

Entanglement-Enhanced Optical Ion Clock

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Entangled states hold the promise of improving the precision and accuracy of quantum sensors. We experimentally demonstrate that spectroscopy of an optical clock transition using entangled states can outperform its classical counterpart. Two $^{40}\text{Ca}^+$ ions are entangled in a quantum state with vanishing first-order magnetic field sensitivity, extending the coherence time of the atoms and enabling near-lifetime-limited probe times of up to 550 ms. In our protocol, entangled ions reach the same instability as uncorrelated ions, but at half the probe time, enabling faster cycle times of the clock. We run two entangled $^{40}\text{Ca}^+$ ions as an optical clock and compare its frequency instability with a ^{87}Sr lattice clock. The instability of the entangled ion clock is below a clock operated with classically correlated states for all probe times. We observe instabilities below the theoretically expected quantum projection noise limit of two uncorrelated ions for interrogation times below 100 ms. The lowest fractional frequency instability of $7 \times 10^{-16}/\sqrt{\tau/1\text{ s}}$ is reached for 250 ms probe time, limited by residual phase noise of the probe laser. This represents the lowest instability reported to date for a $^{40}\text{Ca}^+$ ion clock.

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Introduction—Since the seminal proposal to use quantum-correlated squeezed states to overcome the shot noise limit in optical interferometry for gravitational wave detectors [1], many more quantum sensing applications have been proposed to benefit from using entangled states [2–4]. Among them are optical clocks, which are the most accurate measurement devices available today, reaching systematic uncertainties at the 10^{-18} level [5–17]. Metrological gain has recently been demonstrated using entangled neutral atoms in optical lattices and optical tweezers [18–20]. Ion clocks are typically limited by quantum projection noise (QPN) [21], but increasing the number of ions poses a significant technological challenge [12,22–27]. Improving the signal-to-noise ratio using entangled states is therefore particularly attractive for ion clocks [28–30]. Toward this goal, two-ion Rabi and 14-ion Ramsey spectroscopy on an optical transition using entangled ions [31,32] have been demonstrated, optimal quantum states for optical clock operation have been implemented on a programmable quantum computer

[33], and a distributed two-ion entangled state has been employed for optical clock operation [34]. However, a quantum gain in optical clock comparisons exceeding the performance of a competitive version of the clock operating with uncorrelated atoms has not yet been demonstrated. Besides suppressing projection noise, correlated many-particle states can be used to tailor additional desirable features of atomic clocks. In particular, carefully designed entanglement can realize a decoherence-free subspace (DFS) and mitigate systematic or coherence-limiting effects [34,35].

In this Letter, we report the implementation of an entanglement-enhanced optical $^{40}\text{Ca}^+$ ion clock with reduced projection noise, employing a generalized Ramsey protocol [30] with a two-ion Bell state encoded in a DFS immune to magnetic field fluctuations. We demonstrate lifetime-limited coherence time in the DFS and clock instability beyond the best demonstrated $^{40}\text{Ca}^+$ clock [36] by comparing to a Sr optical lattice clock [14].

Entanglement-enhanced protocol—We implement a Cirac-Zoller-type entangling gate [37,38] using resonant interactions between two different pairs of Zeeman states $|S_{\pm}\rangle = |S, m_j = \pm 1/2\rangle \leftrightarrow |D_{\pm}\rangle = |D, m_j = \pm 1/2\rangle$, where m_j is the magnetic quantum number, on the $^{40}\text{Ca}^+ 2S_{1/2} \leftrightarrow 2D_{5/2}$ clock transition at 729 nm. A partial level scheme is shown in Fig. 1(a). For the entangled DFS (enDFS) scheme, we start from the state $|S_{-}\rangle|S_{+}\rangle = |S_{-}S_{+}\rangle$ to prepare the Bell state

