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Phase locking a clock oscillator to a coherent atomic ensemble

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The sensitivity of an atomic interferometer increases when the phase evolution of its quantum superposition state is measured over a longer interrogation interval. In practice, a limit is set by the measurement process, which returns not the phase, but its projection in terms of population difference on two energetic levels. The phase interval over which the relation can be inverted is thus limited to the interval $[-\pi/2, \pi/2]$; going beyond it introduces an ambiguity in the read out, hence a sensitivity loss. Here, we extend the unambiguous interval to probe the phase evolution of an atomic ensemble using coherence preserving measurements and phase corrections, and demonstrate the phase lock of the clock oscillator to an atomic superposition state. We propose a protocol based on the phase lock to improve atomic clocks under local oscillator noise, and foresee the application to other atomic interferometers such as inertial sensors.

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From the first observations of Huygens on the coordinated motion of coupled nonlinear oscillators [1], phase synchronization has evolved to an indispensable tool for time and frequency metrology and starts to be investigated for quantum systems [2–4]. Phase lock loops (PLLs) [5], where a local oscillator (LO) is phase locked to a reference signal, are widely used for the generation of atomic time scales [6, 7], the synchronization in telecommunication [8] or in radio navigation [9]. On the contrary, nowadays in atomic frequency standards, only the frequency of the local oscillator is locked on the atomic resonance. This is directly linked to the quantum nature of the reference system, *i.e.*, the quantum superposition of two internal states of an ensemble of atoms, molecules or ions, which is destroyed by the detection at the end of the interrogation process. A similar limitation exists for any measurement of a quantum system as in magnetometers or inertial sensors [10]. Adopting PLLs in such systems will improve their long term stability, as the phase is the integral of the frequency, and reduce the constraints on the stability of the LO or of the measured signal. Here, we demonstrate the direct phase lock of a LO to an atomic ensemble, based on repeated coherence preserving measurements of the atomic ensemble. We also study how this technology could improve atomic clocks subject to local oscillator noise.

Phase locking a classical system to a quantum system would give a direct link in metrology to the fundamental oscillations of quantum particles and could lead to enhanced sensitivities and new applications in precision measurements. In an atomic clock, the frequency of a LO is repeatedly referenced to an atomic transition frequency by comparing their respective phase evolutions in an interrogation time T and applying a feedback correction. During the interrogation, the atoms are in a superposition state, and the projection of the relative phase

between the LO and the atomic ensemble is measured as a population imbalance of the two clock levels. This leads to a sinusoidal signal, and the phase drift can therefore only be unambiguously determined if it stays within the $[-\pi/2, \pi/2]$ interval, hereafter called inversion region. Hence, for a given LO noise, the interrogation time of the atomic transition must be kept short enough such that phase drifts beyond the inversion region are avoided. Currently, LO noise limits the interrogation time in ion [11] and optical lattice clocks [12–14] and is expected to become a limit for microwave clocks with the recently discovered spin-self rephasing effect [15]. The standard approach to tackle this issue consists in improving the quality of local oscillators [16–20]. As an alternative, it has been recently proposed to track and stabilize the LO phase evolution using several atomic ensembles probed with increasing interrogation time [21, 22], or by enhancing the Ramsey interrogation interval by stabilizing the LO either via cascaded frequency corrections [23] or by coherence preserving measurements on the same atomic ensemble and feedback [24, 25]. We follow the latter proposal, and show how the LO limitations can be bypassed by implementing a phase lock between the LO and the atomic system. We begin with a minimally destructive measurement of the LO phase drift when it is within the inversion region; the measurement readout is then used to correct the LO phase so as to reduce the phase drift. The cycle is repeated using the residual atomic coherence. This results in successive, phase related measurements of the relative phase evolution, and the feedback keeps the phase in the inversion region, leading to an effectively longer interrogation time. We demonstrate this approach with a trapped ensemble of neutral atoms probed on a microwave transition.

