Phase stabilization of a frequency comb using multi-pulse quantum interferometry

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From the interaction between a frequency comb and an atomic qubit, we derive quantum protocols for the determination of the carrier-envelope offset phase, using the qubit coherence as a reference, and without the need of frequency doubling or an octave spanning comb. Compared with a trivial interference protocol, the multi-pulse protocol results in a polynomial enhancement of the sensitivity $O(N^{-2})$ with the number $N$ of laser pulses involved. We present specializations of the protocols using optical or hyperfine qubits, Λ-schemes and Raman transitions, and introduce methods where the reference is another phase-stable cw-laser or frequency comb.

Quantum Physics has experienced a dramatic revolution in the last decades, with unprecedented and universally recognized [1] progress in the control and observation of individual quantum systems. In this respect, trapped ions [2, 3] is one of the most mature setups, with unbeaten precision in the realization of single- [4] and two-qubit [5] unitaries and measurements [6, 7], closely followed by neutral atoms [8]. This spectacular progress underlies a number of “spin-off”, such as the characterization of atomic properties using entanglement [9] or the development of quantum algorithms and protocols [10–12] for studying molecular ions. The synergy is even more advanced in the field of metrology, with accurate atomic clocks assisted by quantum gates [13, 14] or the use of atomic squeezing for enhanced magnetometry [15, 16].

Despite the exquisite precision of AMO systems, the times scales involved in control and detection (∼10 μs to 1 ms) have so far prevented exporting these techniques to study ultrafast processes. In this work, we show that the speed of AMO setups is not a limiting factor to accurately stabilize the carrier-envelope offset (CEO) phase of a frequency comb. CEO phase effects are relevant for few-cycle pulses, though effects in multicycle pulses have also been reported [17]. The first observation of such an effect was reported in the spatial asymmetry of above-threshold ionization (ATI) from Kr gas [18] and in the x-ray emission from Ne [19]. The direction of photocurrents injected in semiconductors is also controlled by the CEO phase [20, 21] and more recently, the absolute CEO phase of single pulses has been measured in ATI experiments [22]. In general, the study of the CEO phase has been focused on its spectral components as these determine the temporal evolution of the electric field within a pulse [23], while only a few reports have addressed time-domain measurements of the relative phase of successive pulses in a train [24, 25]. The methods presented below follows this less-beaten path.

Let us introduce the notion of “multi-pulse quantum interferometry” (MPQI), where an atom acts as a nonlinear, fast-response detector that efficiently measures the differences between ultrashort laser pulses. Modelling the atom-pulse interaction as a sequence of unitaries, $\{U_i\}_{i=1}^N$, through a suitable reordering of the pulses, additional gates and measurements, we build protocols that accurately determine the differences among the pulses, or the properties of the individual pulses themselves. Compared with cw laser interferometry, this approach profits from a polynomial enhancement of the sensitivity because a single atom accumulates many interferometric events.

A direct application of MPQI is the characterisation and stabilisation of a frequency comb. The comb [26, 27] is a device that produces an uninterrupted train of laser pulses with a fixed duration, $\tau$, and a regular spacing, $T$ [cf. Fig. 1a]. Stabilising a comb is ensuring that the offset frequency, $\nu_0$, remains a constant and well-known value, and that the spectrum is a collection of regularly spaced teeth with frequencies $f_n = n/T + \nu_0$. This problem was solved by Haensch and Hall [28, 29] in frequency space, interferometrically comparing different teeth in the limit of many pulses. Note that this requires a comb whose spectrum spans at least an octave, as some Ti:Sa lasers, or broadening the light with a nonlinear fiber. This stabilisation enables direct frequency comb spectroscopy, accurately revealing the (unknown) atomic level structure of neutral atoms [30, 31] and ions [32, 33].

We will rather work on the time-domain image of the pulse train. Here, the effect of the offset frequency, $\nu_0$, is to change the CEO phase from pulse to pulses, as in $\phi_{n+1} - \phi_n = \Delta \phi = \nu_0 T$ [cf. Fig. 1a]. To address the problem of comb stabilisation we will use MPQI, designing protocols that detect the phase difference between pulses with the greatest accuracy possible. The resulting method does not require an octave-spanning comb, broadening or frequency doubling. It is thus useful for a wider variety of lasers, demands less power to perform the stabilisation, and it may profit from the ever-growing precision in atomic interferometry.

Single-pulse unitary.- We start by determining the unitaries associated to each laser pulse and how they depend on the carrier-envelope phase, $\phi_n$. The interaction of multi-level atoms with a frequency comb has been studied previously [34]. For simplicity and in order to produce analytical estimates, we will model this inter-
action in the Rotating Wave Approximation (RWA) \[35\]

\[H_{\text{RWA}} = \frac{1}{2}(\omega_{\text{at}} - \tilde{\omega})\sigma_z + s(t) \left(e^{-i\phi_m}\sigma^+ + H.c.\right),\]

(1)

Here, \(m\) is the pulse index, \(s(t) \geq 0\) is the pulse envelope, \(\tilde{\omega} = 2\pi\nu\) is the comb carrier frequency, \(\omega_{\text{at}}\) is the atomic transition frequency \((\hbar = 1\) throughout\), there is an unknown (constant) phase \(\phi_m\) for each pulse \[36\], and \(\sigma_{x,y,z}\) are the Pauli matrices. The RWA works \[37\] for pulses which contain more than 30 periods of the carrier frequency, \(\tau > 30/\nu\), and allows us to explicitly write the pulse unitaries

\[U_m = \cos(\theta_m) + i\sin(\theta_m)\sigma_{\phi_m} = e^{-i\phi_m}\sigma_z U_0 e^{i\phi_m}\sigma_z,\]

(2)

in terms of the total Rabi flip angle of a single pulse, \(\theta = \int_{-\tau/2}^{\tau/2} s(t) dt\), with \(\sigma_{\phi_m} = \cos(\phi_m)\sigma_x + \sin(\phi_m)\sigma_y\). In what follows, we assume that the comb is almost resonant, \(\tilde{\omega} = \omega_{\text{at}}\), and has uniform intensity, that is \(\theta_m = \theta\). These assumptions imply that we only need to stabilize the pulse-to-pulse phase difference \(\Delta\phi\).

