



Heisenberg-Limited Atom Clocks Based on Entangled Qubits

Citation

Kessler, E. M., P. Kómár, M. Bishof, L. Jiang, A. S. Sørensen, J. Ye, and M. D. Lukin. 2014. "Heisenberg-Limited Atom Clocks Based on Entangled Qubits." *Physical Review Letters* 112 (19) (May).

Published Version

doi:10.1103/PhysRevLett.112.190403

Permanent link

<http://nrs.harvard.edu/urn-3:HUL.InstRepos:12242255>

Terms of Use

This article was downloaded from Harvard University's DASH repository, and is made available under the terms and conditions applicable to Open Access Policy Articles, as set forth at <http://nrs.harvard.edu/urn-3:HUL.InstRepos:dash.current.terms-of-use#OAP>

Share Your Story

The Harvard community has made this article openly available.
Please share how this access benefits you. [Submit a story](#).

[Accessibility](#)

Heisenberg-limited atom clocks based on entangled qubits

E. M. Kessler,^{1,2,*} P. Kómár,^{1,*} M. Bishof,³ L. Jiang,⁴ A. S. Sørensen,⁵ J. Ye,³ and M. D. Lukin¹

¹*Physics Department, Harvard University, Cambridge, MA 02138, USA*

²*ITAMP, Harvard-Smithsonian Center for Astrophysics, Cambridge, MA 02138, USA*

³*JILA, National Institute of Standards and Technology, Department of Physics, University of Colorado, Boulder, Colorado 80309-0440, USA*

⁴*Department of Applied Physics, Yale University New Haven, CT 06520, USA*

⁵*QUANTOP, Danish National Research Foundation Centre of Quantum Optics, Niels Bohr Institute, DK-2100 Copenhagen, Denmark*

(Dated: October 24, 2013)

We present a quantum-enhanced atomic clock protocol based on groups of sequentially larger Greenberger-Horne-Zeilinger (GHZ) states, that achieves the best clock stability allowed by quantum theory up to a logarithmic correction. The simultaneous interrogation of the laser phase with such a cascade of GHZ states realizes an incoherent version of the phase estimation algorithm that enables Heisenberg-limited operation while extending the Ramsey interrogation time beyond the laser noise limit. We compare the new protocol with state of the art interrogation schemes, and show that entanglement allow a significant quantum gain in the stability for short averaging time.

PACS numbers: 03.65.-w 42.50.Dv 06.20.-f 06.30.Ft

Keywords: ...

High precision atomic frequency standards form a cornerstone of precision metrology, and are of great importance for science and technology in modern society. Currently, atomic clocks based on optical transitions achieve the most precise [1, 2] and accurate [3, 4] frequency references. Additionally, the development of optical frequency combs [5–8] – establishing a coherent link between the optical and radio frequencies – enabled the application of optical frequency standards to a wide range of scientific and technological fields including astronomy, molecular spectroscopy and global positioning systems (GPS).

The improvement of frequency standards using quantum resources, such as entanglement [9–13], has been actively explored in recent years. A characterization of the improvement obtainable by using entanglement requires a detailed investigation of the decoherence present in the system. Previous studies have focused on two kind of noise sources: i) single particle decoherence resulting from the interaction of the atoms with the environment and ii) fluctuation in the local oscillator (LO) used to interrogate the atoms. It is well known that fully entangled states (e.g., Greenberger-Horne-Zeilinger (GHZ) states) allow for improved spectroscopic sensitivity, but in the same way that these states benefit from their increased sensitivity in the laser interrogation, they are generically prone to various types of noise sources canceling any quantum gain. It has therefore been long believed that such states fail to increase clock stability in the standard Ramsey type protocol regardless of the noise model being used [12, 14–16]. On the other hand, it has been shown that for clocks with local oscillator (LO) noise limited stability, the use of moderately squeezed

atomic states can yield a modest improvement over the standard quantum limit (SQL) [10, 11]. A recent study demonstrated further that, in principle, highly squeezed states could achieve Heisenberg-limited stability using a complex adaptive measurement scheme [13]. At the same time, it has been shown that the single particle decoherence-limited regime can be reached for long averaging time at a logarithmic cost in the number of qubits by interrogating uncorrelated atomic ensembles for suitably chosen times [17, 18].

In this Letter, we introduce a protocol involving groups of sequentially larger GHZ states to estimate local oscillator deviations from the atomic reference in a manner reminiscent of the phase estimation algorithm [19]. Furthermore, we unify previous treatments of decoherence for atomic clocks and incorporate previous proposals involving uncorrelated atoms to effectively narrow the LO linewidth [17, 18] and thereby identify ultimate limits to the stability of atomic clocks based on entangled atoms. The central results of our work are illustrated in Fig. 1, which compares the performance of the proposed protocol with other known approaches as a function of averaging time. Specifically, for LO-noise limited clocks, the proposed quantum protocol is found to be nearly optimal, realizing the Heisenberg limit of clock stability up to a logarithmic correction in the particle number. Importantly, it reaches the fundamental noise floor (dashed line) resulting from individual dephasing of the clock qubits N times faster than the best known classical schemes, where N is the total number of particles employed.

The central idea of our approach can be understood as follows. In modern atomic clocks, the frequency of a LO is locked to an ultra-narrow optical (or radio-frequency) transition of the clock atoms serving as the frequency reference. The long-term stability of such a clock af-

*These authors contributed equally to this work

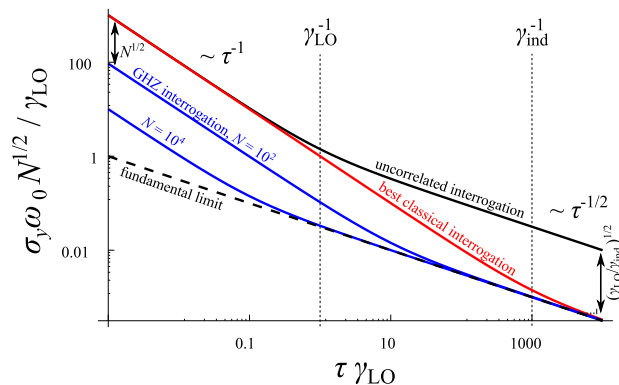


FIG. 1: Allan deviation σ_y for different protocols as a function of averaging time τ , normalized to the standard quantum limit, for $\gamma_{LO}/\gamma_{ind} = 10^3$. The solid black line corresponds to the standard scheme using a single uncorrelated ensemble. It fails to reach the fundamental noise floor set by the atomic transition linewidth (cf. Eq. (5), broken line). A more sophisticated classical scheme which uses exponentially increasing Ramsey times in each cycle [17, 18] allows to extend the regime of linear scaling with $1/\tau$ up to the point where the bound (5) is met. In comparison, the proposed cascaded GHZ protocol (blue solid curves) enables an $\sim N$ times faster convergence. For short averaging times the stability is enhanced by a factor \sqrt{N} as compared to classical protocols.

ter a given total averaging time τ is directly related to the precision by which the accumulated laser phase relative to the atoms can be determined. To this end, in the standard Ramsey protocol, the phase is repeatedly measured over cycle times $T < \tau$, followed by a correction of the laser frequency according to the measurement outcome. Since each Ramsey sequence introduces measurement noise, it is optimal to extend the Ramsey time T as much as possible, ideally to its maximum value $T \rightarrow \tau$. A single GHZ state consisting of N entangled atoms – whose state after the interrogation is $|\text{GHZ}\rangle_T \propto |0\rangle^{\otimes N} + \exp(-iN\Phi_{LO})|1\rangle^{\otimes N}$ – accumulates the laser phase (denoted by Φ_{LO}) N times faster than an uncorrelated state. Thus, in principle, it allows to measure the phase with the best precision allowed by quantum mechanics [20]. However, fluctuations in the laser frequency account for the fact that the laser phase we aim to measure is a random variable with a probability distribution that grows in width as we increase the Ramsey time T . Whenever the laser phase realized in a particular Ramsey cycle induces a full phase wrap on the state [i.e., the atomic phase $N\Phi_{LO} \notin [-\pi, \pi]$] a subsequent measurement yields a 2π error in the estimation of the true laser phase. For a single GHZ state this accounts for a strict limitation on the maximally allowed Ramsey time in order to limit the initial variance of Φ_{LO} , and the resulting laser stability is found to yield no improvement over classical protocols [16].

To address this problem, we use a protocol reminiscent of the *phase estimation algorithm* [19] that allows a direct assessment of the number of 2π phase wraps during the

interrogation, thus enabling the Ramsey time to extend to its maximum value to guarantee optimal laser stability. Let us assume for the moment that the accumulated laser phase after the interrogation time T lies in the interval $\Phi_{LO} \in [-\pi, \pi)$, and has an exact binary representation $(\Phi_{LO} + \pi)/2\pi = \sum_{j=1}^M Z_j/2^j$, with digits $Z_j \in \{0, 1\}$ (both conditions will be relaxed below). One can then readily show, that a GHZ state consisting of 2^{M-1} atoms picks up the phase $\Phi_{M-1} = 2^{M-1}\Phi_{LO} \bmod [-\pi, \pi) = \pi(Z_M - 1)$. Thus, by measuring if the phase is 0 or π , the last digit of the laser phase can be determined. However, as stated above, without knowledge of the remaining digits (i.e., the number of phase wraps) this information is useless. In our protocol, these digits are found by an additional simultaneous interrogation with successively smaller GHZ states of $2^{M-2}, 2^{M-3}, \dots$ entangled atoms (see Fig. 2). Each of these states picks up a phase proportional to its size $\Phi_j = 2^j\Phi_{LO} \bmod [-\pi, \pi)$, and in analogy to the first state this phase gets a contribution of $\pi(Z_j - 1)$. By distinguishing whether the phase is shifted by π or not, we can thus determine the value of the bit Z_j . The combined information provides an estimate with an accuracy given by the largest GHZ state, while the cascade increases the total number of atoms employed only modestly by a factor of two: $\sum_{j=0}^{M-1} 2^j \approx 2^M = 2 \times 2^{M-1}$.

However, in the limit of large averaging times, the assumption $\Phi_{LO} \in [-\pi, \pi)$ is not justified anymore. Here, the optimal Ramsey time $T \sim \tau$ can attain values that induce phase wraps of the laser itself, causing the binary representation of the laser phase to contain digits $Z_j \neq 0$ for $j \leq 0$ which are inaccessible to the technique discussed so far. To achieve the optimal laser stability in this regime, we extend the cascade to the classical domain, and employ additional groups of uncorrelated atoms that interrogate the laser with successively decreasing interrogation times, or alternatively, using dynamical decoupling techniques [17, 18, 21]. Each of these ensembles acquires a phase that again is reduced by multiples of two from the laser phase, and thus, following the arguments from above, allows one to gain information on the lower digits Z_j with $j \leq 0$. The information of all digits combined provides the total number of phase wraps, which in turn yields a Heisenberg-limited estimate of the laser phase. By this, the protocol effectively eliminates all limitations arising from the LO noise, and allows the Ramsey time to extend to its optimal value to achieve the best laser stability allowed by quantum mechanics (up to a logarithmic correction as shown below).