The experimental scheme shown in Fig. 1 has been described in [26]. A cloud of cold ^{87}Rb atoms is trapped

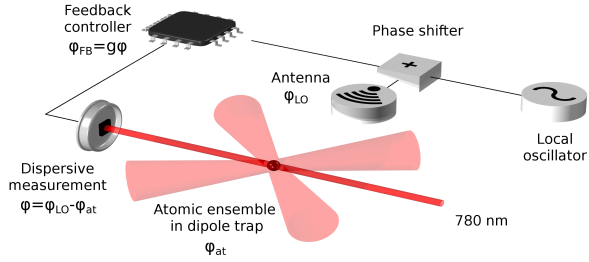


FIG. 1: **Experimental scheme.** The evolution of the LO phase φ_{LO} is compared to the phase φ_{at} of an atomic ensemble in a superposition state using a coherence preserving measurement in a Ramsey spectroscopy sequence. The relative phase is obtained from the read out of the population difference, and is used to implement the phase lock between the two oscillators by applying feedback on a phase actuator on the LO output. The light shift induced by the optical trap and by the probe has been engineered to have a homogeneous measurement on the atomic ensemble [26].

in an optical potential (see Appendix A), prepared with a $\pi/2$ pulse of a resonant microwave field in a balanced superposition state of two hyperfine levels $|\downarrow\rangle \equiv |F=1, m_F=0\rangle$ and $|\uparrow\rangle \equiv |F=2, m_F=0\rangle$ of the electronic ground state, and probed using a nondestructive detection. Typically, 5×10^5 atoms at a temperature of $10 \mu\text{K}$ are used in the measurements reported here. The phase φ_{at} of the superposition state oscillates at a frequency of 6.835 GHz corresponding to the energy difference between the $|\downarrow\rangle$ and $|\uparrow\rangle$ atomic states, which is the fundamental reference if atoms are protected from perturbations. A microwave LO has a frequency close to the atomic frequency difference, so that the relative phase $\varphi = \varphi_{\text{LO}} - \varphi_{\text{at}}$ between the two oscillators drifts slowly because of the LO noise. φ can be measured using the Ramsey spectroscopy method (see Fig. 2): a second $\pi/2$ microwave pulse (projection pulse) maps it onto a population difference, which we read out with a weak optical probe perturbing the atomic quantum state only negligibly and preserving the ensemble coherence [26–28]. Unlike for destructive measurements, the interrogation of φ can continue in a correlated way, once the action of the projection pulse is inverted using an opposite $\pi/2$ microwave pulse (reintroduction pulse), which brings the atomic state back to the previous coherent superposition. Moreover, after each measurement and reintroduction pulse, the phase read out can be used to correct the LO phase. The evolution and manipulation of the atomic ensemble can be illustrated using the Bloch sphere representation (Fig. 2): the collective state of N_{at} two-level atoms in the same pure single particle state (also called coherent spin state (CSS)) forms a pseudo-spin with length $J = N_{\text{at}}/2$, where J_z denotes

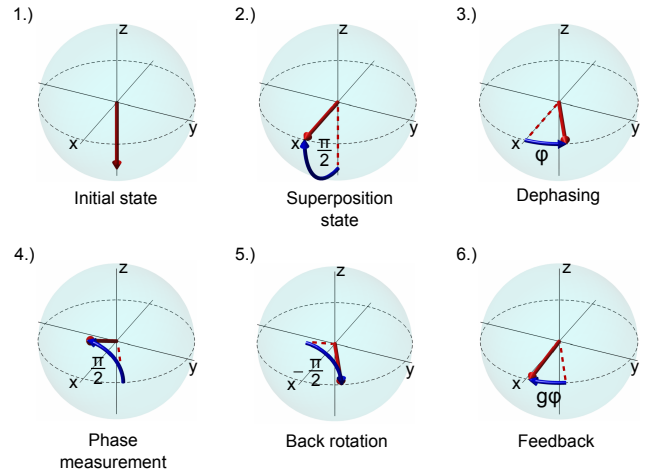


FIG. 2: **Bloch’s sphere representation of the phase lock between the LO and the atomic superposition state.**