**Multi-pulse unitaries.** We want a protocol that efficiently detects the difference between the sequence of unequal pulses \(U_{\text{tot}} = \prod_{i=1}^N U_i\), and the ideal case \(U_1^N\). Let us first assume an ideal qubit, seeking an ordering of pulses with which the fidelity \(|\text{tr}(U_i^N|U_{\text{tot}})|\) decreases most rapidly with \(N\). The simplest protocol (1A) applies \(N\) consecutive pulses [cf. Fig. 2a] with low intensity, \(\theta \ll 1\), on the qubit, which adiabatically follows the phase

\[U_{\text{tot}}^{(1A)} \approx \mathbb{1} + i\theta \frac{\sin(N\Delta\phi)}{\sin(\Delta\phi)} \left[e^{i(N+1)\Delta\phi}\sigma^+ + H.c.\right] + \ldots \]

(3)

Note how the pulse-to-pulse phase difference \(\Delta\phi\) decreases the amplitude of the Rabi oscillations and can be measured. However, as we will show later on, the functional dependence on \(\Delta\phi\) implies a low sensitivity on the phase in practical implementations of the protocol.

We can do much better by changing the intensity regime to \(\theta = \pi/2\) (protocol 1B), where each comb pulse is capable of completely flipping the state of the atom. Under these conditions, for an even set of pulses we get

\[U_{\text{tot}}^{(1B)} = \prod_{i} U_i = \exp \left[-2i \sum_{k=1}^{N/2} (\phi_{2k} - \phi_{2k-1})\sigma_z\right],\]

(4)

which for constant \(\Delta\phi\) implies \(U_{\text{tot}}^{(1B)} = \exp(-iN\Delta\phi\sigma_z^2)\).

Now \(\Delta\phi\) appears in a favorable position for the interferometric detection with an enhancement proportional to the number of pulses, \(N\).

From Eq. (4) it is obvious that the sensitivity can be increased by maximizing the phase difference between consecutive pulses, \(\phi_{2k} - \phi_{2k-1}\). To take advantage of this, we design a set of protocols that split the original pulse train, extracting a sequence of \(N/2\) pulses that will be delayed a time \(T_d > T\). This sequence of pulses will then be interleaved with the original one, as shown in Fig. 2b. The phases then read \(\phi_{2k} = k\Delta\phi + \Delta\phi T_d/T\) and \(\phi_{2k-1} = k\Delta\phi\). Introducing this in Eq. (4) we obtain the unitary corresponding to protocol 2B:

\[U_{\text{tot}}^{(2B)} = \exp(-i\sigma_z^2\Delta\phi \times N T_d/T),\]

(5)

which contains an enhancement factor, \(N_d = T_d/T\). This is optimal with respect to any rearrangement of the pulses, using each pulse only once.

**Interferometry and sensitivity.** We must now transfer the information of the acquired phase to a measurable quantity: the populations of the atomic states. For this, we will complete the previous unitaries with additional operations and measurements that allows us to estimate \(\Delta\phi\) and \(\theta\). Out of \(2M\) atoms, \(M\) are subject to the following steps [cf. Fig. 2c]: (i) initialize the atoms to the ground state, \([0]\), (ii) apply a Hadamard gate, \(U_H = \frac{1}{\sqrt{2}}(\sigma_x + \sigma_z)\), (iii) apply a reference phase \(\xi\) onto the level \([1]\), (iv) let the atom interact with the comb as described before, (v) undo the Hadamard gate and

![FIG. 1.](image1)

![FIG. 2.](image2)
\[\begin{array}{|c|c|c|}
\hline
\text{Two levels} & \theta \ll 1 & \theta \approx \pi/2 \\
\hline
1 sequence & \sqrt{M} & N\sqrt{M} \\
\hline
2 sequences & \Delta \phi & \frac{N_d\sqrt{M}}{N_d\sqrt{M}} \\
\hline
\Lambda-\text{scheme} & \theta \ll 1 & \theta \approx \pi/2 \\
\hline
1 delay & N_{d}\sqrt{M} & - \\
\hline
2 delays & |N_{d2} - N_{d1}|\sqrt{M}/M & |N_{d2} - N_{d1}|\sqrt{M}/M \\
\hline
\end{array}\]

TABLE I. Experimental sensitivity, \(\sigma^{-1}\) of a set of \(2M\) two-level or three-level atoms to each of the protocols described in the main text. \(N\) is the number of pulses in a sequence, which in the delayed case are combined with \(N\) pulses from a later time, \(T_d = N_dT\).

measure the state of the atom, \(s \in \{0, 1\}\). The measurement outcome is described by the probability distribution, \(P_1(s|\theta, \Delta\phi)\). For the remaining \(M\) atoms we skip (ii), obtaining the distribution, \(P_2(s|\theta, \Delta\phi)\). It is important to remark that we need no phase coherence between the comb and the lasers that implement the Hadamard gates, and that the reference phase, \(\xi\), is computed a priori to maximize the sensitivity of \(P_{1,2}\) to the phase.

The functions \(P_1\) and \(P_2\) convey all the information that is accessible in the lab: from the measurements of \(s\) in \(P_1\) and \(P_2\) experiments, one should compute different estimators and use them to infer the values of \(\theta\) and \(\Delta\phi\), with uncertainties \(\sigma_\theta\) and \(\sigma_{\Delta\phi}\). Using error propagation and the Fisher information we obtain fundamental lower bounds and practical estimates \([37]\) of the sensitivity \((\sigma_{\Delta\phi}^{-1}\) and \(\sigma_\theta^{-1}\)) of each protocol \((1A, 1B, 2B, \) plus another combination, labelled \(2A\)). As summarized in Table I, it is possible to build estimators of minimal variance for \(\theta\) and \(\Delta\phi\), which saturate the fundamental lower bounds. Moreover, we observe that all protocols but \(1A\) improve over the standard statistical sensitivity, \(\sqrt{M}\), thanks to the large number of pulses or to the use of pulses from well-separated times. In practical implementations both \(N\) and \(N_d\) span several orders of magnitude, providing a sensitivity comparable to the state of the art.

Three-level schemes.- In real atoms, if the qubit states 0 and 1 are dipole-coupled by a comb, spontaneous emission may severely limit the total duration of the experiment. One solution is to use dipole-forbidden transitions restricted in practice to the \(\theta \ll 1\) regime. A more attractive configuration is the \(\Lambda\)-scheme shown in Fig. 3, where two long-lived states of the atom, \(|0\rangle, |1\rangle\), talk via an intermediate level, \(|e\rangle\). Applying combs or other lasers with orthogonal polarizations on the legs of the \(\Lambda\)-scheme, we can create effective Rabi oscillations between the qubit states, \(|0\rangle\) and \(|1\rangle\), while still keeping the population of \(|e\rangle\) so small that spontaneous emission is negligible.