In the following, we provide a detailed derivation of the above results. Modern clocks periodically measure the fluctuating LO frequency $\omega(t)$ against the frequency standard ω_0 of a given ensemble of clock atoms (qubits) to obtain an error signal. After each Ramsey cycle of duration T [i.e., at times $t_k = kT$ ($k = 1, 2, \dots$)], the measurement data yield an estimate of the relative phase $\Phi_{LO}(t_k) = \int_{t_k-T}^{t_k} dt [\omega(t) - \omega_0]$ accumulated by the LO. This estimate in turn is used to readjust the frequency

of the LO: $\omega(t_k) \rightarrow \omega(t_k) - \alpha \Phi_{\text{LO}}^{\text{est}}(t_k)/T$, where $\Phi_{\text{LO}}^{\text{est}}(t_k)$ represents a suited estimator of the phase $\Phi_{\text{LO}}(t_k)$ [22], and $\alpha < 1$ is an suitably chosen gain.

The stability of the actively stabilized LO after a total averaging time τ is characterized by the Allan deviation (ADEV) which is directly proportional to the measurement uncertainty $\Delta\Phi_{\text{LO}}(t_k)$ after each Ramsey cycle [23],

$$\sigma_y(\tau) \equiv \frac{1}{\omega_0 \tau} \sqrt{\sum_{k=1}^{\tau/T} \sum_{l=1}^{\tau/T} T^2 \langle \delta\bar{\omega}_k \delta\bar{\omega}_l \rangle} \approx \frac{1}{\omega_0 \sqrt{\tau T}} [\Delta\Phi_{\text{LO}}(T)]. \quad (1)$$

Here, $\delta\bar{\omega}_k = \Phi_{\text{LO}}(t_k)/T$ is the average detuning of the (stabilized) LO during the k th cycle. To obtain Eq. (1) we use the fact that after the frequency feedback the detuning averages become approximately uncorrelated for realistic laser spectra, $\langle \delta\bar{\omega}_k \delta\bar{\omega}_l \rangle \approx \langle \delta\bar{\omega}^2 \rangle \delta_{kl}$ [4, 18, 24]. Other noise sources (such as the bias of the linear estimator, the Dick effect, or a sub-optimal gain α [25]) are not fundamental, and neglected in the following.

The ultimate precision by which the accumulated Ramsey phase after each cycle can be estimated is in principle (i.e., in the limit of small phases) determined by the Cramér-Rao bound [20, 26] which, e.g., is achieved by the use of GHZ states. However, as a consequence of the LO frequency fluctuations, in general, large phases can occasionally be acquired, leading to uncontrolled phase wraps of the atomic phase, $\Phi(t_k) = N\Phi_{\text{LO}}(t_k) \notin [-\pi, \pi)$. To suppress these events, the cycle time T has to be chosen such that the prior distribution of Φ is well localized within $[-\pi, \pi)$. This limits the maximally allowed Ramsey time to a value $T_{\text{max}} \sim \gamma_{\text{LO}}^{-1}/N^2$, where we assumed a white frequency noise spectrum of the LO, $S_\omega(f) = \gamma_{\text{LO}}$ (for $1/f$ -noise one finds the less stringent condition $T_{\text{max}} \sim \gamma_{\text{LO}}^{-1}/N$). In most cases, this value lies below the optimal (i.e., maximal) value implied by Eq. (1) $T \sim \tau$, resulting in a laser stability for GHZ states which shows no improvement over the stability achieved with uncorrelated atoms [12, 16].

However, unlike the individual particle noise resulting in the finite atom linewidth γ_{ind} , the LO frequency fluctuations affect all clock atoms alike, and this *collective noise* does not represent a fundamental metrological limitation. Assuming for the moment that the prior distribution of the LO phase Φ_{LO} after the Ramsey time is localized in $[-\pi, \pi)$ (we will relax this condition below), we can use groups of GHZ states of varying size to measure the Φ_{LO} in a binary representation, as discussed above. In general, however, the phase does not have an exact binary representation ending at the digit Z_M . We therefore employ n_0 duplicates of the GHZ states at each level of the cascade ($n_0 = N/\sum_{j=0}^{M-1} 2^j \approx N/2^M$) to improve the precision. In the case where all digits Z_j ($j = 1 \dots, M-1$) are determined correctly according to the relation

$$Z_j = [2(\Phi_{j-1} + \pi) - (\Phi_j + \pi)]/2\pi, \quad (2)$$

the last group ($j = M-1$) then yields a Heisenberg-

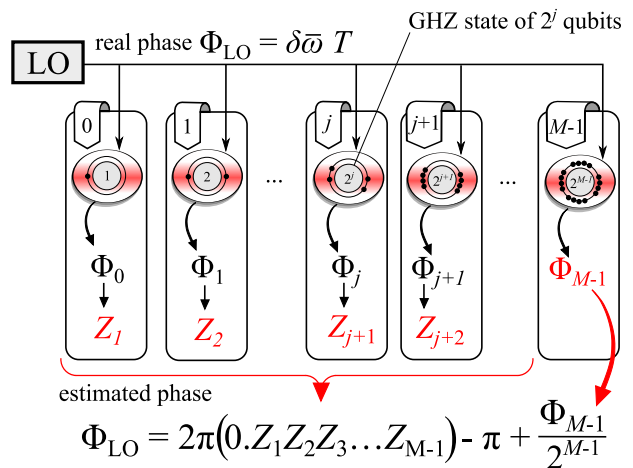


FIG. 2: The proposed clock operation scheme employs M different groups of clock atoms prepared in correlated states of varying size to interrogate the relative phase Φ_{LO} of the LO field. A single group j contains n_0 independent instances of GHZ-like states, each entangling 2^j qubits, and therefore accumulating a phase $\Phi_j = 2^j \Phi_{\text{LO}} \bmod [-\pi, \pi]$ during a single cycle. Each group is then used to measure this phase, which gives a direct estimate on the digit Z_{j+1} in a binary representation of the LO phase $(\Phi_{\text{LO}} + \pi)/2\pi = (0.Z_1 Z_2 Z_3 \dots)$. This estimate is subsequently used to feedback the LO frequency. This clock operation protocol achieves the best stability allowed by quantum mechanics up to a logarithmic correction.

limited estimate of the LO phase with accuracy $(\Delta\Phi_{\text{LO}})_{\text{pr}} = 1/(2^{M-1}\sqrt{n_0}) = 2\sqrt{n_0}/N$, and with a Ramsey time that can exceed the laser noise limit T_{max} .

However, in general the estimation of the binary digits Z_j is not perfect. A rounding error occurs whenever $|\Phi_{j-1}^{\text{est}} - \Phi_{j-1}| > \pi/2$ (where Φ_j^{est} represents a suitable estimator derived from the n_0 measurement outcomes), leading to a wrong digit Z_j . The variance contribution of such an event to the total measurement uncertainty is $(2\pi 2^{-j})^2$. Since these errors are small and independent, we can approximate their total variance contribution by the sum $(\Delta\Phi_{\text{LO}})_{\text{re}}^2 = P_{\text{re}} \sum_{j=1}^{M-1} (2\pi 2^{-j})^2$. The corresponding probability in a single cycle is $P_{\text{re}} = 2 \int_{\pi/2}^{\infty} d\phi \rho(\phi)$, where $\rho(\phi)$ is the Gaussian probability distribution of the error $\Phi_j^{\text{est}} - \Phi_j$ with a width proportional to $1/\sqrt{n_0}$ [23]. Consequently, rounding errors can be exponentially suppressed by choosing a sufficiently large value for n_0 , and for $P_{\text{re}} \ll 1$, the total measurement uncertainty of this estimation scheme is thus $(\Delta\Phi_{\text{LO}})^2 = (\Delta\Phi_{\text{LO}})_{\text{pr}}^2 + (\Delta\Phi_{\text{LO}})_{\text{re}}^2$. In [23] we show that the optimal allocation of resources is achieved for the choice $n_0^{\text{opt}} \sim \frac{16}{\pi^2} \log(N)$ for which rounding errors are negligible, yielding the total measurement accuracy

$$\Delta\Phi_{\text{LO}} \approx (\Delta\Phi_{\text{LO}})_{\text{pr}} = \frac{8}{\pi} \sqrt{\log(N)}/N. \quad (3)$$

This measurement precision obtains the fundamental Cramér-Rao bound (up to a logarithmic correction resulting from the cost to suppress rounding errors) despite

it being applicable to a general (typically large) phase.

So far we have assumed that $\Phi_{\text{LO}} \in [-\pi, \pi)$ in each cycle. However, for realistic laser noise spectra there is always a finite probability that the LO phase Φ_{LO} lies outside the interval $[-\pi, \pi)$ after the interrogation time. Such phase wraps of the laser phase itself add to the final measurement uncertainty in Eq. (3) by the amount $(\Delta\Phi_{\text{LO}})_{\text{slip}}^2 = (2\pi)^2 P_{\text{slip}}$. The corresponding probability after a single Ramsey cycle is $P_{\text{slip}} = 2 \int_{\pi}^{\infty} d\phi \rho_{\text{LO}}(\phi)$, where ρ_{LO} is the Gaussian prior distribution of Φ_{LO} . Its width depends directly on the product $\gamma_{\text{LO}} T$, which puts a constraint on the maximally allowed Ramsey time $T \leq \frac{\pi^2}{4} \gamma_{\text{LO}}^{-1} [\log(\gamma_{\text{LO}} \tau N)]^{-1}$, and thus the achievable ADEV σ_y ($\propto 1/\sqrt{T}$) as we demonstrate in [23].

As discussed above, this does not represent a fundamental limitation as we can extend the scheme by adding additional classical measurements with a shorter Ramsey periods (or alternatively, by employing dynamical decoupling techniques) [17, 18, 21] to assess the number of phase slips of the laser phase itself ... $Z_{-3} Z_{-2} Z_{-1} Z_0$. As demonstrated in [23] this allows extending the Ramsey time by a factor k adding only a negligible number of atoms $N^* \approx \frac{8}{\pi^2} \log(kN^2) \log_2(k) \ll N$.