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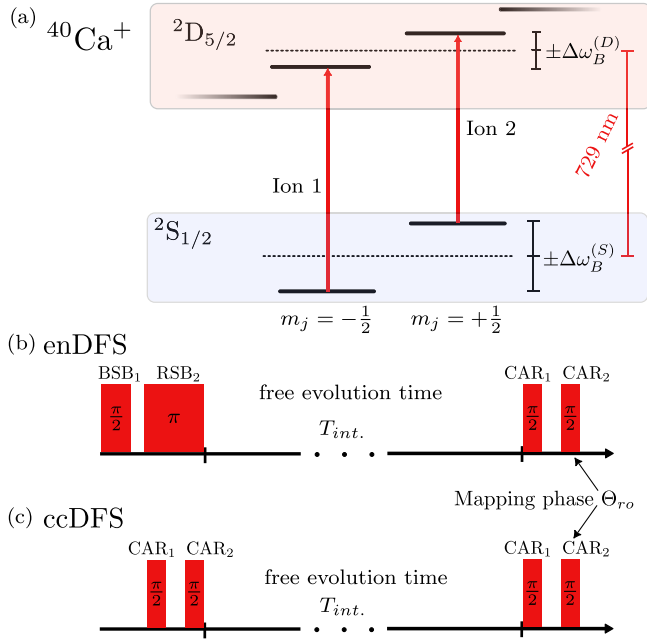


FIG. 1. (a) Partial level scheme of the $2S_{1/2} \leftrightarrow 2D_{5/2}$ transition in $^{40}\text{Ca}^+$. The interferometer is initialized on two different $\Delta m = 0$ transitions. Because of different g factors in the $2S_{1/2}$ and $2D_{5/2}$ states, the Zeeman shifts $\Delta\omega_B^{(S)}$ and $\Delta\omega_B^{(D)}$ result in distinct resonance frequencies for all transitions. (b) Implementation of the entangled Ramsey interferometer (enDFS). Sideband pulses are employed to prepare the state $|\Phi_{\text{Bell}}\rangle$ (see main text for details). (c) Implementation of the classically correlated Ramsey interferometer (ccDFS). The ions are independently prepared by two $(\pi/2)$ carrier pulses. The readout sequence is identical to (b).

$$|\Phi_{\text{Bell}}\rangle = \frac{1}{\sqrt{2}}(|S_-S_+\rangle + e^{-i\Theta}|D_-D_+\rangle). \quad (1)$$

The phase evolution $\Theta(t) = (\omega_- + \omega_+)t$ of the two-ion wave function during time t is given by the energy difference between the two parts of the wave function, where $\omega_{\pm}$ denote the respective transition frequencies between $|S_{\pm}\rangle \leftrightarrow |D_{\pm}\rangle$. Since each part of the wave function consists of states that experience an equal but opposite linear Zeeman shift, the overall state $|\Phi_{\text{Bell}}\rangle$ is free of this shift [35].

During a Ramsey spectroscopy sequence with interrogation time T_{int} , and in the limit where the duration of the excitation pulses can be neglected, the relative phase evolution $\Theta_{\text{rel}}(T_{\text{int}})$ of the spectroscopy laser with frequency $\omega_L(t)$ and the state $|\Phi_{\text{Bell}}\rangle$ is given by

$$\Theta_{\text{rel}}(T_{\text{int}}) = \int_0^{T_{\text{int}}} 2\omega_L(t) - (\omega_- + \omega_+)dt.$$

In the readout stage, this phase is mapped onto multi-ion states of different parity using resonant $\pi/2$ pulses on both atoms [39]. A parity measurement $\hat{P} = \hat{\sigma}_z^{(1)}\hat{\sigma}_z^{(2)}$ of the

resulting quantum state has an expectation value $\langle\hat{P}\rangle = \cos[\Theta_{\text{rel}}(T_{\text{int}}) + \Theta_{\text{RO}}]$, where $\hat{\sigma}_z^{(i)}$ is the Pauli spin operator z for ion i . The phase Θ_{RO} contains all relevant laser phases and can be scanned to obtain the contrast of the signal (see Supplemental Material [40]) or obtain a feedback signal for laser stabilization.