A simple way to minimize the spontaneous emission is to turn the \(\Lambda\)- into a Raman scheme, detuning the lasers that couple \(|0\rangle, |1\rangle\) with \(|e\rangle\). Such Raman processes mix well with our algorithms. To start, if we have already stabilized the phase of a cw laser, we can combine it with the pulses from the comb [cf. Fig. 3a]. This process enables an accurate determination of the pulse phase with respect to the cw source. The result will be a sequence of effective unitaries with some average Rabi angle, \(\theta'\), and a pulse phase \(\phi'_m = \phi_m - \phi_{\text{ref}}\), where \(\phi_{\text{ref}}\) is the phase of the stabilized source. The identifications \(\theta \rightarrow \theta'\) and \(\phi_m \rightarrow \phi'_m\) directly translate all protocols above to this new setup. Likewise, one may combine the frequency comb with a stabilized one [cf. Fig. 3b] and use our protocols to reconcile both sources.

A more interesting use of Raman transitions is to achieve self-referencing with hyperfine qubits. For this, we repeat the scheme from Fig. 3b, combining two pulses from the same comb, but with a relative delay, \(T_d\). This amounts to a self-referenced interferometric scheme based on time shifts, not requiring frequency shifting nor shearing \([23]\). The phases of both pulses effectively combine in a nontrivial way in the unitary associated to the Raman process, \(\phi'_m = \phi_m - \phi_{m-N_d} = N_d\Delta\phi\). We can apply a sequence of \(N\) pulse pairs with total angle which should optimally lay around \(N\theta' \approx \pi/4\)

\[
U(1A, \text{Raman}) = e^{-iN_d\Delta\phi\sigma_z}e^{i\theta'\sigma_x}e^{iN_d\Delta\phi\sigma_z},
\]

and use Ramsey interferometry to measure both \(\theta'\) and \(\Delta\phi\). A generalization of protocols \(2A\) and \(2B\) is also possible using a linear optics circuit with two delay lines, so that each atom is hit by pairs of pulses with alternating phases \((\phi_m, \phi_{m-N_d})\) and \((\phi_m, \phi_{m-N_d})\). This leads to the sensitivities shown in the second half of Table I.

Note that the use of Raman schemes demands the setup to be interferometrically stable up to a fraction of a wavelength. When a single pulse interacts with a two-level atom it does not matter whether the delay is a multiple of the comb period, or fails by a small amount, \(\delta T = T_d - N_dT\). This is so because only the phase of the pulse relative to the envelope enters the unitary and this only contains information on
\(\nu_0N_dT\). However, in Raman schemes, where two pulses overlap in time, their relative delay appears as a new parameter that influences the operations, not only in the accumulated Rabi angle, \(\theta^r\), but also in the phase. In particular, the measured phase difference is actually \(\Delta\phi = \Delta\phi + \omega_0T\), with a contribution due to the interferometric path \(\delta dT\), which must be separately stabilized.

To remove the need for interferometric stability we can use a different \(\Lambda\)-scheme in which the comb only interacts with one leg, performing \(\pi/2\) rotations. The unperturbed and delayed pulses arrive closely in pairs, but without temporal overlap, implementing the sequence \(|1\rangle \rightarrow -e^{i(\phi_m - \phi_m - \phi_d)} |1\rangle\). Due to the lack of overlap, the delay errors drop and the effective operation is a phase gate in the qubit space. Spontaneous emission simply lowers the visibility and it is small because the excited state will only be populated a brief time \(T_e = \mathcal{O}(\tau)\). Denoting by \(\gamma\) the spontaneous emission rate of the \(|e\rangle\) state, we may afford \(N = -\log(\epsilon)/\gamma T_e\) pulses before we lower the visibility by \(\epsilon\). Using a typical value of \(1/\gamma = 8\text{ns}\), and a safe value \(T_e = 100\text{ps}\), spontaneous emission would be just 10\% for about 200 pulses, sufficient to implement the last protocol in Table I.

**Errors.** An actual experiment will have sources of error which influence the atom’s phase: ac Stark and Zeeman shifts induced by scattered light and stray magnetic fields, and also small detunings of the laser. In all cases the effect is similar: a random term \(\epsilon(t)\sigma_t\) makes the atomic levels fluctuate on a time scale which is much longer than \(\tau\). Mathematically, this induces a time-dependent uncertainty in \(\Delta\phi\) of order \(\sigma_t \times (t_{m+1} - t_m)\), where \(\sigma_t\) is the standard deviation of \(\epsilon(t)\) from its (zero) average, and \(t_m\) the arrival time of each pulse. This error can be cancelled using the spin-echo technique \(\text{[38]}\) or, more directly, in protocols 2A and 2B one can calibrate the delays so that the pulses in a Raman pair arrive closely spaced, say 10 ps apart \(\text{[39]}\). In this way, a pessimistic ac Stark shift \(\sigma_\phi \sim 100\text{Hz}\) induces an error of only \(10^{-9}\) radians in the determination of \(\Delta\phi\).

Another source of error is temperature: when atoms move between pulses, they sample the spatial variations of phase and intensity in the laser. We can eliminate both types of error \(\text{[37]}\) by (i) working in a Raman configuration which does not transfer net momentum to the atom and (ii) ensuring that the lasers are not tightly focused. Both techniques also suppress any motion-qubit entanglement and the associated decoherence.

The protocols discussed so far admit many different practical implementations: from neutral atoms to trapped ions. For concreteness, we will discuss this last setup, which has witnessed an astonishing progress in the field of ultrafast laser-ion interaction \(\text{[40, 41]}\). The long coherence times in these setups, \(t_{\text{coh}} \sim 1\text{s}\) \(\text{[42]}\), allow in principle to consider trains of up to \(N = t_{\text{coh}}/T \sim 10^8\) pulses for a typical comb with \(f_{\text{rep}} \sim 100\text{MHz}\) \(\text{[43]}\). Within the Raman schemes, using a single ion and one delay line, this already allows us to detect a pulse-to-pulse phase difference around \(\Delta\phi \sim 10^{-8}\) rad and thus calibrate the comb offset below \(\nu_0 \leq \Delta\phi/T \sim 1/t_{\text{coh}} \sim 1\text{Hz}\) —a remarkable precision for 1 s interrogation time!