The phase lock between the classical oscillator and the atomic spin is obtained using repeated, time correlated Ramsey interrogations and feedback. The sequence begins by preparing the atomic CSS in the $|\downarrow\rangle$ state via optical pumping (step 1.). The measurement of the relative phase between the CSS and the LO starts when a $\pi/2$ microwave pulse around the y axis brings the CSS into a balanced superposition of the $|\downarrow\rangle$ and $|\uparrow\rangle$ states, depicted as a vector on the equatorial plane of the Bloch sphere (step 2.). The x axis is chosen to represent the phase of the local oscillator, and φ the relative phase between the LO and the atomic superposition that evolves because of the LO noise (step 3.). After an interrogation time T , the projection of φ is mapped onto a population difference by a projection $\pi/2$ pulse around the x axis and read out with the coherence preserving detection (step 4.). The CSS is rotated back to the equatorial plane by a reintroduction $\pi/2$ pulse around the x axis (step 5.), and feedback is applied on the phase of the LO (step 6.). The PLL between the LO and the atomic ensemble consists in the repetition of the steps from 3.) to 6.), potentially till the atomic ensemble shows a residual coherence.

the population difference and $\varphi = \arcsin(J_y/J_x)$ is the phase difference between the phase of the LO and that of the superposition state. A resonant microwave pulse determines the rotation of \mathbf{J} around an axis in the equatorial plane of the Bloch sphere, and the axis direction is set by the phase of the microwave signal. The repetition of the manipulation, measurement and feedback cycle implements the phase lock of the LO on the atomic superposition state, as shown in steps 2.-6. of Fig. 2.

We first show that we can reconstruct the time evolution of the relative phase between the LO and the CSS by monitoring the population difference and without applying feedback. For this purpose, we frequency offset the LO by 100 Hz from the nominal resonance, and periodically measure the projection of the relative phase ($\sin(\varphi)$) with only a small reduction of the atomic ensemble coherence (Fig. 3). Every 1 ms φ is mapped to a population difference via a projection $\pi/2$ microwave

pulse around the x-axis, a weak measurement of J_z is performed, and the collective spin is rotated back to the equatorial plane of the Bloch sphere via a reintroduction $\pi/2$ pulse around the x-axis. The $\pi/2$ microwave pulses are derived from an amplified version of the LO at 6.835 GHz leading to a pulse length of $\tau_{\pi/2}=47 \mu\text{s}$, and the rotation axis is controlled with a quadrature phase shifter. The coherence preserving, dispersive measurement relies on frequency modulation spectroscopy (see Appendix B). Fig. 3 shows in a single experimental run how the spin state evolves around the equator of the Bloch sphere, with the phase φ mapped on the normalized population difference J_z/J . The S/N of the weak measurements is 20 for a full state coherence and each readout of the relative phase drift reduces the state coherence by 2%. The destructivity from the probe is the main decoherence source till 10 ms, then the inhomogeneous light shift of the dipole trap on the clock states becomes the dominant decoherence source.

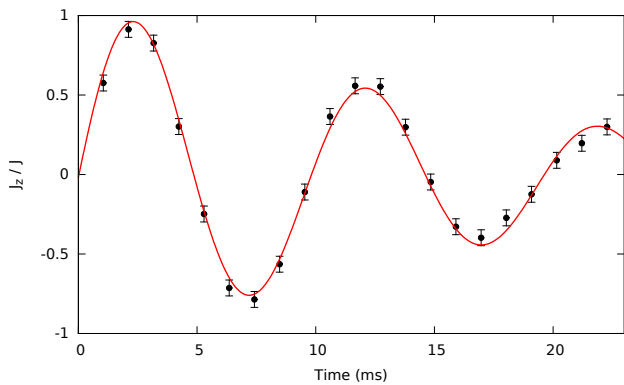


FIG. 3: Coherence preserving measurement of the relative phase between the LO and the atomic superposition. Real-time measurement of the vertical spin projection to which the relative phase φ between the local oscillator and the atomic superposition state is periodically mapped via microwave rotation pulses. The phase precession is induced by setting the LO frequency 100 Hz off the nominal atomic transition frequency. The experimental points are fitted with a sinusoidal evolution, damped by the effect of the J_z measurement and by the residual differential light shift on the clock transition.