With all phase wraps counted correctly, the Ramsey time is only limited by individual noise processes. The finite linewidth of the atomic clock transition γ_{ind} gives rise to the fundamental constraint $T \leq \gamma_{\text{ind}}^{-1}/2^{M-1}$. For averaging times smaller than this value, $\tau \leq \gamma_{\text{ind}}^{-1}/2^{M-1}$, we can choose $T \approx \tau$, and using the optimized value for n_0 found above the resulting clock stability is obtained from Eq. (1)

$$\sigma_y(\tau)^{(1)} \approx \frac{2}{\omega_0 \tau} \frac{\sqrt{n_0^{\text{opt}}}}{N} \approx \frac{8}{\pi \omega_0 \tau} \frac{\sqrt{\log(N)}}{N}. \quad (4)$$

It scales linearly with the averaging time τ , and realizes the Heisenberg bound of laser stability up to a logarithmic correction. In contrast, in the regime $\tau \geq \gamma_{\text{ind}}^{-1}/2^{M-1}$, T is limited by the presence of individual particle noise to a value $T \approx \gamma_{\text{ind}}^{-1}/2^{M-1} = 2\gamma_{\text{ind}}^{-1} n_0/N$, and we find

$$\sigma_y(\tau)^{(2)} \approx \frac{1}{\omega_0} \sqrt{\frac{\gamma_{\text{ind}}}{\tau N}}. \quad (5)$$

Eq. (5) represents the fundamental noise floor for laser stability resulting from quantum metrological bounds in the presence of individual particle noise [30]. As we have seen, the proposed protocol reaches this optimal value rapidly after the averaging time $\tau_0 \sim \gamma_{\text{ind}}^{-1} \log(N)/N$ (cf. Fig. 1), $N/\log(N)$ times faster than any classical scheme.

In the following, we benchmark the stability of our protocol against different approaches by comparing the lowest achievable ADEV as a function of averaging time τ (cf. Fig. 1). First, we consider the standard procedure in which all atoms are interrogated in an uncorrelated fashion. The scheme is identical to N independent measurements of Φ_{LO} , and therefore the ADEV is limited by the standard quantum limit: $\sigma_y \sim \frac{1}{\omega_0 \tau \sqrt{N}}$ for $\tau < \gamma_{\text{LO}}^{-1}$. Since

the Ramsey time is limited by the LO noise to $T < \gamma_{\text{LO}}^{-1}$ due to uncorrected phase wraps, this naive protocol fails to achieve the fundamental bound Eq. (5) in the long time limit $\tau > \gamma_{\text{LO}}^{-1}$, and we find the suboptimal ADEV $\sigma_y(\tau) \sim \frac{1}{\omega_0} \sqrt{\frac{\gamma_{\text{LO}}}{\tau N}}$. Second, we discuss the recently published classical protocol which interrogates the LO with uncorrelated atoms for exponentially increasing Ramsey times in each cycle [17, 18]. This protocol can be understood as the classical part of the cascaded interrogation proposed here ($j \leq 0$). It eliminates the constraint of the LO linewidth, and allows to extend the interrogation time T to its maximum value, enabling a linear scaling with τ up to the point where the fundamental bound (5) is reached. However, using an uncorrelated interrogation, the scheme displays a standard-quantum-limited scaling (i.e. $\propto 1/\sqrt{N}$), for short averaging times.

The above analysis illustrates the quantum gain of the proposed clock operation protocol using cascaded GHZ states. As compared to the best known classical scheme, our scheme provides a $\sqrt{N/\log(N)}$ enhancement for short averaging times. As a result it reaches the fundamental noise floor for laser stability in the presence of single particle decoherence [Eq. (5)] $\sim N/\log(N)$ times faster. In the limit of $N \rightarrow \infty$, our scheme attains this optimal stability allowed by quantum mechanics for all values of τ . This results identifies the possible advantage of using entanglement previously debated in the literature [9–14, 16, 31]: While the long term limitation may be set by atomic decoherence, entangled atoms reaches this limit faster thus improving the bandwidth of the stable oscillator. Our results motivate the development of atomic clocks based on entangled ions and neutral atoms. Furthermore this motivates and lays the foundations for a network of quantum clocks which operates by interrogating entangled states of all atoms in the network collectively, therefore achieving a stability set by the Heisenberg limit of all atoms [32]. Such a network can be used to construct a real-time world clock. Modifications of the scheme, such as employing optimized numbers of copies on each level of the cascade, or conditional rotations of the measurement basis, might allow to overcome the logarithmic correction in the achievable stability, and are subject to future investigations.

We are grateful to Till Rosenband and Johannes Borregaard for enlightening discussions. This work was supported by NSF, CUA, ITAMP, HQOC, JILA PFC, NIST, DARPA QUSAR, the Alfred P. Sloan Foundation, the Quiness programs, ARO MURI, and the ERC grant QIOS (grant no. 306576); MB acknowledges support from NDSEG and NSF GRFP.

SUPPLEMENTARY INFORMATION

Appendix A: Figure of merit: Allan-variance

Provided N qubits, we aim to devise an efficient interrogation scheme that provides input for the feedback

mechanism, using Ramsey spectroscopy. After the k th Ramsey cycle, of length T , an estimate $\Phi_{\text{LO}}^{\text{est}}(t_k)$ is obtained for the accumulated phase of the LO, $\Phi_{\text{LO}}(t_k) = \int_{t_k-T}^{t_k} dt \delta\omega(t)$, ($t_k = kT$, $k = 1, 2, \dots$, and $\delta\omega(t) = \omega(t) - \omega_0$), which differs from the real value by $\Delta\Phi_{\text{LO}}(t_k) = \Phi_{\text{LO}}^{\text{est}}(t_k) - \Phi_{\text{LO}}^{\text{real}}(t_k)$. Using the obtained estimate, the feedback mechanism corrects the phase or frequency of the LO after every cycle, and thus creates a LO signal with better stability. The figure of merit for stability is the Allan-variance, $\sigma_y^2(\tau) = \frac{1}{\omega_0^2} \langle \delta\bar{\omega}^2(t_0) \rangle_{t_0}$ where $\delta\bar{\omega}(t_0)$ denotes the time-average of $\delta\omega(t_0 + t)$ over $t \in [0, \tau]$, where τ is the available averaging time, $\langle \cdot \rangle_{t_0}$ stands for time-average over t_0 , which is much longer than τ , ω_0 is the frequency of the chosen clock transition. Consequently, one readily shows that the Allan variance can be written as,

$$\sigma_y^2(\tau) = \frac{1}{\omega_0^2 \tau^2} \sum_{i=1}^{\tau/T} \sum_{j=1}^{\tau/T} \left\langle \Delta\Phi_{\text{LO}}(t_0 + iT) \Delta\Phi_{\text{LO}}(t_0 + jT) \right\rangle_{t_0}. \quad (\text{A1})$$

By assuming that $\Delta\Phi_{\text{LO}}$ is a stationary random process, we substitute the average over t_0 with the average over many realizations. With the notation $\Delta\Phi_{\text{LO}}(t_0 + jT) = \Delta\Phi_{\text{LO},j}$, we can write this average as

$$\langle \Delta\Phi_{\text{LO},i} \Delta\Phi_{\text{LO},j} \rangle \approx \langle \Delta\Phi_{\text{LO}}^2 \rangle \delta_{ij}, \quad (\text{A2})$$

where we further used a white noise assumption, such that phase accumulations in consecutive Ramsey cycle are approximately uncorrelated. Also for realistic $1/f$ laser noise spectra, numerical studies show that this factorization assumption leads to only negligible corrections. Earlier results show that this is the case for initial LO frequency noise spectra, $S_\nu(f)$ that are less divergent than $1/f^2$ at low frequencies [24]. As a result, the Allan-variance simplifies to,

$$\sigma_y^2(\tau) = \frac{1}{\omega_0^2 \tau T} \langle \Delta\Phi_{\text{LO}}^2 \rangle, \quad (\text{A3})$$

linking the achieving stability directly to the frequency measurement uncertainty during the interrogation. Eq. (A3) serves as our starting point in finding the optimal measurement protocol that minimizes $\sigma_y^2(\tau)$ for fixed N and τ . In the following, we investigate and compare different classical and quantum mechanical strategies for the interrogation of the (from cycle to cycle fluctuating) quantity Φ_{LO} , and we demonstrate that the proposed interrogation protocol using cascaded GHZ states is optimal up to a logarithmic correction.

Appendix B: Single-step Uncorrelated ensemble

First, we consider the case of naive interrogation using a Ramsey protocol with N uncorrelated atoms.

1. Sub-ensembles and projection noise

Single ensemble Ramsey spectroscopy is limited to estimating either the real or the imaginary part of $e^{i\Phi_{\text{LO}}}$. However, by dividing the available qubits into two sub-ensembles, X and Y , preparing their individual qubits in different states,

$$X : \quad [|0\rangle + |1\rangle]/\sqrt{2}, \quad (\text{B1})$$

$$Y : \quad [|0\rangle + i|1\rangle]/\sqrt{2}, \quad (\text{B2})$$

and performing the same Ramsey measurement on them, we can get estimates on both the real and imaginary parts of $e^{i\Phi_{\text{LO}}}$ and deduce the value of Φ_{LO} up to 2π shifts, instead of π . At the end of the free evolution time, each qubit in ensemble X (Y) is measured in the x -basis ($|\pm\rangle = [|0\rangle \pm |1\rangle]/\sqrt{2}$) and yields the '+' outcome with probability $p_x = [1 - \cos \Phi_{\text{LO}}]/2$ ($p_y = [1 - \sin \Phi_{\text{LO}}]/2$).

After performing the measurement with N total qubits, we obtain $\Phi_{\text{LO}}^{\text{est}}$ from the estimates of p_x and p_y . Since both provide information on $\Phi_{\text{LO}}^{\text{est}}$ equivalent of $N/2$ measurement bits, this results in a total information of N measurement bits, which gives an uncertainty of

$$\langle \Delta\Phi_{\text{LO}}^2 \rangle_{\text{pr}} = \frac{1}{N}, \quad (\text{B3})$$

up to 2π phase shifts, that are fundamentally undetectable. This method is identical to the one described in [17].

2. Effects of laser fluctuations: Phase slip errors

Random fluctuations in the laser frequency (characterized by the laser spectrum noise spectrum $S_\nu(f) = 2\gamma_{\text{LO}}/f$) result in the fact that the laser phase itself has to be considered as a random variable after each cycle. Whenever in a given cycle the phase $\Phi_{\text{LO}}(t_k)$ falls outside the interval $[-\pi, \pi]$, the aforementioned technique leads to an estimate deviating from the true value by $\sim 2\pi$. As the variance s^2 of the prior distribution of Φ_{LO} grows with the interrogation time T (one finds $s^2 = \gamma_{\text{LO}}T$ ($s^2 = (\gamma_{\text{LO}}T)^2$) for a white ($1/f$) noise frequency spectrum, where γ_{LO} denotes the laser linewidth of the free-running (non-stabilized) LO) these undetected *phase slips* pose a fundamental limitation on the allowed Ramsey time T , and thus on the overall achievable laser stability.