The numbers improve, \(\Delta\phi \sim 10^{-15}\) using the same pulses in a \(\Lambda\) scheme, thus reaching a limit in which the previous errors become relevant. The numbers decrease marginally, \(\Delta\phi \sim 10^2 - 10^{10}\), using shorter duty cycles \(\sim 1\text{ms}\). These are optimistic sensitivities that go beyond what many existing lasers can hold: in existing combs, the phase may fluctuate violently after a shorter number of pulses. Our scheme cannot cope with that: we may only increase the delays \(T_d\) and improve the accuracy up to a time scale at which these jumps happen. However, in practice such slow fluctuations set a limit on how well those combs may be stabilized by any method.

Summing up, in this work we have presented an exciting line of research: the development of quantum algorithms for the characterization and stabilization of pulsed light sources using trapped atoms. Our work relies on the following key ideas. First, a single atom may accumulate the effect of multiple laser pulses, computing their differences through the appropriate choice in the order of pulses, the intermediate gates and the final measurements. Second, there is a polynomial enhancement in the sensitivity of pulsed interferometric protocols with respect to conventional interferometry, arising from the combination of multiple events in a single atom. Third, these ideas can be used to detect changes in the phase of the pulses emitted by a frequency comb. This is possible because the unitary implemented by a single comb pulse is sensitive to both the intensity and the CEO phase, and not to the time at which the pulse arrives.

The combination of all those ideas results in a variety of quantum protocols for stabilizing frequency combs. The schemes are particularly suitable for non-octave spanning combs with a low intrinsic phase noise, such as amplified high-power Ti:Sapphire laser systems, where significant phase noise is introduced by the amplification stages. In a near future, we expect to develop further pulse combination and shaping protocols, and obtain formal results about the optimality of the algorithms, comparing their performance with existing linear and nonlinear interferometric methods. Our protocols can be generalized also to work beyond RWA and to include and characterize other properties of the comb, such as pulse-to-pulse intensity fluctuations. We anticipate MPQI will enable new progress in fields as diverse as ultrafast science, frequency metrology and direct frequency-comb spectroscopy, or coherent control of molecular processes.

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SUPPLEMENTARY MATERIAL

In the following pages we provide more details for the following concepts which are spelled in the body of the paper:

§A Justification of the Rotating Wave approximation and its influence in the following calculations.

§B Study of the Raman transitions and three level systems.

§C Composition of unitaries for the different metrology protocols.

§D Analysis of the sensitivity of the protocols using Fisher information and error propagation.

§E Modelization of experimental errors and their influence in the quantum gates.

§F Practical implementation with trapped ions and estimation of achievable sensitivities.

§G Ultimate limits of these ideas based on pulse shaping techniques.

§A. RWA AND CONVENTIONS

We model the atom interaction with a single laser pulse in the semiclassical limit

\[ H = \frac{\omega_{at}}{2} \sigma_z + s(t) \cos(\bar{\omega}t + \phi_m) \sigma_x. \]  

(7)

Here, \( s(t) \geq 0 \) is the pulse envelope, and \( \bar{\omega} = 2\pi\nu \) is the comb carrier frequency. The evolution under this Hamiltonian is described by a unitary operator that satisfies the Schrödinger equation \( i\frac{d}{dt}W(t) = H(t)W(t) \). We are going to split out an evolution with the spin operator \( \sigma_z \) as \( W(t) = \exp(-i\bar{\omega}\sigma_zt/2)U(t) \). This operator evolves now according to

\[ i\frac{d}{dt}U = \frac{\omega_{at} - \bar{\omega}}{2} \sigma_z + s(t)(e^{i\bar{\omega}t+i\phi_m} + c.c.)(e^{i\bar{\omega}t} + H.c.). \]  

(8)

Assuming that the RWA is valid, in this Hamiltonian we can neglect the counter rotating terms, such as \( e^{i2\bar{\omega}t+i\phi_m} \sigma^+ \), and keep only those that are slowly varying.

The result is \( H_{\text{RWA}} \) in the body of the paper.

But when is the RWA valid? We have performed numerical simulations of the evolution of the qubit under Eq. (7), varying the duration of the pulse or number of oscillations it contains, as well as the intensity and detuning. In Fig. 4a we show the fidelity, \( F \), of a resonant pulse, with a pulse area \( \theta = \pi/4 \), and a variable pulse length, \( \tau \). The validity of RWA is also challenged by the inaccuracy of the control parameters, and in particular the driving frequency: as Fig. 4b shows, the unitary is affected by the detuning, and the differences between the RWA and the full model increase as the pulse length decreases. In practice this is not a problem, for we expect the detuning of the comb to be smaller than 1%. As a final check, we show in Fig. 4c that the phase of the unitary is indeed proportional to the phase of the pulse. The main message is that for pulses above 30 oscillations, we are safe using the RWA Hamiltonian.

§B. RAMAN TRANSITIONS

Our Raman protocols are developed assuming that we can use ultrashort pulses to implement Raman transitions between two states, \( a \) and \( b \), mediated by a third one, \( c \), which remains unpopulated at the end of the pulse. (Due to the very short duration of the interroga-
tion sequence, it is not necessary that $c$ be unpopulated at all times, as is usual in STIRAP processes.) Importantly, we need that such operations implement the same quantum gates and carry the same phase information as the original designs. We are going to discuss both requirements and how they are achieved.

Note that Raman transitions with very short pulses have been demonstrated experimentally by the group of C. Monroe et al in a series of works that implement quantum gates with trapped ions and pulsed lasers [44, 45].

In these references, an interpretation based on Raman transitions induced by all the comb teeth is provided, but here we will discuss a different one.

For us the key aspect of a Raman transition is the fact that the intermediate state, $c$, is completely depopulated at the end of the process. In order for this to happen, we need that the energy of the final state is similar to the energy of the original one. Intuitively, this implies that the inverse of the duration of the process has to be smaller than $\delta = \omega_{at} - \omega$, the detuning of the laser from the atomic transitions $\{a, b\} \leftrightarrow c$, but larger than the difference $|\omega_{ac} - \omega_{bc}|$. As shown in Figs. 3a-b, this qualitative appreciation remains true even for rather extreme cases. In those exaggerated plots, we see that pulses with a detuning $\delta \sim 0.2\omega_{at}$ work fine even when they only contain 10–20 oscillations of the laser. In this regime the excited state $c$ is significantly populated during the pulse, but it has a population of less than $10^{-3}$ at the end.