A direct comparison between the performance of the Bell state and a nonentangled two-ion state for an optical clock is complicated by the fact that the simplest product state $(|S_+\rangle + |D_+\rangle)^{\otimes 2}$ is highly magnetic field sensitive and therefore does not allow for the same long Ramsey times as the Bell state. However, starting from the state $|S_-S_+\rangle$, one can also obtain a classically correlated state in the DFS (ccDFS) using a simplified version of the entangled protocol [39].

Two independent Ramsey $\pi/2$ pulses on the two ions result in the state

$$\begin{aligned} |\psi_{\text{cc}}\rangle &= \frac{1}{2}(|S_- \rangle + e^{-i\omega_-t}|D_- \rangle) \otimes (|S_+ \rangle + e^{-i\omega_+t}|D_+ \rangle) \\ &= \frac{1}{\sqrt{2}}(|\Phi_{\text{Bell}}\rangle + |\Psi_{\text{Bell}}\rangle), \quad \text{with} \\ |\Psi_{\text{Bell}}\rangle &= \frac{1}{2}(|S_-D_+\rangle + e^{-i(\omega_- - \omega_+)t}|S_+D_-\rangle). \end{aligned}$$

The state is split into two parts with equal probability. The first part consists of the desired Bell state, $|\Phi_{\text{Bell}}\rangle$, in the DFS. The second part, $|\Psi_{\text{Bell}}\rangle$, evolves at the difference frequency between the two parts of its wave function, which is dominated by their magnetic field splitting. Consequently, this component dephases for Ramsey times exceeding the coherence time of magnetic-field-sensitive states, resulting in a mixed state described by the density operator $\rho = \frac{1}{2}|\Phi_{\text{Bell}}\rangle\langle\Phi_{\text{Bell}}| + \frac{1}{4}|S_-D_+\rangle\langle S_-D_+| + \frac{1}{4}|D_-S_+\rangle\langle D_-S_+|$. While this state exhibits a component of the Bell state $|\Phi_{\text{Bell}}\rangle$, the density operator overall represents a separable, which is classically correlated, state only. A parity measurement using this density operator will therefore result in the same phase evolution as using the entangled state $|\Phi_{\text{Bell}}\rangle$, but with half the measurement amplitude [34,39].

Spontaneous decay of the excited clock state will decrease the measurement contrast for interrogation times T_{int} approaching the lifetime of the excited state $t_{\text{sp}} = (1/\Gamma)$, where Γ is the decay rate from the excited state. Incorporating the spontaneous decay for standard Ramsey spectroscopy with N uncorrelated ions on a transition with frequency ω_0 yields a fractional frequency instability after an integration time τ of

$$\sigma_y(\tau) = \frac{e^{\Gamma T_{\text{int}}}}{\omega_0 \sqrt{N}} \frac{1}{\sqrt{T_{\text{int}}\tau}},$$

which is the QPN limit for uncorrelated atoms. The lowest instability is achieved for the optimal Ramsey time

$T_{\text{int}} = 1/\Gamma$ [41,42]. Two-particle Bell states exhibit a reduced lifetime $1/\Gamma_{\text{Bell}} = 1/(N\Gamma)$, which accelerates the decay of measurement contrast. Consequently, the optimal interrogation time is given by $T_{\text{int}} = 1/\Gamma_{\text{Bell}}$, resulting in the same instability as for N uncorrelated particles, but at N -times-shorter optimal Ramsey times [43] (see also Fig. 4). Well below the lifetime limit, an entangled two-ion clock can reach a gain of $\sqrt{2}$ in stability compared to uncorrelated ions for fixed interrogation times.