If we assume a constant rate of phase diffusion, resulting in a Gaussian prior distribution of Φ_{LO} , the probability of a phase slip single cycle of length T can be

estimated as

$$\mathcal{P}_{\text{slip}} = 2 \int_{\pi}^{\infty} d\Phi_{\text{LO}}^{\text{real}} \frac{1}{\sqrt{2\pi s^2}} \exp \left[-\frac{(\Phi_{\text{LO}}^{\text{real}})^2}{2s^2} \right] \quad (\text{B4})$$

$$\begin{aligned} &= 1 - \text{erf} \left(\frac{\pi}{\sqrt{2}s} \right) \\ &= \left(\frac{\sqrt{2}s}{\pi^{3/2}} + \mathcal{O}(s^2) \right) \exp \left[-\frac{\pi^2}{2s^2} \right], \quad (\text{B5}) \end{aligned}$$

where erf denotes the error function. As a phase slip in an early Ramsey cycle will remain undetected in the following cycles, its error contribution will accumulate over the total averaging time τ , in the worst case by a factor τ/T . Using this upper bound, and assuming $\mathcal{P}_{\text{slip}} \ll 1$ we write the variance contribution of phase slips as

$$\begin{aligned} \langle \Delta \Phi_{\text{LO}}^2 \rangle_{\text{slip}} &= (2\pi)^2 \frac{\tau}{T} \mathcal{P}_{\text{slip}} \quad (\text{B6}) \\ &\approx (2\pi)^2 \frac{\tau}{T} \frac{\sqrt{2}}{\pi^{3/2}} \sqrt{\gamma_{\text{LO}} T} \exp \left[-\frac{\pi^2}{2\gamma_{\text{LO}} T} \right], \end{aligned}$$

where the $(2\pi)^2$ prefactor sets the absolute contribution of a manifested slip event to $\pm 2\pi$, and in the second step we approximated $\mathcal{P}_{\text{slip}}$ with the first term of its asymptotic series from Eq. (B5).

3. Optimal Ramsey time

While (A3) suggests increasing Ramsey times improve the laser stability, we have seen in the previous section that they also lead to an increased occurrence of phase slips yielding a significant contribution to the measurement uncertainty.

In order to find the optimal Ramsey time we add the contributions from quantum projection noise [Eq. (B3)] and phase slip noise [Eq. (B6)] under the assumption that the probability of phase slips is small, and obtain an expression for the Allan-variance,

$$\sigma_y^2(\tau) = \frac{1}{\omega_0^2 \tau} \Gamma, \quad (\text{B7})$$

where

$$\Gamma = \frac{1}{TN} + \sqrt{32\pi} \frac{\tau \gamma_{\text{LO}}^{1/2}}{T^{3/2}} \exp \left[-\frac{\pi^2}{2\gamma_{\text{LO}} T} \right]. \quad (\text{B8})$$

In order to find the optimal Ramsey time T_{opt} , that minimizes Γ , we introduce the new variable $x = \frac{2}{\pi^2} \gamma_{\text{LO}} T$, and write

$$\Gamma = \frac{2}{\pi^2} \frac{\gamma_{\text{LO}}}{Nx} + \frac{16}{\pi^{5/2}} \frac{\tau \gamma_{\text{LO}}^2}{x^{3/2}} e^{-1/x}. \quad (\text{B9})$$

Taking the derivative with respect to x results in

$$\frac{d}{dx} \Gamma = -\frac{2}{\pi^2} \frac{\gamma_{\text{LO}}}{Nx^2} + \frac{16}{\pi^{5/2}} \tau \gamma_{\text{LO}}^2 \left(-\frac{3}{2} \frac{1}{x^{3/2}} + \frac{1}{x^{7/2}} \right) e^{-1/x}, \quad (\text{B10})$$

which, after using the (self-consistent) assumption $x_{\text{opt}} \ll 1$, results in the following transcendental equation for x_{opt} ,

$$x_{\text{opt}}^{3/2} = ((8/\sqrt{\pi}) \gamma_{\text{LO}} \tau N) e^{-1/x_{\text{opt}}}. \quad (\text{B11})$$

Below, we provide the derivation of the asymptotic solution,

$$x_{\text{opt}} = [\log((8/\sqrt{\pi}) \gamma_{\text{LO}} \tau N)]^{-1} \approx [\log(\gamma_{\text{LO}} \tau N)]^{-1} \quad (\text{B12})$$

yielding directly

$$T_{\text{opt}} \approx \gamma_{\text{LO}}^{-1} \frac{\pi^2}{2} [\log(\gamma_{\text{LO}} \tau N)]^{-1}. \quad (\text{B13})$$

Self-consistently we confirm that already for values $\gamma_{\text{LO}} \tau N \geq 10^4$, the approximation in Eq. (B6) is well justified, so that the above value represents a true local minimum. For larger values of T the phase slip errors grow rapidly, and numerical studies confirm that Eq. (B13) indeed represents a global minimum.

The optimal interrogation time is mainly set by the LO coherence time γ_{LO}^{-1} , and shows a weak dependence on the total number of qubits N , and the averaging time τ (Note, that if we model the LO with a $1/f$ frequency noise spectrum, only the exponent of the log term changes to $-1/2$ in this result). Using this optimized Ramsey time we find for the minimal value of Γ is

$$\Gamma_{\text{min}} = \frac{2}{\pi^2} \frac{\gamma_{\text{LO}}}{Nx_{\text{opt}}} + \frac{16}{\pi^{5/2}} \frac{\tau \gamma_{\text{LO}}^2}{x_{\text{opt}}^{3/2}} e^{-1/x_{\text{opt}}} \quad (\text{B14})$$

$$= \frac{2}{\pi^2} \frac{\gamma_{\text{LO}}}{N} \left(\frac{1}{x_{\text{opt}}} + 1 \right) \quad (\text{B15})$$

$$\approx \frac{2}{\pi^2} \frac{\gamma_{\text{LO}} \log(\gamma_{\text{LO}} \tau N)}{N}. \quad (\text{B16})$$

This result is valid as long as the averaging time τ is longer than the proposed T_{opt} from Eq. (B13). If this is not the case, then $T_{\text{opt}} = \tau$, the phase slip noise becomes negligible, and we end up with

$$\Gamma_{\text{min}} = \frac{1}{\tau N}. \quad (\text{B17})$$

We approximate the crossover region (around $\tau \sim \gamma_{\text{LO}}^{-1}$) by adding leading terms from Eq. (B16) & (B17) and obtain

$$[\sigma_y(\tau)]_{\text{min}} \approx \frac{1}{\omega_0 \sqrt{N} \tau} \sqrt{\frac{1}{\tau} + \frac{2}{\pi^2} \gamma_{\text{LO}} \log(\gamma_{\text{LO}} \tau N)}. \quad (\text{B18})$$

In summary, in the region $\tau < T_{\text{opt}}$, the LO noise is negligible leading to a linear scaling of the ADEV with the total averaging time τ . For large averaging times $\tau > T_{\text{opt}}$, phase slips of the laser phase pose a limitation to the maximal possible Ramsey time which results in a $1/\sqrt{\tau}$ scaling of the laser stability. Since we employ uncorrelated atoms, the ADEV displays in both regimes the $1/\sqrt{N}$ scaling of the standard quantum limit (SQL). As modern atomic clocks typically are laser noise limited, $\gamma_{\text{LO}} \gg \gamma_{\text{ind}}$ (where γ_{ind} represents the clock atom linewidth), we neglected the effects of individual atomic dephasing in the above considerations.

Appendix C: Cascaded interrogation using GHZ states

In this Section, we discuss the possibility of using quantum correlated states, namely GHZ states of the form

$$[|\mathbf{0}\rangle + e^{i\chi}|\mathbf{1}\rangle]/\sqrt{2}, \quad (\text{C1})$$

where $|\mathbf{0}\rangle$ and $|\mathbf{1}\rangle$ are product states of all qubits being in $|0\rangle$ or $|1\rangle$, respectively, and χ will be referred to as the phase of the GHZ state. Such a state, once prepared, is more sensitive to the accumulated phase of the LO, Φ_{LO} , by a factor of N' , the number of qubits entangled:

$$\left(\prod_{j=1}^N \hat{U}_j \right) [|\mathbf{0}\rangle + e^{i\chi}|\mathbf{1}\rangle] / \sqrt{2} = [|\mathbf{0}\rangle + e^{i(\chi + N'\Phi_{\text{LO}})}|\mathbf{1}\rangle] / \sqrt{2}, \quad (\text{C2})$$

where $\hat{U}_j = |0\rangle\langle 0| + e^{i\Phi_{\text{LO}}}|1\rangle\langle 1|$ is the time propagation operator for the interrogation time acting on the j th qubit. This property promises an enhancement in phase resolution, and therefore a better stability for quantum clocks.

1. Parity measurement

Using the idea with the two sub-ensembles [see Eq. (B1) and (B2)], we imagine dividing the qubits into two equal groups, and preparing two separate GHZ states:

$$|X\rangle := [|\mathbf{0}\rangle + |\mathbf{1}\rangle]/\sqrt{2}, \quad (\text{C3})$$

$$|Y\rangle := [|\mathbf{0}\rangle + i|\mathbf{1}\rangle]/\sqrt{2}, \quad (\text{C4})$$

each entangling N' qubits. After the free evolution time, we imagine measuring each qubits in the x -basis ($|\pm\rangle = [|\mathbf{0}\rangle \mp |\mathbf{1}\rangle]/\sqrt{2}$) separately. In this basis, the above states are written as

$$\left[\left(\frac{|+\rangle - |-\rangle}{\sqrt{2}} \right)^{\otimes N'} + e^{i\phi_\nu} \left(\frac{|+\rangle + |-\rangle}{\sqrt{2}} \right)^{\otimes N'} \right] / \sqrt{2}, \quad (\text{C5})$$

where $\phi_\nu = \chi_\nu + N'\Phi_{\text{LO}}$, for $\nu \in \{x, y\}$ and $\chi_x = 0$, while $\chi_y = \pi/2$ for the two groups, respectively. The above state can be written as

$$\frac{1}{2^{(N'+1)/2}} \sum_{\mathbf{q} \in \{+, -\}^{\times N'}} \left[\left(\prod_{j=1}^{N'} q_j \right) + e^{i\phi_\nu} \right] |q_1, q_2, \dots, q_{N'}\rangle. \quad (\text{C6})$$

Once the qubits are measured one by one, the probability to measure a certain outcome $\mathbf{q} = (q_1, q_2, \dots, q_{N'})$, ($q_j \in \{+, -\}$) is

$$\mathcal{P}(\mathbf{q}) = \frac{1}{2^{N'+1}} |1 + p(\mathbf{q})e^{i\phi_\nu}|^2, \quad (\text{C7})$$

where $p(\mathbf{q}) = \prod_{j=1}^{N'} q_j$ is the parity of the sum of all measurement bits. This parity is the observable that is

sensitive to the accumulated phase, since its distribution is

$$\mathcal{P}(p = \pm 1) = \frac{1 \pm \cos(\phi_\nu)}{2}. \quad (\text{C8})$$

This is identical to the parity measurement scheme described in [15]. By interrogating $n_0/2$ instances of $|X\rangle$ and $|Y\rangle$, respectively, we can measure the phase of the GHZ state, $N'\Phi_{\text{LO}}$, with uncertainty $1/\sqrt{n_0}$, since each instance provides a single measurement bit, which can be combined the same way as we described in the case of uncorrelated ensembles. The resulting measurement uncertainty, $\Delta\Phi_{\text{LO}}$, is

$$\langle \Delta\Phi_{\text{LO}}^2 \rangle_{\text{pr}} = \frac{1}{(N')^2 n_0} = \frac{n_0}{N^2}, \quad (\text{C9})$$

which is a factor of N/n_0 smaller than the variance contribution of projection noise for the uncorrelated ensemble protocol, ($N = n_0 N'$).