We next introduce feedback and demonstrate that we can phase lock the LO on the atomic superposition state, and increase the Ramsey interrogation time beyond the limit set by the inversion region between J_z and the relative phase. We apply on the local oscillator two types of signals, first a frequency offset, and second periodic phase jumps, and use the output of the coherence preserving measurements to actively minimize φ . The phase lock is obtained by controlling the phase of the local oscillator by means of a digital phase shifter (see Appendix D). The feedback is performed after the atomic spin is rotated back to the equatorial plane of the Bloch sphere. When the disturbance applied on the local oscillator con-

sists of a frequency offset, there is a linear phase drift between the LO and the atomic phase (Fig 4, red). The phase evolution in open loop is reconstructed from the data of Fig. 3 by taking into account the damping on the sinusoidal signal due to the decoherence sources, and knowing that a constant frequency offset is applied on the LO. We remark that sudden sign inversions of the applied frequency offset when $\varphi = \pm\pi/2$ would produce exactly the same evolution of the population difference, illustrating the need to keep φ in the inversion region. In closed loop, the phase drift due to the 100 Hz frequency offset on the LO is periodically reset to zero, with a precision set by the $\pi/32$ step size of the digital phase shifter and the uncertainty of the coherence preserving measurements. This results in a saw-tooth-like signal for J_z/J (Fig. 4, blue signal). Without phase lock, the phase drift leaves the inversion region after 2.5 ms and rotates several times around the Bloch sphere, whereas with phase lock it stays in the inversion region for all the 22 ms interval shown in the image. When the feedback is active the total phase drift results as the phase measured at the end of the Ramsey interferometer, added to the correction phase shifts on the LO via the feedback controller. We next apply periodic phase jumps of $\pi/3$ back and forth on the LO using a second phase shifter. The signal obtained in open loop is shown at the top of Fig. 5. When the feedback controller is active, the jumps detected on the relative phase are corrected to zero, with a precision set by the resolution of the phase shifter and the uncertainty of the weak measurements (Fig. 5, bottom). The solid lines in Fig. 4 and 5 are drawn from the known timing for the applied phase signal and the feedback on the phase. For a combination of a phase drift and phase jumps, the relative phase can leave the inversion region while the noise action cannot be predicted from previous measurements. This is the commonly encountered situation in atomic clocks, and highlights the requirement of feedback on the LO phase to keep track of the relative phase drifts. Without feedback, the Ramsey interrogation time should be kept sufficiently short to avoid ambiguities for the measured phase shift.

We propose now a protocol to efficiently use the phase lock to improve an atomic clock. In a conventional atomic clock, the phase drift φ is destructively read out after a single interrogation and feedback is performed on the LO by the addition of a frequency $\omega_{\text{FB}} = -\varphi/T$ considering unity gain. Our protocol of using the PLL between the LO and the atomic superposition state in an atomic clock is based on the reconstruction of the phase drift experienced by the LO over the extended interrogation time $T_{\text{tot}}=N \times T$ (N is the number of phase coherent interrogations) by combining the phase shifts applied by feedback and the final phase readout (Fig. 6). The known phase corrections applied to the LO phase serve the dual purpose of keeping the relative phase in the inversion region and giving a coarse estimate of the phase drift during T_{tot} . The final phase measurement φ_f , together with the phase corrections $\varphi_{\text{FB}}^{(i)}$, gives a precise

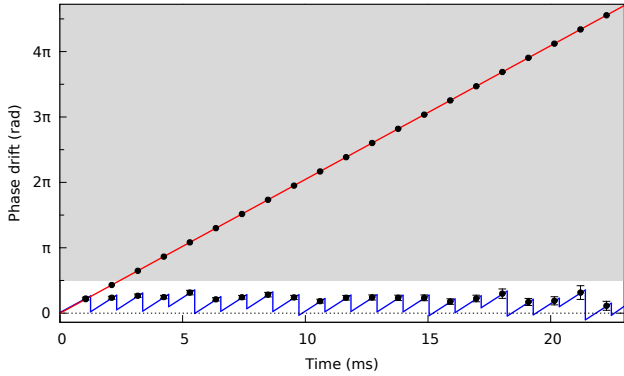


FIG. 4: **Phase lock between the LO and the atomic superposition state.** The evolution of the LO-atom relative phase is reconstructed from the J_z signal in Fig. 3 (red, solid line) and when feedback is applied on the phase of the LO after each measurement (blue, solid line). In the open loop case, the points are reported at the values given by the fit in the previous figure. In the closed loop case, the relative phase is always smaller than $\pi/2$, and does not enter in the region represented in grey where it cannot be univocally determined from the measurement.