Experimental implementation—A detailed description of the experimental apparatus is given in [44] and the Supplementary Material [40]. In brief, we trap two $^{40}\text{Ca}^+$ ions in a linear Paul trap with single-ion trap frequencies $(\omega_z, \omega_x, \omega_y) = 2\pi \times (0.8, 1.8, 1.9)$ MHz. A stabilized magnetic field of 0.356 mT aligned with the axial trap axis lifts the degeneracy of the S - and D -state Zeeman sublevels, spectrally separating the two used transitions by 4 MHz. Residual magnetic field fluctuations broaden the linewidth of these transitions to approximately 100 Hz.

Before the interferometry sequence, the ions are cooled via Doppler and successive electromagnetic transparency cooling [45,46]. The axial out-of phase mode at a frequency of 1.4 MHz is used for the entangling operation and therefore further cooled by resolved sideband cooling close to the ground state of motion. The initial state $|\psi_i\rangle = |S_-S_+\rangle$ is prepared starting with both ions optically pumped into the $|S_-S_- \rangle$ state and transferring one of the ions to the $|S_+\rangle$ state via the $|D_+\rangle$ state using single-ion addressing and a composite pulse sequence (see Supplemental Material [40] and Ref. [47]).

Cooling, state preparation, detection, and clock feedback contribute to a total dead time of 12.5 ms per experimental clock cycle. We use red (RSB) and blue (BSB) sideband pulses which, respectively, add and remove one quantum of motion while driving the ion from S to D during the entangling operations. The Bell state $|\Phi_{\text{Bell}}\rangle$ for the enDFS scheme is prepared by a $\pi/2$ BSB₁ pulse on ion 1, followed by a π RSB₂ pulse on ion 2 [see Fig. 1(b)] [38]. The ions are selectively addressed using a global laser beam exploiting their frequency separation. The acquired phase $\Theta_{\text{rel}}(T_{\text{int}})$ is mapped by $(\pi/2)$ carrier pulses on both ions (CAR_{1,2}) onto different parity states. The clock laser is derived from an extended cavity diode laser (ECDL) locked to a high-finesse cavity; its filtered transmission seeds a second ECDL, whose amplified output is used in the experiment [48,49]. The laser frequency is further stabilized via a transfer oscillator lock [50] to an ultrastable reference laser using an optical frequency comb. This reference laser [51] has a coherence time longer than the excited state lifetime of the atoms of $t_{\text{sp}} \approx 1.17$ s.

The coherence time is typically limited by the first-order Zeeman shift, which is ≈ 2 MHz at a sensitivity of 5.6 MHz mT for the two transitions used within this experiment. We determine the coherence time for the

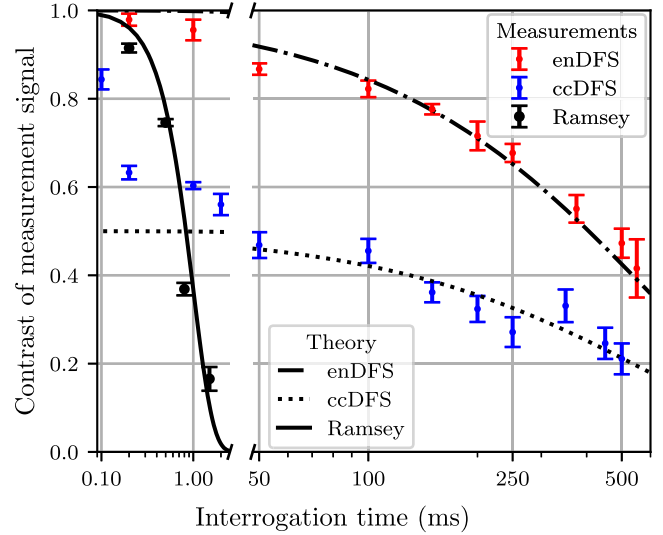


FIG. 2. Coherence time for different probe schemes. Contrast for the two-ion entangled (enDFS, red) and correlated schemes (ccDFS, blue) as well as a Ramsey measurement with a single ion (black). The contrast was inferred from fringe measurements of parity (DFS schemes) and excitation (Ramsey) signals for different interrogation times and fitting a sine function with varying amplitude. The theoretical lifetime-limited contrast is shown for the enDFS (dash-dotted) and ccDFS (dotted) measurement schemes. For short interrogation times, the contrast of the single ion drops in good agreement with a Gaussian noise model with 1 ms time constant. In contrast, interrogation within the DFS ensures close to lifetime-limited coherence.