2. Failure of the maximally entangled GHZ

Motivated by the increased phase resolution provided by the interrogation of GHZ states, we evaluate the stability of such a protocol. We find that it fails to provide improvement compared to the single-step uncorrelated protocol due to an increased phase slip rate. This is in agreement with earlier results [12, 16].

The probability, that the phase accumulated by $|X\rangle$ ($|Y\rangle$) during the interrogation time T , $N'\Phi_{\text{LO}}$ lies outside the interval $[-\pi, \pi]$, is

$$\mathcal{P}_{\text{slip}} = 2 \int_{\pi/N'}^{\infty} d\Phi_{\text{LO}}^{\text{real}} \frac{1}{\sqrt{2\pi\gamma_{\text{LO}}T}} \exp \left[-\frac{(\Phi_{\text{LO}}^{\text{real}})^2}{2\gamma_{\text{LO}}T} \right], \quad (\text{C10})$$

which, due to the much lower slipping threshold of π/N' [instead of π in the uncorrelated case, compare Eq. (B4)] will become significant for much shorter T cycle times. The resulting variance contribution (following the same argument as before) is

$$\langle \Delta\Phi_{\text{LO}}^2 \rangle_{\text{slip}} = \sqrt{32\pi} \frac{\tau}{T} \sqrt{\gamma_{\text{LO}}T} N' \exp \left[-\frac{\pi^2}{2\gamma_{\text{LO}}T(N')^2} \right] \quad (\text{C11})$$

Neglecting the individual qubit noise by the same argument as before, we simply add the contributions from Eq. (C9) and Eq. (C11) to obtain the Allan-variance, $\sigma_y^2(\tau) = \frac{1}{\omega_0^2 \tau} \Gamma$, where

$$\Gamma = \frac{1}{NN'T} + \sqrt{32\pi} \frac{\tau\gamma_{\text{LO}}^{1/2}}{T^{3/2}} N' \exp \left[-\frac{\pi^2}{2\gamma_{\text{LO}}T(N')^2} \right]. \quad (\text{C12})$$

After optimizing T , we find

$$T_{\text{opt}} \approx \frac{\pi^2}{2} \frac{1}{\gamma_{\text{LO}}(N')^2} \frac{1}{\log[\gamma_{\text{LO}}\tau N(N')^3]}, \quad (\text{C13})$$

which results in the minimal Allan-variance,

$$[\sigma_y(\tau)]_{\min} \approx \frac{1}{\omega_0} \frac{\sqrt{2}}{\pi} \sqrt{\frac{\gamma_{\text{LO}} N' \log[\gamma_{\text{LO}} \tau N (N')^3]}{\tau N}}, \quad (\text{C14})$$

which is at least a factor of $\sqrt{N'}$ bigger than the smallest obtainable Allan-variance with the single-step uncorrelated protocol [Eq. (B16)]. In case of a $1/f$ LO frequency noise spectrum, $T_{\text{opt}} \propto \frac{1}{N}$ (up to logarithmic terms), and the resulting Allan-variance is equal to Eq. (B16), up to logarithmic corrections, yielding effectively no advantage over the uncorrelated scheme.

3. Cascaded GHZ scheme

As demonstrated in the previous Section, single GHZ states fail to improve clock stability because the increase in sensitivity to the laser detuning, at the same time, leads to a drastic increase of phase slip errors originating from laser frequency fluctuations. These fluctuations, however, affect all clock qubits in identical manner, and therefore represents a *collective noise*. As such (and unlike, e.g., the individual dephasing of the clock qubits), they do not represent a fundamental limitation for the phase estimation. In the following, we show that this problem can be efficiently addressed using a cascade of GHZ states of varying sizes (and classical states with varying interrogation times) in an incoherent version of the phase estimation algorithm [19]. To this end, we reformulate the problem of estimating Φ_{LO} in a more suitable language.

The laser phase after a given Ramsey cycle can be expressed in a base- D numeral system as

$$(\Phi_{\text{LO}} + \pi)/2\pi = \sum_{j=-\infty}^{\infty} Z_j/D^j, \quad (\text{C15})$$

with base- D digits $Z_j \in \{0, 1, \dots, D-1\}$. Let us for the moment assume that the laser phase $\Phi_{\text{LO}} \in [-\pi, \pi]$, such that $(\Phi_{\text{LO}} + \pi)/2\pi = \sum_{j=1}^{\infty} Z_j/D^j \equiv 0.Z_1Z_2Z_3\dots$

Provided with N qubits, we imagine dividing them into M different groups, the j th group ($j = 0, 1, \dots, M-1$) contains n_0 instances of GHZ states with D^j number of entangled qubits. One readily shows that a GHZ state consisting of D^{M-1} particles picks up the phase

$$\Phi_{M-1} = D^{M-1} \Phi_{\text{LO}} \pmod{[-\pi, \pi]} \quad (\text{C16})$$

$$= 2\pi(0.Z_M Z_{M+1} Z_{M+2} \dots) - \pi, \quad (\text{C17})$$

which depend only on digits Z_M and higher of the laser phase to be measured. This insensitivity of the GHZ state with regard to the lower digits Z_1 to Z_{M-1} restates the problems of phase slips. Only if the latter are known, a measurement of the phase of the GHZ state Φ_{M-1} yields useful information to determine Φ_{LO} . In other words, the natural number $Z_1 Z_2 \dots Z_{M-1}$ represents the number of phase slips of the largest group

of GHZ states ($j = M-1$). These lower digits can be determined one by one from the accumulated phases $\Phi_j = D^j \Phi_{\text{LO}} \pmod{[-\pi, \pi]}$ of the smaller GHZ ensembles $j = 0, \dots, M-2$ by using the relation

$$\begin{aligned} & [D(\Phi_{j-1} + \pi) - (\Phi_j + \pi)]/(2\pi) \quad (\text{C18}) \\ & = (Z_j.Z_{j+1}Z_{j+2}\dots) - (0.Z_{j+1}Z_{j+2}\dots) = Z_j. \end{aligned}$$

Combining all measurement results we find that the best estimate for Φ_{LO} is given by

$$\Phi_{\text{LO}}^{\text{est}} = 2\pi \sum_{j=1}^{M-1} Z_j^{\text{est}}/D^j + \Phi_{M-1}^{\text{est}}/D^{M-1}, \quad (\text{C19})$$

the precision of which is mostly determined by the uncertainty of the phase of the last group ($j = M-1$), which contains the GHZ states with the most entangled qubits. Since there are n_0 independent instances of these GHZ states, their phase is known up to the uncertainty, $\langle \Delta \Phi_{M-1}^2 \rangle_{\text{pr}} = \frac{1}{n_0} \approx \frac{\delta D^{M-1}}{N}$, where $\delta = \frac{D}{D-1}$, and therefore we find

$$\langle \Delta \Phi_{\text{LO}}^2 \rangle_{\text{pr}} = \frac{\langle \Delta \Phi_{M-1}^2 \rangle_{\text{pr}}}{D^{2(M-1)}} = \frac{n_0 \delta^2}{N^2}. \quad (\text{C20})$$

This would be the total uncertainty if we could tell with certainty that all phase slips of the lower levels had been detected correctly. However, the occurrence of an error in the estimation of any Z_j (in the following referred to as *rounding error*) has non-zero probability.

4. Rounding errors: finding the optimal n_0

If Φ_j is determined with poor precision, the estimate of Z_{j+1} will have a significant uncertainty, causing the final estimate of Φ_{LO} to be uncertain as well. Whenever $|\Phi_j^{\text{est}} - \Phi_j^{\text{real}}| > \pi/D$, we make a mistake by under- or overestimating the digit Z_{j+1} . To minimize the effect of this error, we need to optimize how the qubits are distributed on various levels of the cascade. In other words, for a given total particle number N and basis D we need to find the optimal value of n_0 , the number of copies of GHZ states in each step [33].