The other aspect we demand from these pulses is the fact that they must carry information on the phase of the laser. To check this, we analyze the interaction between the three-level atom and the light using a simple Hamiltonian,

$$H = s(t) \cos(\omega t + \phi_1) |c\rangle \langle a| + H.c. \tag{9}$$

$$+ s(t) \cos(\omega t + \phi_2) |c\rangle \langle b| + H.c$$

$$+ \omega_{at} |c\rangle \langle c|,$$

which under the RWA becomes

$$H_{RWA}(\phi_1, \phi_2) = s(t) e^{i\phi_1} |c\rangle \langle a| + H.c. \tag{10}$$

$$+ s(t) e^{i\phi_2} |c\rangle \langle b| + H.c$$

$$+ (\omega_{at} - \omega) |c\rangle \langle c|.$$

Note how $H_{RWA}(\phi_1, \phi_2)$ is related to $H_{RWA}(0, 0)$ through a unitary transformation $\exp(-i\phi \sigma^z_{ab})$ in the $\{a, b\}$ subspace, with the relative phase $\phi = \phi_2 - \phi_1$. In other words, according to the RWA the phase of the laser is mapped onto the relative phase between the states. The question is whether this behavior also follows from the original Eq. (9). We have performed numerical simulations of the three level system in Eq. (9) and the conclusions are: (i) There is always a small deviation between the real phase and the RWA approximation. (ii) This deviation decreases with decreasing detuning, as in the two-level system. (iii) The actual phase experienced by the atom is a monotonic function of the laser phase, that is $\phi_s(\phi_1)$ grows with $\phi_1$. These properties are exemplified in Fig. 5c for a case with 2% detuning, where the deviations from the RWA are small, below 1%, but the nonlinear behavior is clear in the inset.

It would seem that, since we are striving for large accuracies in the stabilization protocol, errors of 1% would be enough to discard the protocols. However, we have to remember that we are not actually measuring the absolute phase, but the phase difference between pulses. Hence,
stabilizing $\phi$, which is a smooth, monotonic function of the laser phase, is equivalent to (and as accurate as) stabilizing $\phi_t$.

§C. ANALYTICAL PULSE ESTIMATES

We summarize some of the arguments in the body of the article regarding the composition of pulses.

Pulse composition

Let us start with the case of a sequence of pulses with $\theta \ll 1$ and a uniform carrier-envelope frequency mismatch, $\phi_m = m\Delta \phi$. The combination of pulses reads

$$U_{tot}^{(1A)} = \prod_{m=1}^{N} e^{-i\phi_m \sigma_z} e^{i\theta \sigma_x} e^{i\phi_m \sigma_z}$$

$$= \cos(\theta) + i \sin(\theta) \sum_{m=1}^{N} e^{-i\phi N \sigma_z} \sigma_x e^{i\Delta \phi \sigma_z}$$

$$\simeq \mathbb{1} + i \theta e^{-i(N+1)\Delta \phi} \frac{\sin(N\Delta \phi)}{\sin(\Delta \phi)} \sigma^x + \text{H.c.} + \mathcal{O}(\theta^2)$$

Note that in this context $\Delta \phi$ has the effect of a detuning and suppresses for long trains any excitation probability induced by the pulses. When we work around $\theta = \pi/2$ we obtain instead

$$U_{tot}^{(1B)} = \prod_{m=1}^{N} i e^{-i\phi_m \sigma_z} \sigma_x e^{i\phi_m \sigma_z}$$

If we assume that the number of pulses is even, we can use the anticommutation $\sigma_x \sigma_z = -\sigma_z \sigma_x$ and

$$e^{-i\phi_m \sigma_z} \sigma_x e^{i\phi_m \sigma_z} = -e^{-i\phi_m - 1 \sigma_z} \sigma_x e^{i\phi_m - 1 \sigma_z}$$

recovering the formula

$$U_{tot}^{(1B)} = \exp \left[ -2i \sum_{k=1}^{N/2} (\phi_{2k} - \phi_{2k-1}) \sigma_x \right]$$

from the paper.

Some optimality considerations

We now prove that the sequence for protocol 2B (2 sequence of pulses split from the original train with a time delay) is optimal when our only resource is the comb laser. As seen before, if we work around $\theta = \pi/2$ we obtain the analytical formula

$$U_{tot}^{(1B)} = \exp \left[ -2i \sum_{k=1}^{N/2} (\phi_{2k} - \phi_{2k-1}) \sigma_x \right]$$

and our protocol accumulates phase quite fast, about $\mathcal{O}(NN_d)$ for $N$ pulses, where $N_d$ depends on the delay. It is possible to prove that for any rearrangement of the same set of pulses (that is, with the same phases and intensity as before) this is the largest accumulation that can be detected.

If $\sigma$ is a permutation for a certain arrangement of initial pulses, we can use the analytical expression for the arbitrary product of a train of pulses with different phases to compute product of the unitaries after the permutation

$$\prod_{i=1}^{M} U_{\sigma(i)} = e^{2i \sum_{i=1}^{M} (-1)^{\sigma(i)} \phi_{\sigma(i)} \sigma^x}$$

It is possible to find all permutations $\sigma$ such that they maximize $|\sum_{i=1}^{M} (-1)^{\sigma(i)} \phi_{\sigma(i)}|$. Suppose the original pulses are ordered in terms of their carrier-envelope phase $\phi_n$, then it is quite straightforward to see how to construct optimal rearrangements of these pulses. Consider $\alpha$ a permutation of the first half of pulses and $\beta$ a permutation of the second half of pulses. Then, the optimal set of rearrangements will be those formed by pulses labelled according to their carrier-envelope phase as $\{\phi_{\alpha(1)}, \phi_{\beta(N/2+1)}\}_{i=1,...,N/2}$ or of the form $\{\phi_{\beta(N/2+i)}, \phi_{\alpha(i)}\}_{i=1,...,N/2}$.

In particular, our proposed protocol corresponds to $\alpha$ and $\beta$ being the identity permutation. This protocol accumulates the largest possible amount of phase after the action of the pulses onto the ion.