enDFS and ccDFS schemes from the contrast of parity oscillations at several interrogation times by scanning the mapping phase. Similarly, for the single-ion Ramsey scheme, we extract the coherence time from the oscillations in the excitation signal. Figure 2 shows that the contrast for a single ion is well described by a Gaussian noise process $\exp(-t^2/t_c^2)$ with a coherence time $t_c \sim 1$ ms. In contrast, the effective transition frequency of the DFS state has a strongly suppressed first-order Zeeman shift extending the coherence time toward the lifetime limit with a contrast given by the envelope $\exp(-t\Gamma_{\text{Bell}})$. A measurement contrast of 0.98 and 0.46 for the enDFS and ccDFS schemes, respectively, could be achieved, limited by high frequency laser noise, magnetic field fluctuations, and crosstalk between the excitation pulses. The measurements show that the DFS extends the possible interrogation times by more than 2 orders of magnitude.

Clock stability—We operate an optical frequency reference using the enDFS and ccDFS protocols and compare it to a ^{87}Sr lattice clock [14], providing a reference stability of $1.1 \times 10^{-16} \times (\tau/1 \text{ s})^{-1/2}$ well suited for a high stability frequency comparison. All fibers involved in the clock comparison are length stabilized [52]. The clock laser is stabilized to the parity signal using a two-point sampling method analogous to [41]. During the frequency comparison, the contrast is on average 0.92 (0.43) for the enDFS

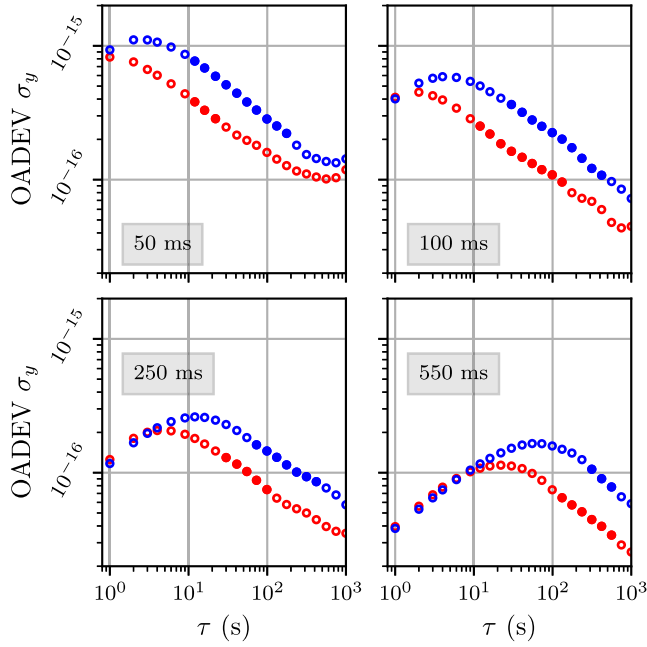


FIG. 3. Fractional stability of the frequency ratio of the $^{40}\text{Ca}^+ / ^{87}\text{Sr}$ frequency ratio. Data for the enDFS (red) and ccDFS (blue) measurement schemes are shown for four different interrogation times. The fractional stability is given as the overlapping Allan deviation of the data traces (see text for details). Error bars are smaller than the symbol size; filled markers indicate the data range taken for the stability fit.