The probability that a rounding error occurs during the estimation of Z_j is

$$\begin{aligned} \mathcal{P}_{\text{re}} &= 2 \int_{\pi/D}^{\infty} d\phi \rho(\phi - \Phi_j^{\text{real}}) \leq 2 \int_{\pi/D}^{\infty} d\phi n_0^{3/2} \exp\left[-\frac{n_0 \phi^2}{2}\right] \\ &\approx \frac{2}{\pi} n_0^{1/2} D \exp\left[-\frac{n_0 \pi^2}{2D^2}\right], \quad (\text{C21}) \end{aligned}$$

where $\phi = \Phi_{j-1}^{\text{est}} - \Phi_{j-1}^{\text{real}}$, and ρ is the conditional distribution of Φ_{j-1}^{est} for given Φ_{j-1}^{real} . The employed upper bound is obtained in the last section, with the assumption $\gamma_{\text{LO}}/\gamma_{\text{ind}} \gg N/n_0$ (γ_{ind} is the individual qubit dephasing rate), so that the projection noise is the dominant

noise term. Due to the fixed value of n_0 across different levels of the cascade, this probability is independent of j , however the phase shift imposed on Φ_{LO} by a manifested rounding error of Z_j is $2\pi D^{-j}$ ($j = 1, \dots, M-1$), as rounding errors early in the cascade are more harmful than later ones. This results in the total variance contribution,

$$\langle \Delta \Phi_{\text{LO}}^2 \rangle_{\text{re}} = \mathcal{P}_{\text{re}} \sum_{j=1}^{M-1} (2\pi D^{-j})^2 \quad (\text{C22})$$

$$\approx 8\pi \sqrt{\frac{N}{\delta}} D^{-\frac{M-3}{2}} \exp\left[-\frac{n_0 \pi^2}{2D^2}\right] \frac{1}{D^2 - 1}. \quad (\text{C23})$$

By adding the two error contributions from Eq. (C20) and Eq. (C22), we obtain the total uncertainty, $\langle \Delta \Phi_{\text{LO}}^2 \rangle$ and the corresponding Allan-variance [according to Eq. (A3)]

$$\begin{aligned} \sigma_y^2(\tau) &= \frac{1}{\omega_0^2 \tau} \left[\frac{\delta}{NTD^{M-1}} \right. \\ &\quad \left. + \frac{8\pi}{T} \sqrt{\frac{N}{\delta}} D^{-\frac{M-3}{2}} \exp\left[-\frac{n_0 \pi^2}{2D^2}\right] \frac{1}{D^2 - 1} \right] \\ &=: \frac{1}{\omega_0^2 \tau} [\Gamma_1 + \Gamma_2] \end{aligned} \quad (\text{C24})$$

We find the optimal value of n_0 by minimizing this quantity. Introducing the new variable $x \equiv \frac{2D^2}{n_0 \pi^2}$, and using $n_0 \approx N/(\delta D^{M-1})$ we write

$$\Gamma_1 + \Gamma_2 = \frac{2}{\pi^2} \frac{\delta^2 D^2}{N^2 T x} + \frac{\sqrt{128} D^2}{T(D^2 - 1)} \frac{1}{x^{1/2}} \exp\left[-\frac{1}{x}\right] \quad (\text{C25})$$

Taking the derivative with respect to x and equating it with 0, while using the (self-consistent) assumption $x_{\text{opt}} \ll 1$, results in the condition $\Gamma_2 \approx x_{\text{opt}} \Gamma_1 \ll \Gamma_1$ and the transcendental equation

$$x_{\text{opt}}^{1/2} \approx \frac{\sqrt{32} \pi^2 N^2}{\delta^2 (D^2 - 1)} \exp\left[-\frac{1}{x_{\text{opt}}}\right] \quad (\text{C26})$$

for x_{opt} . The asymptotic solution in the case of $x_{\text{opt}} \ll 1$ is

$$x_{\text{opt}} \approx \left[\log\left(\frac{\sqrt{32} \pi^2 N^2}{\delta^2 (D^2 - 1)}\right) \right]^{-1} \sim [\log(N^2)]^{-1} \quad (\text{C27})$$

yielding directly the optimal number of instances of GHZ states per level

$$n_0^{\text{opt}} \sim \frac{2}{\pi^2} D^2 \log(N^2). \quad (\text{C28})$$

This choice guarantees rounding errors yield a negligible contribution to the total measurement uncertainty, and we find for the corresponding value of $\Gamma_1 + \Gamma_2$

$$\begin{aligned} [\Gamma_1 + \Gamma_2]_{\text{min}} &\approx \Gamma_1(x_{\text{opt}}) \\ &= \frac{n_0^{\text{opt}} \delta^2}{N^2 T} \sim \frac{2}{\pi^2} \frac{\delta^2 D^2}{N^2 T} \log(N^2), \end{aligned} \quad (\text{C29})$$

where the factor $\delta = D/(D-1) \in (1, 2]$. Obviously, the use of a binary basis ($D=2$) is optimal, and the effect of rounding errors lead to a logarithmic correction to the Heisenberg limit.

5. Phase slip errors: limitations to the Ramsey time T from laser noise

Although the cascade is designed to detect phase slips at levels $j = 1, 2, \dots, M-1$, when we relax the condition $\Phi_{\text{LO}} \in [-\pi, \pi]$, and allow $\Phi_{\text{LO}} \in (-\infty, +\infty)$, the possible phase slips of level $j=0$ (Z_0) remains undetected. Once this happens, it introduces a 2π phase shift in Φ_{LO} , and therefore contributes to its overall uncertainty with

$$\begin{aligned} \langle \Delta \Phi_{\text{LO}}^2 \rangle_{\text{slip}} &= (2\pi)^2 \frac{\tau}{T} \mathcal{P}_{\text{slip}} = \\ &= \sqrt{32} \pi \frac{\tau \gamma_{\text{LO}}^{1/2}}{T^{1/2}} \exp\left[-\frac{\pi^2}{2\gamma_{\text{LO}} T}\right], \end{aligned} \quad (\text{C30})$$

where we assumed $\gamma_{\text{LO}} T \ll 1$. This adds an extra noise term $\Gamma_3 := \langle \Delta \Phi_{\text{LO}}^2 \rangle_{\text{slip}}/T$ to the already optimized $[\Gamma_1 + \Gamma_2]_{\text{min}}$ expression, yielding

$$\begin{aligned} [\Gamma_1 + \Gamma_2]_{\text{min}} + \Gamma_3 &= \\ &= \frac{2}{\pi^2} \frac{\delta^2 n_0^{\text{opt}}}{N^2 y} + \frac{16}{\pi^{5/2}} \tau \gamma_{\text{LO}}^2 \frac{1}{y^{3/2}} \exp\left[-\frac{1}{y}\right], \end{aligned} \quad (\text{C31})$$

where $y = \frac{2}{\pi^2} \gamma_{\text{LO}} T$. After taking the derivative with respect to y and equating it with zero, the assumption $y_{\text{opt}} \ll 1$ results in the condition $\Gamma_3 \approx y_{\text{opt}} [\Gamma_1 + \Gamma_2]_{\text{min}} \ll [\Gamma_1 + \Gamma_2]_{\text{min}}$ and the following transcendental equation,

$$y_{\text{opt}}^{3/2} \approx \frac{8 \gamma_{\text{LO}} \tau N^2}{\sqrt{\pi} \delta^2 n_0^{\text{opt}}} \exp\left[-\frac{1}{y_{\text{opt}}}\right], \quad (\text{C32})$$

for y_{opt} . The asymptotic solution is

$$y_{\text{opt}} = \left[\log\left(\frac{8 \gamma_{\text{LO}} \tau N^2}{\sqrt{\pi} \delta^2 n_0^{\text{opt}}}\right) \right]^{-1} \sim [\log(\gamma_{\text{LO}} \tau N^2)]^{-1} \quad (\text{C33})$$

$$T_{\text{opt}} = \frac{\pi^2}{2} \frac{y_{\text{opt}}}{\gamma_{\text{LO}}} \sim \frac{\pi^2}{2} \frac{[\log(\gamma_{\text{LO}} \tau N^2)]^{-1}}{\gamma_{\text{LO}}} \quad (\text{C34})$$

in the realistic limit of $\gamma_{\text{LO}} \tau N^2 \gg 1$. The corresponding minimal value of $\Gamma_1 + \Gamma_2 + \Gamma_3$ is

$$\left[[\Gamma_1 + \Gamma_2]_{\text{min}} + \Gamma_3 \right]_{\text{min}} \approx \Gamma_1(x_{\text{opt}}, y_{\text{opt}}) = \frac{n_0^{\text{opt}} \delta^2}{N^2 T_{\text{opt}}} \quad (\text{C35})$$

$$\sim \gamma_{\text{LO}} \frac{4\delta^2 D^2}{\pi^4} \frac{\log(\gamma_{\text{LO}} \tau N^2) \log(N^2)}{N^2}. \quad (\text{C36})$$

Since Γ_3 grows exponentially with T , interrogation times exceeding T_{opt} drastically reduce the resulting laser stability. For averaging times $\tau > T_{\text{opt}}$ this limit on the maximal interrogation time imposed by phase slip errors, leads to sub-optimal values of the ADEV $\sigma_y(\tau) \propto 1/\sqrt{\tau}$

according to Eq. (B7) & (C36). However, this limitation can be overcome as we demonstrate in the following section.

If the averaging time τ is shorter than the interrogation time suggested by Eq. (C34), Γ_3 is negligible compared to Γ_1 , and the corresponding effective linewidth is

$$[\Gamma_1 + \Gamma_2]_{\min} + \Gamma_3 \approx \frac{n_0^{\text{opt}} \delta^2}{N^2 T} = \frac{2}{\pi^2} \frac{\delta^2 D^2}{N^2 T} \log(N^2), \quad (\text{C37})$$

and the real optimum is at $T = \tau$. The resulting τ^{-1} scaling indicates that this is the noise-free measurement regime, and results in a Heisenberg-limited ADEV (up to the logarithmic correction arising from n_0^{opt}).

6. Extending the Ramsey time beyond the laser noise limit

As we have seen in the previous Section, for long $\tau > T_{\text{opt}}$, the cascaded GHZ scheme is limited by the LO linewidth γ_{LO} yielding a sub-optimal laser stability [Eq. (C36)]. In the following we demonstrate a method to efficiently circumvent this problem, by employing additional classical interrogations with varying (effective) interrogation times [17, 18]. This allows us to directly assess the digits $Z_0, Z_{-1}, Z_{-2} \dots$, thus effectively countering the problem of phase slips on this level. As such it represents a direct extension of the cascaded GHZ states scheme in the classical domain.

We assume we have additional M^* groups of n_0^* particles at our disposal. Using dynamical decoupling techniques [21], we realize that each ensemble ($j = -1, -2 \dots, -M^*$) during the interrogation picks up a phase $\Phi_j = D^j \Phi_{\text{LO}} \bmod [-\pi, \pi]$ (alternatively, this can be achieved by choosing varying interrogation times for each ensemble according to $T_j = D^j T$, for $j < 0$ [17]). This implies that these ensembles evolve successively slower for decreasing j , and thus, in the spirit of the section on cascaded GHZ, directly assess the digits left from the point in the D-numeral representation of Φ_{LO} [compare Eq. (C15)] according to Eq. (C18), where we now allow negative values of j .

If all digits are correctly estimated, this accounts for all phase slips up to the last ensemble $j = -M^*$. One readily shows in an analogous calculation to the one in the section on phase slip errors that for such a procedure with M^* classical stages the optimal Ramsey time (i.e., the optimized interrogation time of the GHZ states) is exponentially prolonged

$$T_{\text{opt}}^{(M^*)} = D^{M^*} T_{\text{opt}}. \quad (\text{C38})$$

Note, that here we assumed that the total number of particles employed in the classical part of the scheme is negligible with regard to the total number of particles, $N^* = M^* n_0^* \ll N$. This is a well justified assumption, as in order to prolong the optimal Ramsey time by a factor of k from the original optimum T_{opt} we need a logarithmic

number of groups only, $M^* \approx \log_D(k)$, as implied by Eq. (C38). Furthermore, following the argumentation outlined in the section on rounding errors, we find that only

$$n_0^* \geq \frac{2}{\pi^2} D^2 \log(kN^2), \quad (\text{C39})$$

particles per level are sufficient to ensure that the rounding errors induced by the classical part of the cascade ($j < 0$) are negligible.