The fastest phase-accumulation protocol: phase referencing

If we allow for more gates, performing unitaries in between the pulses, we can measure not only the phase difference, but also the total sum of the carrier-envelope phases $\sum_i \phi_i$. In order to do so, the new set of gates and unitaries, considered in order, would be $\{\sigma_x, U(\phi_1), \ldots, \sigma_x, U(\phi_N)\}$, for which the overall product is

$$\prod_{i=1}^{M} \sigma_x U(\phi_{\sigma(i)}) = \prod_{i=1}^{M} \sigma_x e^{-i\phi_{\sigma(i)} \sigma_x} e^{i\phi_{\sigma(i)} \sigma_x}$$

$$= \prod_{i=1}^{M} e^{i\phi_{\sigma(i)} \sigma_x^2} e^{i\phi_{\sigma(i)} \sigma_x} = \prod_{i=1}^{M} e^{i\phi_{\sigma(i)} \sigma_x} e^{i\phi_{\sigma(i)} \sigma_x}$$

$$= \prod_{i=1}^{M} e^{2i\phi_{\sigma(i)}} = e^{2i \sum_i \phi_{\sigma(i)}}$$

where we use both $\sigma_x e^{-k \sigma_x} = e^{k \sigma_x} \sigma_x$ and $\sigma_x^2 = Id$.

Note however this protocol demands $\sigma_x$ gates in between the pulses. Since the phase of these gates is stable, we can thus view this extra protocol as the referencing of the comb to the device that implements the $\sigma_x$ gates, which can itself be a laser or a microwave beam, in the case of hyperfine qubits.
§ D. FISHER INFORMATION AND SENSITIVITY

We are interested in estimating the sensitivity of the interferometric protocols that we have developed with respect to changes in the parameters they depend on. A measure of the information that one can extract about one or several parameters from a given probability distribution is the so-called Fisher Information [46, 47].

In our protocols, we want to estimate the precision of the pulses \( \theta \) (considered constant throughout the whole experiment) and the pulse-to-pulse phase difference \( \Delta \phi \). They will be related to some physical observables which measure the population of the excited state after applying certain protocols. The precision of the parameters \( \theta \) and \( \Delta \phi \) is determined by the fluctuations of these observables and their variance can be obtained using standard error propagation theory. The Fisher Information will yield a measure of the available precision in the estimation of the parameters. Also, the variance of the estimation of a given parameter will be limited by the Cramer-Rao bound [48], which sets the ultimate limit for the precision that we can achieve.

Let us see how to compute both the Fisher Information and the Cramer-Rao bound. Let \( X \) be a sample of observations with joint probability distribution given by \( P(X|k) \) depending on a vector parameter \( k = (k_1, k_2, \ldots, k_i)^T \) and \( h(k) \), a real valued function of \( k \). Then, under suitable regularity conditions (see [46, 47]), for any unbiased estimator \( \hat{h}(X) \) of \( h(k) \)

\[
\text{Var}(\hat{h}) \geq \delta^T [I(k)]^{-1} \delta
\]

where \( \delta \) is the vector of derivatives of \( h(k) \), i.e.

\[
\delta = \left( \frac{\partial}{\partial k_1} h(k), \frac{\partial}{\partial k_2} h(k), \ldots, \frac{\partial}{\partial k_i} h(k) \right)
\]

(17)

and the matrix \( I(k) \) is the Fisher Information matrix with \( (i, j) \)th element

\[
I_{ij}(k) = \mathbb{E}(S_i(X)S_j(X)) = -\mathbb{E} \left( \frac{\partial^2}{\partial k_i \partial k_j} \log P(X|k) \right)
\]

where \( \mathbb{E}(\cdot) \) denotes the expectation value and

\[
S_i(X) = \frac{\partial}{\partial k_i} \log P(X|k).
\]

(18)

(19)

In particular, if \( \hat{k}(X) \) is an unbiased estimator of the \( r \)th parameter \( k_r \), then, under the same regularity conditions

\[
\text{Var}(\hat{k}_r) \geq J_{rr}(k),
\]

where \( J_{rr}(k) \) is the \( r \)th diagonal element of the inverse matrix \( [I(k)]^{-1} \).

We have both used standard error propagation theory and computed the Fisher Information as explained before in order to get fundamental lower bounds and practical estimates of each of the proposed protocols, which we have shown in Table I of the main text. According to the theory [48], this implies the possibility of finding estimators of minimal variance for both \( \Delta \phi \) and \( \theta \).

§E. EXPERIMENTAL ERRORS

Dephasing

We want to clarify with greater detail our estimates of the errors induced by small detunings and energy shifts. Our starting point is the Rabi model in the RWA, but now with an extra field,

\[
H_e = \frac{\epsilon}{2} \sigma_z + s(t) \left( e^{-i\phi_m} \sigma^+ + \text{H.c.} \right).
\]

(20)

\( \epsilon(t) \) may be random but always so smoothly varying, \( \frac{d}{dt}\epsilon(t) \ll \tau^{-1} \), that it may be assumed constant over a single pulse. If we make an interaction picture over this field, the result is that the phases of the pulses change in time

\[
H_{e,1} = s(t) \left( e^{-i\hat{\phi}_m} \sigma^+ + \text{H.c.} \right),
\]

(21)

where the new phase depends on the arrival times of the pulses, \( t_m \), as

\[
\hat{\phi}_m - \hat{\phi}_{m-1} \simeq \phi_m - \phi_{m-1} + \int_{t_{m-1} - \tau/2}^{t_m + \tau/2} \epsilon(t)dt.
\]

(22)

On average, such a field contributes to the phase difference an amount which is of the order \( \sigma_e (t_m - t_{m-1}) \), where \( \sigma_e \) is the standard deviation of these random errors (assuming they average to zero). We can thus upper bound the error in our phase estimation simply by decreasing the interval between pairs of consecutive pulses. This fine tuning is quite well suited for protocols 2A an 2B in the manuscript, where we are free to control the time separation between one pulse, \( t_{2k} \), and the pulse coming from the delayed beam, \( t_{2k+1} \).

Temperature

Temperature can also induce dephasing: if the atom is not still enough and it has time to move between pulses, it will see different phases of the pulse which depend on the distance traveled as \( 2\pi \Delta x/\lambda \). There are various ways to address this issue. We can make a simple estimate for a free atom in space, assuming that it is Doppler cooled. The temperature of the atom is

\[
k_B T \simeq \hbar \Gamma,
\]

(23)
where $\Gamma$ is the natural linewidth of the cooling transition. Let us pessimistically assume that all this energy goes to the kinetic part, $\frac{1}{2}mv^2$, giving us an average velocity

$$v \simeq \sqrt{\frac{2k_BT}{m}} \simeq \sqrt{\frac{2\hbar\Gamma}{m}}.$$ (24)

From this we can estimate the phase errors as

$$\delta \phi \simeq \frac{2\pi}{\lambda} v(t_m - t_{m-1}).$$ (25)

Also pessimistically we will take $\Gamma \approx 200$ MHz, and a light atom such as Be, obtaining $v \sim 5$ m/s, which for a pulse separation of 10 ps gives $10^{-3}$ (or actually a bit larger if we consider the additional velocity due to the photon recoil).

