Heisenberg-Limited Atom Clocks Based on Entangled Qubits

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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. Importantly the protocol is designed to work under realistic conditions where the drift of the phase of the laser interrogating the atoms is the main source of decoherence. The simultaneous interrogation of the laser phase with a cascade of GHZ states realizes an incoherent version of the phase estimation algorithm that enables Heisenberg-limited operation while extending the coherent interrogation time beyond the laser noise limit. We compare and merge the new protocol with existing state of the art interrogation schemes, and identify the precise conditions under which entanglement provides an advantage for clock stabilization: it allows a significant gain in the stability for short averaging time.

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Currently, atomic clocks based on optical transitions achieve the most precise [1–3] and accurate [2,4] 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. While clock protocols based on uncorrelated atoms at best achieve a stability scaling \( \propto \frac{1}{N} \), where \( N \) is the number of atoms—a scaling commonly known as the standard quantum limit (SQL) [14]—the use of entangled resources, in principle, allows one to surpass this limit. However, 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) frequency fluctuations in the laser used to excite the clock transition [in the following also referred to as local oscillator (LO)]. 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 regardless of the noise model being used [12,16–18]. On the other hand, it has been shown that for clocks with local oscillator noise limited stability, the use of moderately squeezed atomic states can yield a modest improvement over the SQL [10,11]. A recent study demonstrated further that, in principle, highly squeezed states could achieve Heisenberg-limited stability (i.e., a \( 1/N \) scaling with the available resources representing the ultimate limit allowed by the laws of quantum mechanics [19]) 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 \( N \) by interrogating uncorrelated atomic ensembles for suitably chosen times [20,21].

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 [22]. Furthermore, we unify previous treatments of decoherence for atomic clocks and incorporate previous proposals involving uncorrelated atoms to effectively narrow the LO linewidth [20,21] and thereby identify ultimate limits to the stability of atomic clocks based on entangled atoms. We find that 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 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 ultranarrow transition of the clock atoms serving as the frequency reference. The long-term stability of such a clock after a given total averaging time $\tau$ is directly related to the precision by which the accumulated laser phase relative to the atoms can be determined. To this end, the phase is repeatedly measured in a standard Ramsey protocol [15]; using the LO, the clock qubits are prepared in a superposition of $|1\rangle$ and $|0\rangle$, denoting the levels of the clock transition. After the qubits evolve freely for a time $T$ (Ramsey interrogation time), they are subsequently measured in an orthogonal basis ($|\pm\rangle \equiv |1\rangle \pm |0\rangle$), which yields an estimate of the accumulated phase difference between the LO and the atomic frequency reference. It is known, that since each of these Ramsey sequences introduces measurement noise, it is optimal to extend the Ramsey time $T$ to its maximum value $T \rightarrow \tau$ [23].

A single GHZ state consisting of $N$ entangled atoms—whose state after the interrogation is $|\text{GHZ}\rangle \propto |0\rangle^\otimes N + \exp(-iN\Phi_{LO})|1\rangle^\otimes N$—accumulates the laser phase (denoted by $\Phi_{LO}$) $N$ times faster than an uncorrelated state, allowing a more precise phase measurement [19]. However, fluctuations in the laser frequency renders the laser phase 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 $2\pi$ error in the estimation. 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 $\Phi_{LO}$, and the resulting laser stability is found to yield no improvement over classical protocols [18].

To address this problem, we use a protocol involving an incoherent version of the phase estimation algorithm [22], similar to the one outlined in [24] but adapted to be applicable also when the frequency fluctuates and phases exceed $2\pi$. The phase estimation algorithm has recently been successfully applied experimentally for global interferometric phase estimation [25,26], and its use in clock synchronization protocols has been discussed [27]. Here, we demonstrate how the same techniques can be applied to guarantee optimal laser stability by allowing the Ramsey interrogation time to be extended to its maximum value.

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 $(\Phi_{LO} + \pi)/2\pi = \sum_{j=1}^{M-1} Z_j/2^j$, 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 $\Phi_{M-1} = 2^{M-1}\Phi_{LO}\mod[-\pi, \pi] = \pi(Z_{M-1})$. Thus, by measuring if the phase is 0 or $\pi$, the last digit of the laser phase can be determined. However, without the remaining digits this information is useless. In our protocol, these digits are found by an additional, simultaneous interrogation with successively smaller GHZ states of $2^{M-2}, 2^{M-3}, \ldots$ entangled atoms (see Fig. 1). Each of these states picks up a phase proportional to its size $\Phi_j = 2^j\Phi_{LO}\mod[-\pi, \pi]$, and this phase gets a contribution of $\pi(Z_{j-1})$. By distinguishing whether the phase is shifted by $\pi$ or not, we can 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 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 above. To overcome this, 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 [20,21,28]. Each of these ensembles acquires a phase that is reduced by multiples of two from the laser phase, and thus, following the arguments from above, allows one to gain information on the 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

![FIG. 1 (color online). The proposed clock operation scheme employs $M$ different groups of clock atoms prepared in correlated states of varying size to interrogate the relative phase $\Phi_{LO}$ of the LO field. A single group $j$ contains $n_j$ independent instances of GHZ-like states, each entangling $2^j$ qubits, and therefore accumulating a phase $\Phi_j = 2^j\Phi_{LO}\mod[-\pi, \pi]$ 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 $(\Phi_{LO} + \pi)/2\pi = (0.Z_1Z_2Z_3\ldots)$, subsequently used to feedback the LO frequency.](190403-2)
the LO noise, and allows the Ramsey time to extend to its optimal value.