As seen in the previous Section, when the optimal Ramsey time exceeds the averaging time, $T_{\text{opt}}^{(M^*)} \geq \tau$ the effective linewidth is given by Eq. (C37) (assuming $N^* \ll N$), as we can neglect the phase slips contribution to the measurement uncertainty (Γ_3). Extending the Ramsey time to its then optimal (i.e., maximal) value $T \sim \tau$ we find the ADEV [compare Eq. (B7)]

$$[\sigma_y(\tau)]_{\min} \approx \frac{1}{\omega_0} \frac{\delta \sqrt{n_0^{\text{opt}}}}{N \tau} \quad (\text{C40})$$

$$\approx \frac{\sqrt{2}}{\sqrt{\pi} \omega_0} \frac{D \delta}{N \tau} \sqrt{\log(N^2)}. \quad (\text{C41})$$

This result illustrates that the presented clock protocol achieves Heisenberg-limited clock stability up to a logarithmic correction arising from the number of atoms necessary to compensate for rounding and phase slip errors. The number of particles needed for the extension of the Ramsey time beyond the laser noise limit $T_{\text{opt}} \approx \gamma_{\text{LO}}^{-1}$ is given as $N^* \approx \frac{2}{\pi^2} D^2 \log(kN^2) \log_D(k)$ and thus negligible compared to the total particle number N . For the optimal choice of basis $D = 2$ the constant factor reduces to $D\delta = 4$.

The above procedure of interrogation with varying Ramsey times (for the groups $j < 0$) can be understood as a classical pre-narrowing of the laser linewidth [17] to a value that eliminates the threat of phase slips, before application of the quantum protocol.

7. Individual qubit noise and final result

Up to this point we have neglected individual particle dephasing. However, as we increase the Ramsey time beyond the laser noise limit $T > \gamma_{\text{LO}}^{-1}$ we need to consider their effect.

In general, the clock atoms are subject to individual decoherence processes characterized by the atomic linewidth $\gamma_{\text{ind}} (\ll \gamma_{\text{LO}})$. For the group with the largest GHZ states in our scheme this leads to an uncertainty contribution of $\langle \Delta \Phi_{M-1}^2 \rangle_{\text{dephasing}} = D^{M-1} \gamma_{\text{ind}} T / n_0$ which results in the measurement uncertainty

$$\langle \Delta \Phi_{\text{LO}}^2 \rangle_{\text{dephasing}} = \frac{\gamma_{\text{ind}} T}{n_0 D^{M-1}} \approx \frac{\delta \gamma_{\text{ind}} T}{N}, \quad (\text{C42})$$

which represents a fundamental noise floor in the form of the effective linewidth contribution $\Gamma_4 = \langle \Delta \Phi_{\text{LO}}^2 \rangle_{\text{dephasing}} / T$.

By adding Eq. (C37) and Γ_4 , we obtain an approximation of the total ADEV under single particle noise,

$$[\sigma_y(\tau)]_{\min} \approx \frac{\delta}{\omega_0 \sqrt{\tau N}} \left[\frac{1}{TD^{M-1}} + \gamma_{\text{ind}} \right]^{1/2} \quad (\text{C43})$$

where we used $n_0 = N/\delta D^{M-1}$. This equation suggests that the quantum gain in the estimation becomes lost if $T \sim (\gamma_{\text{ind}} D^{M-1})^{-1}$. This is a well known result [14], and in fact represents a fundamental limitation of the maximal Ramsey time allowed in the presence of single particle noise [30]. We approximate the crossover between the regimes $\tau < (\gamma_{\text{ind}} D^{M-1})^{-1}$ ($\tau > (\gamma_{\text{ind}} D^{M-1})^{-1}$), where Eq. (C43) is dominated by the first (second term) by taking $T = \tau$, and rewrite D^{M-1} in terms of the total particle number N to arrive at the final equation characterizing the stability of the cascaded GHZ scheme

$$[\sigma_y(\tau)]_{\min} \approx \frac{1}{\omega_0 \sqrt{\tau N}} \left[\frac{1}{\tau N} \frac{2\delta^2 D^2}{\pi^2} \log(N^2) + \delta \gamma_{\text{ind}} \right]^{1/2}. \quad (\text{C44})$$

Again, for the choice of a binary basis $D = 2$ the constant factor is given as $\delta D = 4$.

In summary, we find that the cascaded GHZ scheme enables an optimal, Heisenberg-limited laser stability for short averaging times τ up to a logarithmic correction. This stability reaches the fundamental noise floor given by the single particle dephasing for averaging times $\tau_0 \approx (\gamma_{\text{ind}} D^{M-1})^{-1}$. Note, that in the limit $N \rightarrow \infty$ this crossover value goes to zero, and the clock stability is given by the best possible stability allowed by quantum mechanics for all τ . In comparison, classical protocols reach this fundamental limit in the best case [17] at the fixed time $\tau \approx \gamma_{\text{ind}}^{-1}$. For averaging times larger than this value $\tau \geq \gamma_{\text{ind}}^{-1}$ the quantum protocol does not offer an advantage over an optimal classical protocol due to fundamental quantum metrological bounds [30].

Appendix D: Analytic solution of $x^n = A \exp[-1/x]$

To carry out direct optimization of the Allan variance, we need to be able to solve transcendental equations of the following form

$$x^n = A \exp \left[-\frac{1}{x} \right]. \quad (\text{D1})$$

In this Section we obtain an analytic formula for the solution of this equation over the domain $x \in [0, \infty)$, in the limiting case of $A \gg 1$, where n is real. The sign of n determines the number of solutions: In case of $n > 0$, there are three solutions: $x_{s,0} = 0$, $x_{s,1} \ll 1$ and $x_{s,2} \gg 1$. In case of $n \leq 0$, there is always a single solution, $x_{s,1} \ll 1$. We are going to focus on the $x_{s,1} =: x_s$ solution, and give upper and lower bounds, such that $x_l \leq x_s \leq x_u$, and $x_u/x_l \rightarrow 1$ as $A \rightarrow \infty$.

The general method of Taylor-expanding the right side of Eq. (D1) around $x = 0$ fails due to the non-analytic property of $e^{-1/x}$ function at zero, and forces us to choose an alternate route. Here, we use a recursion formula, and prove its stability around x_s . Rearranging Eq. (D1) and turning it into a recursion f yields

$$x_k = \frac{1}{\log A - n \log x_{k-1}} =: f(x_{k-1}), \quad (\text{D2})$$

The iteration of f is stable around the fixed point ($f(x_s) = x_s$), if and only if $1 > |f'(x_s)| = x_s |n|$, which is true in the limit $x_s \ll 1$. Stability implies that the fixed point can be obtained as the limit

$$x_s = \lim_{k \rightarrow \infty} x_k = \lim_{k \rightarrow \infty} f^{[k]}(x_0), \quad (\text{D3})$$

if the x_0 starting point is sufficiently close to x_s , where $f^{[k]}$ denotes k iterations of f .

In case of $n \leq 0$, $f'(x_s) = n x_s \leq 0$ therefore $[f^{[k]}(x_0) - f^{[k-1]}(x_0)]$ is an alternating sequence and we can quickly obtain upper (x_u) and lower (x_l) bounds by applying the recursion f twice on $x_0 = 1$:

$$x_l = x_1 = \frac{1}{\log A}, \quad x_u = x_2 = \frac{1}{\log A + n \log \log A}, \quad (\text{D4})$$

In case of $n > 0$ and $x_0 = 1$, $f^{[k]}(x_s)$ is monotonically decreasing (since $f'(x_s) > 0$), and we can safely choose the upper bound $x_u = x_2$. To obtain a lower bound, we introduce a new variable $\xi = -\log x$, and write Eq. (D2) as

$$\xi_k = \log \log A + \log \left(1 + \frac{n}{\log A} \xi_{k-1} \right) \quad (\text{D5})$$

$$\leq \log \log A + \frac{n}{\log A} \xi_{k-1} =: g(\xi_{k-1}), \quad (\text{D6})$$

where we used that $\log(1+y) \leq y$, $\forall y \in \mathbb{R}^+$ and g is a new recursion. Since g is a monotonically increasing function, the inequality holds for multiple iterations, $\xi_k \leq g^{[k]}(\xi_0)$, and eventually give the upper bound, $-\log x_s = \xi_s \leq \lim_{k \rightarrow \infty} g^{[k]}(\xi_0)$. In the limit of $A \ll 1$, we can assume $\frac{n}{\log A} < 1$, and the sequence of iterations of g becomes convergent. Due to its simple form, we can evaluate its limit in a closed form, which results in the following upper bound for ξ_s and the corresponding lower bound for x_s .

$$\xi_s < \frac{\log \log A}{1 - \frac{n}{\log A}}, \quad \rightarrow \quad x_s > \left(\frac{1}{\log A} \right)^{\frac{1}{1 - \frac{n}{\log A}}}. \quad (\text{D7})$$

We can obtain an even better (and more conventional) lower bound by applying f once more:

$$x_l = f \left[\left(\frac{1}{\log A} \right)^{\frac{1}{1 - \frac{n}{\log A}}} \right] = \frac{1}{\log A + \frac{1}{1 - \frac{n}{\log A}} n \log \log A} \quad (\text{D8})$$

For both signs of n , $x_l \log A \rightarrow 1$, $x_u \log A \rightarrow 1$, and $x_u/x_l \rightarrow 1$ as $A \rightarrow \infty$, from which we conclude that

$$\lim_{A \rightarrow \infty} (x_s \log A) = 1. \quad (\text{D9})$$

For large enough A , we can approximate x_s with $[\log A]^{-1}$, and the relative error is bounded by $|n| \frac{\log \log A}{\log A}$.

Appendix E: Upper bound on the tail of the distribution of the estimated phase

The probability of rounding errors is given an upper bound in order to obtain a more tractable form for optimization.