(ccDFS) scheme due to drifts of the motional and carrier frequencies as well as the diminishing fidelity of the single-ion addressing. The contrast directly affects the achievable frequency stability. Figure 3 shows the stability of the frequency ratios for interrogation times of 0.05, 0.1, 0.25, and 0.55 s determined using the overlapping Allan deviation (OADEV) [53] over averaging intervals ranging from 1 to 1000 s.

An OADEV as low as 2.5×10^{-17} for a 550 ms probe and 1000 s averaging time has been achieved for the enDFS scheme. As theoretically predicted, the enDFS scheme shows significantly shorter servo time constants [42,43] and an improvement in stability compared to the ccDFS scheme for all probe times. The instabilities are constrained for long averaging times by a flicker floor that depends on the interrogation time. The flicker floor level is compatible with a fluctuating differential phase evolution between the two ions when opening and closing the interferometer: Consecutive excitation of the two ions results in short (20 μs) periods of additional phase evolution in magnetic-field-sensitive states. A small and varying detuning (≈ 100 Hz) from magnetic field drifts and power line noise accounts for the measured frequency deviations. Especially for the enDFS scheme, additional frequency instability over long timescales may arise from ac Stark shifts of the global laser.

While we have not performed a systematic shift evaluation, we expect several additional contributions that need to be addressed (for details, see Supplemental Material [40]). We estimate ac Stark shifts from the global addressing beam to be on the order of 1 to 10 Hz for the enDFS and 1 to 10 mHz for the ccDFS scheme. They can be mitigated by employing suppression schemes [54–58]. The effect of static magnetic field gradients (≈ 100 Hz) can be eliminated by averaging over the Zeeman-state assignment of the two ions or eliminating the gradient altogether. The electric quadrupole shift (≈ 2 Hz) can be significantly reduced by aligning the magnetic field at an angle of 54.7° with respect to the trap axis [23] and eliminated by averaging over all Zeeman components in the excited state [59]. Our measurements show that these shifts remain stable to a level of 10 mHz over the timescales investigated.

Instabilities of the frequency comparison $\sigma_{y,1s}$ were obtained by a least-squares fit $\sigma_y(\tau) = \sigma_{y,1s}(1/\sqrt{\tau})$ to the white-frequency-noise-dominated regime of the OADEV [60,61] (see Supplemental Material for details [40]).

We summarize the measured instabilities and compare them with theoretical predictions in Fig. 4. The theory