In the following, we provide a derivation of the above results combined with feedback analysis that allows us to characterize the achievable stability of a clock using our protocol. Modern clocks periodically measure the fluctuating LO frequency \( \omega(t) \) against the frequency standard \( \omega_0 \) of the clock atoms to obtain an error signal. After each Ramsey cycle of duration \( T \) [i.e., at times \( t_k = kT \) \((k = 1,2, \cdots)\)], the measurement data yield an estimate of the relative phase, \( \Phi_{LO}(t_k) = \int_{t_k}^{t_{k+1}} dt \{ \omega(t) - \omega_0 \} \), accumulated by the LO. This estimate in turn is used to adjust the frequency of the LO: \( \omega(t_k) \rightarrow \omega(t_k) - \alpha \Phi_{LO}^{\text{est}}(t_k)/T \), where \( \Phi_{LO}^{\text{est}}(t_k) \) represents a suited estimator of the phase \( \Phi_{LO}(t_k) \) [29], and \( \alpha \ll 1 \) is a suitably chosen gain.

The stability of the actively stabilized LO, after a total averaging time \( \tau \), is characterized by the Allan deviation (ADEV), which is directly proportional to the measurement uncertainty \( \Delta \Phi_{LO}(t_k) \) after each Ramsey cycle (see the Supplemental Material [30]).

\[
\sigma_\gamma(\tau) = \frac{1}{\omega_0^2} \sum_{k=1}^{T} \sum_{i=1}^{T} T^2 \langle \delta \tilde{\omega}_k \delta \tilde{\omega}_i \rangle \approx \frac{1}{\omega_0^2 \sqrt{\tau T}} \Delta \Phi_{LO}(T). \tag{1}
\]

Here, \( \delta \tilde{\omega}_k = \Phi_{LO}(t_k)/T \) 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, \( \langle \delta \tilde{\omega}_k \delta \tilde{\omega}_i \rangle \approx \langle \delta \tilde{\omega}_1 \rangle^2 \delta_{ij} \) [2,21,31]. Other noise sources (such as the bias of the linear estimator, the Dick effect, or a suboptimal gain \( \alpha \) [32]) are not fundamental, and neglected in the following.

For small values of the accumulated Ramsey phase, the ultimate precision by which this phase can be estimated is determined by the Cramér-Rao bound [19,33] which links the estimation error to the quantum Fisher information (QFI) \( \Delta \Phi_{LO} \sim 1/\sqrt{\mathcal{F}} \) (for a review, see [19]). The QFI, \( \mathcal{F} \), is maximized, e.g., by the use of GHZ states for which \( \mathcal{F} \sim N^2 \). In clock stabilization, however, the LO frequency fluctuations account for the fact that the accumulated Ramsey phase is a random variable which can obtain large values, inherently violating the small phase assumption of the Cramér-Rao bound. In particular for a single GHZ states, phase wraps of the atomic phase, \( \Phi(t_k) = N \Phi_{LO}(t_k) \notin [-\pi, \pi] \), cannot be detected. Consequently, the cycle time \( T \) has to be chosen such that the prior distribution of \( \Phi(t_k) \) is well localized within \([-\pi, \pi]\). This limits the maximally allowed Ramsey time to a value \( T_{\text{max}} \sim \gamma_{LO}^{-1}/N^2 \) (see the Supplemental Material [30]), where we assumed a white frequency noise spectrum of the LO, \( S_\omega(f) = \gamma_{LO} \) (for \( 1/f \) noise one finds the less stringent condition \( T_{\text{max}} \sim \gamma_{LO}^{-1}/N \)). 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 [12,18].

However, unlike the individual particle noise resulting in the finite atom linewidth \( \gamma_{\text{rad}} \), the LO frequency fluctuations affect all clock atoms alike, and this collective noise does not represent a fundamental metrological limitation. We can use a cascade of GHZ states of varying size to measure the \( \Phi_{LO} \) in a binary representation, as discussed above. In general, the phase does not have an exact binary representation ending at the digit \( Z_M \). We therefore employ \( n_0 \) duplicates at each level of the cascade (as opposed to sequential procedure suggested in [24]) \((n_0 = N/\sum_{j=0}^{M-1} 2^j \approx N/2^M)\) to improve the precision. In the case where all digits \( Z_j \) \((j = 1, \ldots, M - 1)\) are determined correctly according to the relation,

\[
Z_j = \left[ 2(\Phi_{j-1} + \pi) - (\Phi_j + \pi) \right]/2\pi, \tag{2}
\]

the last group \((j = M - 1)\) then yields a Heisenberg-limited estimate of the LO phase with accuracy \( \Delta \Phi_{LO}^{\text{pr}} = 1/(2^M - \sqrt{n_0}) \approx 2\sqrt{n_0}/N \).