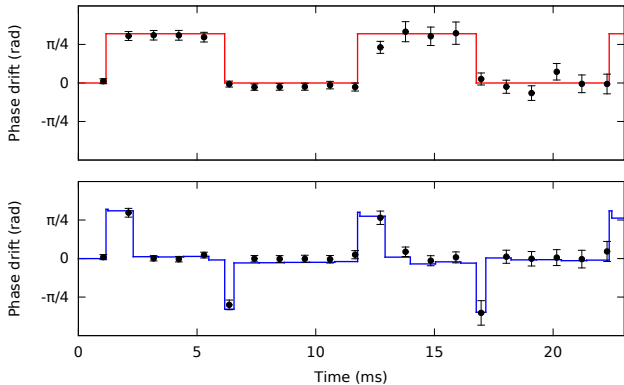


FIG. 5: **Correction of phase jumps between the LO and the atomic ensemble.** Evolution of the LO-CSS phase when periodic phase jumps of $\pi/3$ are applied back and forth on the LO: the points are obtained from the measured population difference, whereas the solid line represents the LO phase measured after the correction phase actuator. The servo controller, implemented using coherence preserving measurements and feedback on a phase actuator, maintains the relative phase close to zero. Above the system is operated in open loop, below in closed loop. In the latter case, the phase corrections are applied 150 μ s after each measurement.

estimate of the phase drift during T_{tot} . The total phase drift can be computed as $\varphi_{\text{tot}} = \varphi_f - \sum_{i=1}^N \varphi_{\text{FB}}^{(i)}$, where the sum is over all the correction phase shifts applied by the feedback controller and stored in the microcontroller during the sequence. The feedback on the frequency is set accordingly to be $\omega_{\text{FB}} = -\varphi_{\text{tot}}/T_{\text{tot}}$. After the final phase read out, the phase shifter is reset to its initial position to avoid any impact of the intermediate phase shifts

on the long term clock stability. The measurement S/N of the total phase drift depends only on the final phase measurement, since it can make use of all the residual ensemble coherence, whereas the S/N of the intermediate measurements has to be only sufficient to keep the atomic state in the inversion region. To further improve the protocol, the clock operation could be optimized by adapting the destructivity of the intermediate measurements to the varying coherence of the atomic sample, or implementing an adaptive measurement protocol for the last measurement [29].

For a proof-of-concept demonstration, we run an atomic clock that exploits the PLL between the LO and the atomic superposition state, and the phase reconstruction protocol. The LO signal was deteriorated so as to have an increased phase drift over the interrogation interval (see Appendix C). During the clock operation, the dead time for the preparation of the new ensemble is $T_d = 1.9$ s and the interrogation time is set to $T = 1$ ms, much shorter than the measured atomic coherence lifetime. As a benchmark, we first run an atomic clock adopting a standard Ramsey interrogation sequence, and without any feedback on the phase. The phase measurement is performed with the coherence preserving detection adopted for the phase lock. The two-sample Allan frequency standard deviation was calculated from the sum of the LO noise and the correction signal applied by the feedback controller. The clock instability reaches a $\tau^{-1/2}$ scaling after a few clock cycles (see Fig. 7, red) at a level consistent with an initial S/N = 20 and considering the coherence decay in the optical trap. The instability is far higher than with state-of-the-art atomic clocks since the experimental setup was not explicitly designed for the operation of an atomic clock, and the Allan frequency standard deviation is 1.5×10^{-9} at 1 s. We then operate an atomic clock making use of a PLL sequence with $N=9$ successive interrogations, and again with $T=1$ ms. For simplicity, the intermediate and the final phase readouts are set to have the same measurement strength. The Allan frequency deviation shows a $\tau^{-1/2}$ scaling as expected for atomic clocks, which demonstrates that the phase reconstruction protocol is working properly. In the opposite case, frequency offsets would be corrected only at a short time with the phase actuator, and the stability of the clock would then diverge because of the non-zero dead time interval. The comparison of the instability shows that the clock adopting the phase lock and reconstruction method is at a lower level by a factor (4.76 ± 0.25) with respect to the clock implementing the standard Ramsey interrogation, as shown in Fig. 7 (blue). This is clearly above the factor 3 expected for a succession of 9 uncorrelated phase drift measurements at each clock cycle [30]. Nevertheless, several detrimental effects prevent us to reach the factor 9 expected for perfectly correlated measurements of the phase drift. The main effects are the reduced S/N due to the cumulated destructivity from the probe and the decay from the optical dipole trap, and the finite phase shifter accuracy,