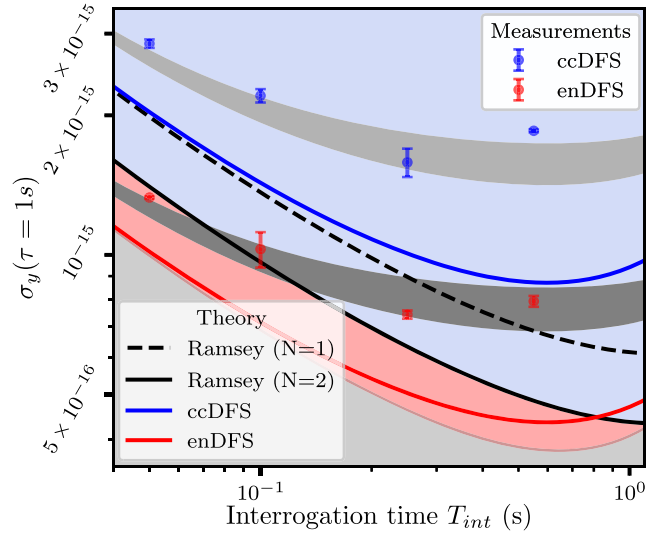


FIG. 4. Overview of measured stabilities and theoretical quantum projection noise limits as a function of the interrogation time for the three interrogation schemes discussed (enDFS, ccDFS, and standard Ramsey). Theoretical values assume ideal signal contrast and perfect laser stability but include a 12.5 ms dead time and a reference stability of $1.1 \times 10^{-16} \times (\tau/1 \text{ s})^{-1/2}$ based on the performance of the ^{87}Sr lattice clock. The colored regions classify different theoretical stability regimes in the presence of spontaneous decay. Classical interrogation protocols are shaded in blue, stabilities only obtainable by entanglement in red, including protocols mitigating feedback errors from spontaneous emission [62] that even exceed the levels of the enDFS protocol. Expected stabilities accounting for contrast loss and a white-frequency-noise term of $6(1) \times 10^{-16}(\tau/1 \text{ s})^{-1/2}$ are shown in dark gray (enDFS) and light gray (ccDFS).

curves have been calculated including the change in duty cycle for varying probe times. We find good agreement with a model including the effective measurement contrast and an additional phase noise (white frequency modulation) with a noise level of around $6(1) \times 10^{-16} \times (\tau/1 \text{ s})^{-1/2}$. As shown in Fig. 4, the theoretical instability of the enDFS scheme is always smaller compared to the ccDFS and uncorrelated two-ion scheme (assuming a perfectly stable magnetic field) for probe times shorter than the optimum probe time. The lowest theoretical instability is identical to a classical two-ion Ramsey scheme but reached at half the optimal probe time [43]. The ccDFS scheme has an identical scaling to the enDFS scheme but is shifted to twice the instability due to the smaller signal contrast. The single-ion curve is larger by a factor of $\sqrt{2}$ compared to the two-ion Ramsey scheme. The experimentally observed increase in instability for an interrogation time of 550 ms is compatible with uncontrolled optical path length fluctuations of the clock laser beam through the optical setup.

Experimentally, the enDFS scheme shows an instability below the QPN for two uncorrelated ions at an interrogation time of 50 ms. The lowest instability of 7.2×10^{-16} at 1 s is achieved for a probe time of 250 ms and surpasses, to our knowledge, the most stable optical frequency demonstrated with $^{40}\text{Ca}^+$ ions (1.15×10^{-15} at 1 s) [63–65], remaining only 45% above the theoretical limit.

Conclusion—We have demonstrated metrological gain beyond uncorrelated atoms for the shortest interrogation time. For longer probe times, entanglement in a DFS provides magnetic field insensitivity which enables near-lifetime-limited probe times and therefore record-low instability. For clocks limited by spontaneous emission from the excited state, a slight variation of the protocol demonstrated here turns out to provide the optimal improvement beyond the standard quantum limit, approaching fundamental limits also for larger ensembles up to several tens of atoms (see red-shaded area below the enDFS limit in Fig. 4) [62]. While in the demonstrated schemes the ultimate instability is the same for GHZ states and uncorrelated atoms, the optimum probe time for GHZ states scales with $1/N$. This supports high-bandwidth feedback schemes, which ease the requirements for long-term clock laser stability and systematic shifts such as second-order Doppler shifts from background heating of the ions in the trap [66,67]. Clocks using species like $^{171}\text{Yb}^+$, $^{115}\text{In}^+$, $^{176}\text{Lu}^+$, and $^{27}\text{Al}^+$ could use an adopted entangled state involving Zeeman and hyperfine components to eliminate not only magnetic field, but also electric quadrupole shifts, while increasing their locking bandwidth. The higher locking bandwidth enables searches for new ultralight scalar bosons, such as dark matter candidates, with larger masses corresponding to larger Compton frequencies [10,68,69].

The systematic uncertainties of the clock will be evaluated in future work. Scaling to more than two ions is complicated by an inhomogeneous electric quadrupole shift of the excited clock state across the ion crystal [26,70]. These shifts can be mitigated by employing dynamical decoupling schemes [22,24,71–75] or more complex multi-ion states in a DFS, enabling multi-ion entangled clocks that perform better than an optimized classical clock of the same species.

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Data availability—The data that support the findings of this article are openly available [76,77].

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