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 \( \Phi_j^\text{est} \) represents a suitable estimator derived from the \( n_0 \) measurement outcomes), leading to the wrong \( Z_j \), and a variance contribution of \((2\pi 2^{-j})^2\) for \( \Phi_{LO} \). We can approximate their total variance contribution with the sum \( \Delta \Phi_{LO}^{\text{re}} = \sum_{j=1}^{M-1} (2\pi 2^{-j})^2 \), where \( P_{re} = 2 \int_{[0,1]} d\varphi \rho(\varphi) \), and \( \rho(\varphi) \) is the Gaussian probability distribution of the error \( \Phi_j^\text{est} - \Phi_j \) with a width proportional to \( 1/\sqrt{n_0} \) (see the Supplemental Material [30]). Consequently, rounding errors can be exponentially suppressed by choosing a sufficiently large value for \( n_0 \). The total measurement uncertainty of this estimation scheme is thus \( \Delta \Phi_{LO}^2 = \Delta \Phi_{LO}^{\text{pr}}^2 + \Delta \Phi_{LO}^{\text{re}}^2 \). In [30], we show that the optimal allocation of resources is achieved for the choice \( n_0^{\text{opt}} \sim (16/\pi^2 \log(N)) \), for which rounding errors are negligible, yielding the total measurement accuracy,

\[
\Delta \Phi_{LO} \approx \Delta \Phi_{LO}^{\text{pr}} = \frac{8}{\pi} \sqrt{\log(N)/N}. \tag{3}
\]

This measurement precision obtains the Heisenberg limit (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 \( \Phi_{LO} \in [-\pi, \pi] \) in each cycle. However, for realistic laser noise spectra there is always a finite probability that the LO phase \( \Phi_{LO} \) 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 \( \Delta \Phi_{LO}^{\text{slip}} = (2\pi)^2 P_{\text{slip}} \), where \( P_{\text{slip}} = 2 \int_{[0,1]} d\varphi \rho_{\Phi_{LO}^\text{slip}}(\varphi) \), and \( \rho_{\Phi_{LO}^\text{slip}} \) is the Gaussian prior distribution of \( \Phi_{LO} \). Its width
The linewidth of the atomic clock transition \( \gamma_{LO} \) is only limited by individual noise processes. The finite atomic limit is reached for \( \gamma_{LO} \) values rapidly after the averaging time \( T \), resulting in the clock stability, \( \sigma_T \), given by

\[
s_T = \frac{2}{\omega_0 \tau N} \sqrt{\frac{n_0}{\pi}} \sqrt{\log(N)},
\]

where \( n_0 \) is the optimization value for \( n_0 \) found above the resulting clock stability. This result identifies the possible advantage of using the proposed cascaded GHZ protocol which interrogates the LO with uncorrelated atoms for exponentially increasing Ramsey times in each cycle [20,21]. This protocol can be understood as the classical part (\( j = 0 \)) of the cascaded interrogation proposed here. 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 \( 1/\tau \) 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. This result identifies the possible advantage of using entanglement previously debated in the literature [9–13,16,18,35]: while the long term limitation is set by atomic decoherence, entangled atoms reach this limit faster thus improving the bandwidth of the stable oscillator. Our results motivate the development of quantum enhanced atomic clocks based on entangled ions [36] and neutral atoms. Furthermore, it lays the foundations for the recently proposed network of quantum clocks [37] which achieves the optimal use of resources in a global network through network-wide entangled states.

\[ \text{FIG. 2 (color online). Allan deviation} \sigma_T \text{ for different protocols as a function of averaging time} \tau, \text{normalized to the standard quantum limit, for} \gamma_{LO}/\gamma_{\text{ind}} = 10^3. \text{The solid black line corresponds to the standard scheme using a single uncorrelated ensemble. It fails to reach the fundamental noise floor set by the atomic transition linewidth (cf., Eq. (5), broken line). A more sophisticated classical scheme which uses exponentially increasing Ramsey times in each cycle [20,21] allows us to extend the regime of linear scaling with} 1/\tau \text{ up to the point where the bound (5) is met. In comparison, the proposed cascaded GHZ protocol (blue solid curves) enables an} \sim N \text{ times faster convergence. For short averaging times the stability is enhanced by a factor} \sqrt{N} \text{ as compared to classical protocols.} \]
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[29] Alternatively, it is also possible to perform direct phase feedback.