1. Upper bound on the tail of the binomial distribution

Here we derive an upper bound for the binomial distribution

$$\mathcal{P}(k) = \binom{n}{k} p^k (1-p)^{n-k}. \quad (\text{E1})$$

Central limit theorem implies that for large enough n , $\mathcal{P}(k)$ can be approximated with the normal distribution, $\mathcal{N}(np, np(1-p))$, however, one can be concerned with the fact that this underestimates the tail of $\mathcal{P}(k)$. Here we give $F(k)$ as a strict upper bound on $\mathcal{P}(k)$,

$$\mathcal{P}(k) < F(k) = \exp \left[-2(n-1) \left(\frac{k}{n} - p \right)^2 \right]. \quad (\text{E2})$$

To see that $F(k)$ is indeed an upper bound of $\mathcal{P}(k)$ for all n, p and k , let us examine the logarithm of the binomial distribution $\mathcal{P}(k = ny)$,

$$L(y) = \log \binom{n}{ny} + ny \log p + n(1-y) \log(1-p), \quad (\text{E3})$$

where $0 \leq y \leq 1$. The properties, we are interested in, are

- $L(y) < 0$ for all y , since $\mathcal{P}(k) < 1$,
- $\frac{\partial}{\partial y} L(y) \Big|_{y=p} = 0$, and positive for $y < p$ and negative for $y > p$, since $L(y) = \max$ at $y = p$.
- $\frac{\partial^2}{\partial y^2} L(y) = \frac{\partial^2}{\partial y^2} \binom{n}{ny} = -n^2 \left(\psi_1[1 + n(1-y)] + \psi_1[1 + ny] \right)$, where $\psi_1(x) = \frac{d}{dx} \log \Gamma(x)$, the first polygamma function.

By analyzing the series expansion of $\psi_1(1+\eta)$ for large and small η arguments,

$$\begin{aligned} \psi_1(1+\eta) &= \frac{1}{\eta} - \frac{1}{2\eta^2} + \mathcal{O}(\eta^{-3}) & \eta \gg 1 \\ \psi_1(1+\eta) &= \frac{\pi^2}{6} - 2.404\eta + \frac{\pi^4}{30}\eta^2 + \mathcal{O}(\eta^3) & \eta \ll 1, \end{aligned}$$

one can show that $\frac{\partial^2}{\partial y^2} L(y)$ is always negative and has a global maximum at $y = 1/2$, where it takes the value

$$\frac{\partial^2}{\partial y^2} L(y) \Big|_{y=1/2} = -4(n-1) - \mathcal{O}(n^{-1}). \quad (\text{E4})$$

Therefore the constant function $f''(y) = -4(n-1)$ is an upper bound of $L'' = \frac{\partial^2}{\partial y^2} L$. Now, let us integrate both L'' and f'' twice, and choose the integration constants, so that $L(y) < f(y)$. Since $L'(y=p) = 0$,

$$L'(y) < \int_p^y d\zeta f''(\zeta) = -4(n-1)(y-p), \quad (\text{E5})$$

which is chosen to be $f'(y)$, and since $L(p) < 0$,

$$L(y) < \int_p^y d\zeta L'(\zeta) < \int_p^y d\zeta f'(\zeta) = -2(n-1)(y-p)^2, \quad (\text{E6})$$

which is chosen to be $f(y)$. Consequently

$$\mathcal{P}(ny) = \exp[L(y)] < \exp[f(y)] = F(ny). \quad (\text{E7})$$

2. Upper bound on the distribution of the estimated phase

Here we give an upper bound on the distribution of the Ramsey phase Φ , as determined by estimating $\cos \Phi$ and $\sin \Phi$ from two sub-groups of GHZ states (X, Y) , each providing $n/2$ measurement (parity) bits. Qubits in group X are prepared in $[|0\rangle + |1\rangle]/\sqrt{2} =: |+\rangle$, and measured in $|+\rangle$ with probability $p_x = [1 + \cos \Phi]/2$ after time T , while qubits in ensemble Y are prepared in $[|0\rangle + i|1\rangle]/\sqrt{2}$, and measured in $|+\rangle$ with probability $p_y = [1 + \sin \Phi]/2$ after time T . The number of $|+\rangle$ outcomes k_x and k_y from groups X and Y , respectively are binomial random variables with the distribution

$$\mathcal{P}(k_\nu) = \binom{n/2}{k_\nu} p_\nu^{k_\nu} (1-p_\nu)^{n/2-k_\nu}, \quad (\text{E8})$$

where $\nu \in \{x, y\}$. Using the upper bound from Eq. (E2), and noting that $n/2 > 1$ we can give the following upper bound on the joint distribution of k_x and k_y ,

$$\mathcal{P}(k_x, k_y) < \exp \left[-\frac{n}{2} \left(\frac{2k_x}{n} - p_x \right)^2 - \frac{n}{2} \left(\frac{2k_y}{n} - p_y \right)^2 \right]. \quad (\text{E9})$$

Let us introduce the polar coordinates r, φ : $r \cos \varphi = \frac{2k_x}{n} - \frac{1}{2}$ and $r \sin \varphi = \frac{2k_y}{n} - \frac{1}{2}$, and smear the distribution into a continuous density function $\rho(r, \varphi)$, and its upper bound accordingly:

$$\rho(r, \varphi) < \frac{n^2}{4} r \exp \left[-\frac{n}{2} \left(r^2 - 2r \cos(\varphi - \Phi^{\text{real}}) + 1 \right) \right]. \quad (\text{E10})$$

The marginal distribution of r is independent of Φ^{real} , which means that r does not hold any information about Φ^{real} . The upper bound on the marginal distribution of φ can be written as

$$\rho(\varphi) < \left(\frac{n}{4} + \frac{n^{3/2}\sqrt{\pi}}{\sqrt{32}} \right) \exp \left[-\frac{n}{2} \sin^2(\varphi - \Phi^{\text{real}}) \right] \quad (\text{E11})$$

$$\sim n^{3/2} \exp \left[-\frac{n}{2} (\varphi - \Phi^{\text{real}})^2 \right], \quad (\text{E12})$$

where in the second line we assumed $|\varphi - \Phi^{\text{real}}| \ll 1$, and $n \gg 1$. This result is an upper bound on the distribution

of φ , which we are going to use to give an upper bound on the rounding error probability,

$$P_{\text{re}} = 2 \int_{\pi/D}^{\infty} d\varphi \rho(\varphi + \Phi^{\text{real}}). \quad (\text{E13})$$

The rigorous upper bound on the tail of ρ is provided by Eq. (E12), as long as $\pi/D \ll 1$, and $n \gg 1$.

-
- [1] T. L. Nicholson, M. J. Martin, J. R. Williams, B. J. Bloom, M. Bishof, M. D. Swallows, S. L. Campbell, and J. Ye, *Phys. Rev. Lett.* **109**, 230801 (2012).
- [2] N. Lemke, A. Ludlow, Z. Barber, T. Fortier, S. Diddams, Y. Jiang, S. Jefferts, T. Heavner, T. Parker, and C. Oates, *Phys. Rev. Lett.* **103**, 063001 (2009).
- [3] C. W. Chou, D. B. Hume, J. C. J. Koelemeij, D. J. Wineland, and T. Rosenband, *Phys. Rev. Lett.* **104**, 070802 (2010).
- [4] B. J. Bloom *et al.* arXiv:1309.1137 (2013).
- [5] J. Eckstein, A. Ferguson, and T. Hänsch, *Phys. Rev. Lett.* **40**, 847 (1978).
- [6] J. Reichert, M. Niering, R. Holzwarth, M. Weitz, T. Udem, and T. Hansch, *Phys. Rev. Lett.* **84**, 3232 (2000).
- [7] D. J. Jones, S. A. Diddams, J. K. Ranka, A. Stentz, R. S. Windeler, J. L. Hall, and S. T. Cundiff, *Science* **288**, 635 (2000).
- [8] J. Ye, J.-L. Peng, R. J. Jones, K. W. Holman, J. L. Hall, D. J. Jones, S. a. Diddams, J. Kitching, S. Bize, J. C. Bergquist, *et al.*, *J. Opt. Soc. Am. B* **20**, 1459 (2003).
- [9] V. Bužek, R. Derka, and S. Massar, *Phys. Rev. Lett.* **82**, 2207 (1999).
- [10] A. André, A. Sørensen, and M. Lukin, *Phys. Rev. Lett.* **92**, 230801 (2004).
- [11] A. Louchet-Chauvet *et al.*, *New J. Phys.* **12**, 065032 (2010).
- [12] T. Rosenband, arXiv:1203.0288 (2012).
- [13] J. Borregaard and A. S. Sørensen, *Phys. Rev. Lett.* **111**, 090801 (2013).
- [14] S. Huelga *et al.*, *Phys. Rev. Lett.* **79**, 3865 (1997).
- [15] J. Bollinger, W. Itano, D. Wineland, and D. Heinzen, *Phys. Rev. A* **54**, R4649 (1996).
- [16] D. Wineland, C. Monroe, W. Itano, D. Leibfried, B. King, and D. Meekhof, *J. Res. Natl. Inst. Stan.* **103**, 259 (1998).
- [17] T. Rosenband and D. R. Leibbrandt, arXiv:1303.6357 (2013).
- [18] J. Borregaard and A. S. Sørensen, *Phys. Rev. Lett.* **111**, 090802 (2013).
- [19] M. A. Nielsen and I. L. Chuang, *Quantum Computation and Quantum Information* (Cambridge University Press, 2011).
- [20] V. Giovanetti, S. Lloyd, and L. Maccone, *Nature Photonics* **5**, 222 (2011).
- [21] G. S. Uhrig, *Phys. Rev. Lett.* **98**, 100504 (2007).
- [22] Alternatively, it is also possible to perform direct phase feedback.
- [23] See Supplemental Information for details.
- [24] A. André, Ph.D. thesis, Harvard University, Cambridge, Massachusetts (2005).
- [25] G. Santarelli, C. Audoin, A. Makdissi, P. Laurent, G. J. Dick, and A. Clairon, *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* **45**, 887 (1998).
- [26] H. Cramér, *Mathematical Methods of Statistics* (Princeton Univ., 1946).
- [27] W. Itano, J. Bergquist, J. Bollinger, J. Gilligan, D. Heinzen, F. Moore, M. Raizen, and D. Wineland, *Phys. Rev. A* **47**, 3554 (1993).
- [28] Φ_j are estimated within the entire $[-\pi, \pi]$ interval by using pairs of sub-ensembles to measure $\cos \Phi_j$ and $\sin \Phi_j$ simultaneously [17].
- [29] G. Giedke, J. M. Taylor, D. D'Alessandro, M. D. Lukin, and A. Imamoglu, *Phys. Rev. A* **74**, 032316 (2006).
- [30] B. M. Escher, R. L. de Matos Filho, and L. Davidovich, *Nature Physics* **7**, 406 (2011).
- [31] D. Meiser, J. Ye, M. J. Holland. *New J. Phys.* **10**, 073014 (2008).
- [32] P. Kómár, E. M. Kessler, *et al.* (in preparation).
- [33] In principle, the clock stability can further be improved by employing different numbers of copies in each step of the Cascade. However, this possibility will not be pursued in this work.