega^{(1)}/\Omega^{(2)}$	
$F_g, m_g \rightarrow F_e, m_e$	$F_g, m_g \rightarrow F_e, m_e$	Measured	Calculated
$-1/2 \rightarrow -1/2^a$	$3, 0 \rightarrow 6, 0$	1.13(8)	1.15
$2, 0 \rightarrow 5, 0$	$3, 0 \rightarrow 6, 0$	0.56(5)	0.54
$3, 0 \rightarrow 4, 0$	$3, 0 \rightarrow 4, +1$	1.66(17)	1.79
$3, 0 \rightarrow 4, 0$	$3, 0 \rightarrow 4, -1$	1.72(17)	1.79
$2, 0 \rightarrow 3, 0$	$3, 0 \rightarrow 6, 0$	2.23(19)	...

^aMeasured on the $^{172}\text{Yb}^+$ isotope (unquenched E3 transition).

TABLE IV. Comparison between experimental and theoretical transition rates. The ratios between the theoretically predicted HFE1 rates $\mathcal{R}_{\text{HFE1}}$ [32] and the effective E1 transition rates $\mathcal{R}_{\text{E1,eff}}$ are calculated, along with the corresponding ratios assuming either destructive ($\mathcal{R}_{\text{E1,dest}}$) or constructive ($\mathcal{R}_{\text{E1,const}}$) interferences between the E3 and HFE1 channels.

$F_g, m_g \rightarrow F_e, m_e$	$(\mathcal{R}_{\text{HFE1}}/\mathcal{R}_{\text{E1,eff}})$	$(\mathcal{R}_{\text{HFE1}}/\mathcal{R}_{\text{E1,const}})$	$(\mathcal{R}_{\text{HFE1}}/\mathcal{R}_{\text{E1,dest}})$
$2, 0 \rightarrow 3, 0$	36.3(7.9)	80.2(25.4)	23.5(4.2)
$3, 0 \rightarrow 2, 0$	29.5(2.7)	52.9(6.0)	18.8(1.5)
$3, 0 \rightarrow 4, 0$	27.4(3.5)	35.4(5.0)	21.8(2.5)

($\vec{e} \parallel \vec{B}$) and $\Delta m_F = \pm 1$ ($\vec{e} \perp \vec{B}$) transitions of the same $^2F_{7/2}$, $F_e = 4$ clock transition. For $\Delta m_F = \pm 1$, the E3 contribution is as low as 2% of the measured Rabi frequency, whereas for $\Delta m_F = 0$ the E3 contributes to 12%. Measurements of transitions with different E3 contributions confirm the approximation made in the main text of neglecting these contributions. This measurement validates the derived equations for HFE1 transitions and confirms the polarization purity of the clock laser (see SM [49]). (3) We coherently excite the $S_{1/2}, F_g = 2 \rightarrow F_{7/2}, F_e = 3$ transition, and compare it with the $F_e = 6$ reference transition. The derived effective E1 transition rate is $\mathcal{R}_{\text{E1,eff}}^{(3 \rightarrow 2)} = 6.9(1.5) \times 10^{-8}/\text{s}$. We include this data and calculate the HFE1 transition rates, which might give hints to the possible origin of the discrepancy between the theory and the experiment.

Besides the effective E1 transition rates, we also calculate the possible HFE1 transition rates (see Table IV). Intriguingly, under the assumption of destructive interference between the HFE1 and E3 channels, the ratios between the derived HFE1 transition rate and the theoretical prediction are nearly identical for all measured transitions. Further theoretical investigation and an independent verification of the $^{171}\text{Yb}^+$ lifetime are needed to resolve this discrepancy.