The situation is very much improved when we arrange the lasers to work in a co-propagating Raman configuration such that there is no net momentum transfer to the atom. This is indeed a solution to the previous problem for, on each pulse, the spatially dependent phase which is acquired from one laser, $\vec{k}\vec{x}$, is cancelled by that of the other laser, $-\vec{k}\vec{x}$, making the whole process independent on the position of the atom. In this favorable circumstance, the only effect that temperature may have is when the intensity of the laser varies spatially. However, by using a laser beam which is not too tightly focused and confining the atoms to a small region, the effect of this inhomogeneity may be safely neglected.

\section*{F. EXPERIMENTAL SENSITIVITIES WITH TRAPPED IONS}

As mentioned earlier, trapped atomic ions \cite{2} constitute one of the most mature systems in the implementation of Quantum Information Processing and Communication (QIPC) protocols and technologies. Precision records have been achieved in the realization of single-qubit \cite{4} and two-qubit \cite{5} unitaries and measurements \cite{6, 49}, even reaching the threshold for fault-tolerant quantum error correction protocols \cite{50}. In the last couple of years, fantastic progress in the controlled interaction between trapped ions and ultrafast lasers has been achieved by the group of C. Monroe at U. Maryland, with the demonstration of two qubit entanglement in the weak-field ($\theta \ll 1$), many-pulses regime \cite{44}, and also in the strong-field ($\theta \sim \pi$), few-pulses regime \cite{41}, where logic gates faster than the trap oscillation period become accessible \cite{40, 51}. Because of this, we think that the technology required to implement the phase-stabilization protocol that we propose is already available.

Typical parameters for the frequency comb are listed in Table II. Pulses with a duration $< 1$ ps are nowadays easily accessible. On the other hand, one has to keep in mind that a pulse duration $\tau$ effectively limits the possible pulse delay times to $T_d > \tau$ in order to avoid an overlap of the electric fields corresponding to different pulses:

\begin{table}[h]
\centering
\begin{tabular}{|c|c|c|c|}
\hline
$f_{\text{rep}}$ (MHz) & $T$ (ns) & $\tau$ (ps) & $T_d$ (ps) \\
\hline
100 & 10 & 10 & 10-100 \\
\hline
\end{tabular}
\caption{Typical parameters of a frequency comb \cite{40, 41}.}
\end{table}

depending on their polarizations, this may lead to several unwanted effects, from excitation of motional sidebands to total cancellation of the Raman transition \cite{40}; in either case, the action of a pulse pair on the qubit would still be described by a unitary transformation $U_i$, but not the ones we have written down earlier, so that our model would break down. Therefore, we will stick to a comb with pulses of 1-10 ps.

In the following, we present details of our calculations to estimate the achievable sensitivity enhancements for the protocols introduced in the main text of the article. At the end, we present an estimation of the ultimate sensitivity limit that can be reached with pulse shaping techniques.

\textbf{Sensitivity enhancements with two-level protocols}

Protocols 1A to 2B consider direct transitions induced only by the comb laser that we want to study. In practice, there are two ways that this can be achieved: dipole and quadrupole transitions. Dipole transitions are, for instance, the $^2S_{1/2} \rightarrow ^2P_{1/2,3/2}$ lines in Yb$^+$ \cite{45}. These transitions have a typical linewidth of a few tens of MHz, which is comparable to $f_{\text{rep}}$. This implies that the time between consecutive pulses could be shorter than the lifetime of the excited state of the ion. A possible solution to this problem will be presented later on in 5G.

An alternative is to rely on quadrupole transitions, as provided by the Ca$^+$ ion using as qubit states the electronic states $|S_{1/2}\rangle$ and $|D_{5/2}\rangle$ \cite{3}. The excited level now has a radiative lifetime $\tau_{\text{rad}} \sim 1$ s which is favorable to implement our ideas. The downside of quadrupole transitions is their lower coupling strength, which demands a more powerful laser to excite them. In practice, depending on the laser, this might imply that we have to work in the limit $\theta \ll 1$ (protocols 1A and 2A) but we will forget this in following discussion.

Following Table II, let us consider a frequency comb composed of 10-ps pulses with $f_{\text{rep}} = 100$ MHz. This pulse duration is much shorter than the trap oscillation period (1 $\mu$s for a typical $\omega_{\text{trap}} = 2\pi \times 1$ MHz rf Paul trap) and allows us to disregard the motional state of the ion in the trap [cf. Eq. (7)] as well as its micromotion, which may affect the performance of coherent protocols at longer times \cite{45}. Such a frequency comb, and a typical ion coherence time for the electronic qubit states of $^{40}\text{Ca}^+$, $\tau_{\text{coh}} \sim 10$ ms \cite{3}, allow up to $\tau_{\text{coh}}f_{\text{rep}} \sim 10^6$ pulses to go through the ion before decoherence becomes
relevant. Using protocol 1B, this leads to an enhancement of the sensitivity by a factor $\chi_{1B} = N = 10^6$, which translates in a stabilization of the frequency offset down to $\delta \nu_0 = f_{\text{rep}}/N \sim 1/\tau_{\text{coh}} \sim 1 \text{kHz}$. (In addition, a probe time of 10 ms combines well with ion state-detection times $\sim 10$ ms for an optimal duty cycle.)

This result can be improved by applying the protocols with two pulse sequences (2A,2B). To be specific, we can pair $N = 5 \times 10^3$ pulses with delays $N_d = 5 \times 10^3$ and reach, with protocol 2B, a resolution $\delta \nu = f_{\text{rep}}/(N N_d) = 0.4 \text{mHz}$.

Again, we remark that the numerical estimates in the main text of the paper and this supplementary material take into consideration only the coherence properties of trapped ions for the stabilization an “ideal frequency comb”, and technical issues inherent to currently available combs are not included in the calculations.

**Sensitivity enhancement with self-referenced Raman schemes**

Use of a Raman scheme lifts the restrictions related to the excited-state lifetime of the qubit as spontaneous decay is of no concern. Such a scheme has been implemented with various systems, e.g., Yb$^+$ with a qubit defined by the $^2S_{1/2}$ hyperfine states $|F = 1, m_F = 0 \rangle = |1 \rangle$ and $|F = 0, m_F = 0 \rangle = |0 \rangle$, which are split by $\omega_{\text{at}} = 12.642815 \text{ GHz}$ [40, 41]. For these states, coherence times larger than 1 second have been measured [42].

Let us consider a pulse train of 1 ms, which provides $10^5$ pulses at $f_{\text{rep}} = 100 \text{ MHz}$, and let us split this train on two lines. We seek to maximize the phase difference between them. To this end, we consider the available $10^5$ pulses into sets of $10^4$ and keep the first set, $S_1 = \{1, \ldots, 10^4\}$, the set $S_2 = \{10^4+1, \ldots, 2 \times 10^4\}$, and the last set, $S_3 = \{9 \times 10^4 + 1, \ldots, 10^5\}$. The first set, $S_1$, will be further split in two, so that half of the pulses are paired with those in $S_2$ ($N_{d1} = 10^4$) and the other half with $S_3$ ($N_{d2} = 9 \times 10^4$). Then, this optical setup yields a sensitivity enhancement of order $\chi = N|N_{d2} - N_{d1}| = 8 \times 10^8$ or $\delta \nu_0 \sim 0.1 \text{ Hz}$. If we allow ourselves a longer interrogation time of 1 s, the figures would improve down to an amazing precision of $10^{-7}$ Hz.

We note that these high sensitivities are achievable almost independently of the underlying physical system used for the qubit: taking into account the continuum spectrum of each pulse, the only requirement is the proximity of $\omega_{\text{at}}$ and $\tilde{\omega}$, a feature that can be engineered, and coherence times which are experimentally available.

**Recursive refinement for large pulse-to-pulse phase shifts**

In our studies we have found that the sensitivity of our metrology protocols can be written in the form $\sigma^{-1} \sim \chi(N)\sqrt{M}$, where the enhancement factor $\chi(N)$ arises from a clever accumulation of the phase. In practice, for a non-stabilized frequency comb with a large $\Delta \phi$ and an excessive number of pulses, the total accumulated phase, $\chi(N)\Delta \phi$, will wrap around the maximum measurable phase, $\pi$, precluding a unique determination of $\Delta \phi$.

The appropriate way to deal with this situation is to do an iterative refinement of the phase measurement. As an example, let us consider a fiber-based frequency comb: these devices have an intrinsic width of the offset frequency of about 200 kHz. This means that, for $f_{\text{rep}} = 100 \text{ MHz}$, the phase $\phi_m$ may wrap around $\pi$ in about $N = 500$ pulses. Hence, on the first iteration, it is meaningless to interrogate the laser for much longer than a few $\mu$s. This iteration allows us already to achieve a precision in $\Delta \phi$ of order $\sqrt{M}/500$ with protocol 1B. This initial value can be used to feed back to the laser setup, to lower $\Delta \phi$ and, on the next iteration, use a larger number of pulses.

Continuing with this example, a similar iterative refinement using protocol 2B and a fixed interrogation time $\sim 1 \mu$s, would lead to an accuracy in the comb offset frequency of $\delta \nu_0 = f_{\text{rep}}/(N N_d) = f_{\text{rep}}/250^2 = 3 \text{ Hz}$ using only one ion.

**§G. ULTIMATE PRECISION LIMITS WITH ADVANCED PULSE-SHAPING TECHNIQUES**

The protocols discussed so far achieve a great efficiency thanks to the number of pulses in a given interrogation time and possible delays among them. Note however, that the comb is mostly “empty”: between every two pulses of about 10 ps, there is a waiting time $\sim 10$ ns in which the ion is idle. It would seem that this empty time, combined with the coherence rates of the ions, set the ultimate limits for precision in our setup. However, if the laser has enough power, we can engineer a clever scheme to fill these empty gaps, increasing the effective repetition rate of the ion-laser interaction.

The trick here will be to “compress” the pulses so that a minimal time elapses between the end of one pulse and the beginning of the following one, but without modifying the phase of any one pulse. Such “compression” could be realized with an optical setup as depicted in Fig. 6b. The key ingredient in this setup is an optical device which we call Beam Splitter and Delayer (BSD) that, given an intense ultrashort pulse, extracts a train of $n$ replica pulses separated by a very short time $\Delta t$ [cf. Fig. 6a]. (An alternative BSD optical setup producing 8 replicas of an initial pulse has been recently implemented in [41].)

We will discuss these ideas in a particular application: doing metrology of the comb with a dipole transition. In this case the qubit of choice will not satisfy the condition $\tau_{\text{coh}} \gg T$. Consider for example Ca$^+$ ions using the dipole-coupled $|S_{1/2}\rangle$ and $|F_{1/2}\rangle$ states for which $\tau_{\text{rad}} \approx 7 \text{ ns}$ [52]. In this setup we can still reach high precision...
taking a relatively long pulse train of duration $\gg \tau_{\text{coh}}$ as long as we ensure that all the pulses pass through the ion within a short time $\lesssim \tau_{\text{coh}}$. On the other hand, we must still fulfill the requirement that different pulses do not overlap in time, that is $\Delta t \geq 2\tau$, which restricts us to use sets of up to $n \leq \min\{\tau_{\text{rad}}, \tau_{\text{coh}}\}/(2\tau) \sim 7$ ns/20 ps = 350 replica pulses. To be concrete, let us use the setup in Fig. 6b to pick up two pulses with a relative delay of $T_d = N_d T = 10 \mu s$ —this corresponds to pulses 1 and $k = N_d = 10^3$ in the previous figure. The pulses will go through the BSD and be recombined, alternating replica pulses from each line. For a conservative $n = 4$ (not to lose too much power in each replica), $\Delta t = 4\tau$, and $N_d \approx 1000$, we obtain a phase sensitivity enhancement by a factor $n N_d \approx 4 \times 10^3$.

The same ideas can be applied to the Raman scheme by ensuring that the replica pulses from both lines arrive simultaneously to the ion. The result is an enhancement of the sensitivity by an additional factor $n$ on top of the formulae derived in the main text of the paper.

We finally note that the very short probe times considered here, allow for the recollection of a large set of statistical data in a very short time. Together with the large sensitivity enhancements calculated, the presented schemes appear as very competitive protocols to measure and stabilize the carrier-envelope offset phase of frequency combs without the need for octave-spanning spectra.