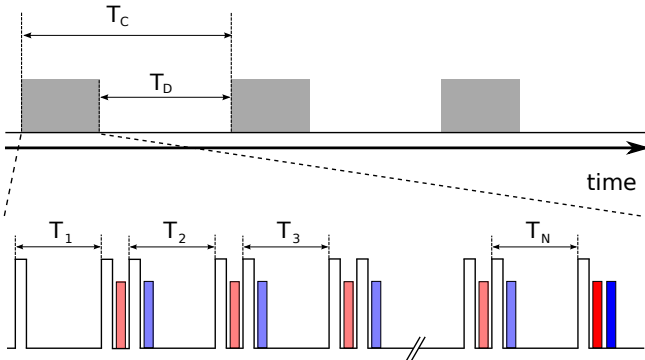


FIG. 6: **Protocol for atomic clock with PLL.** Operation of the clock: at each cycle of duration T_C , the relative phase is repeatedly measured in a coherence preserving way during the phase lock interval (above: shaded grey areas). Each interrogation, represented in the inset by a light red peak between the manipulation $\pi/2$ pulses, is followed by a phase correction $\varphi_{\text{FB}}^{(i)}$ on the LO, represented in the inset by a light blue peak in the right after the reintroduction of the spin on the equatorial plane of the Bloch sphere. The final phase readout φ_f (dark red peak in the inset), whose S/N is set by the residual coherence, together with the previously applied phase shifts on the LO, provides the total phase drift φ experienced during the extended interrogation interval $T_{\text{tot}}=N \times T$. The interrogation sequence ends with the application of a frequency correction on the LO (dark blue peak in the inset), then a new atomic ensemble is prepared in the dead time interval T_D for the next cycle.

equal to 2.2° .

The phase lock can be performed as long as the coherence of the state is maintained. For integration times longer than the coherence lifetime of the trapped ensemble, the atomic phase is lost and our locking scheme becomes again a frequency lock, like when the quantum superposition is destroyed by the detection. In our experiment, the coherence lifetime is limited to 20 ms by the dephasing in the optical dipole trap. Nevertheless, trapped induced dephasing of the atomic state can be suppressed for ^{87}Rb as reported in [15, 31], whereas in an optical lattice it is strongly reduced with the choice of light at the magic wavelength [32]. In the original proposal to lock the local oscillator phase on the atomic phase [24], frequency feedback on the local oscillator after each weak J_z measurement is performed. This leads to a longer effective interrogation time, but to a S/N given by the weak measurements, which is lower than that of a measurement at the quantum projection noise and beyond. Our protocol can overcome this limit, and reach projection limited readout while keeping an extended interrogation time, thanks to the feedback on the LO phase.

In the phase lock sequence, several effects must be considered to maximize the S/N of the last measurement while maintaining a high accuracy on the total phase drift over the increased interrogation time: the rotations

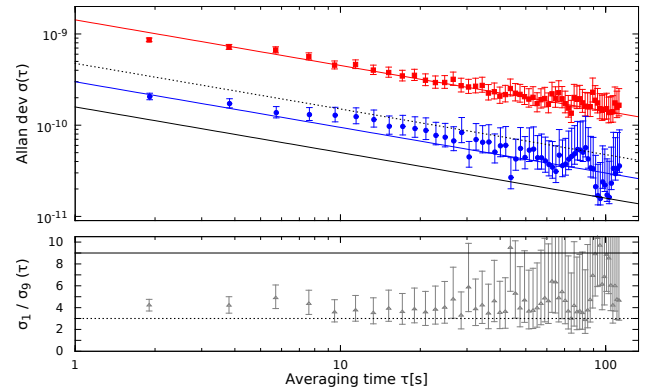


FIG. 7: **Atomic clock implementing a PLL.** (Top) Allan frequency standard deviation σ_1 for a normal Ramsey clock with interrogation time $T=1$ ms (red) and σ_9 for a clock implementing the phase lock between the LO and the atomic superposition state for 9 successive, correlated interrogations on the same atomic ensemble, for a total interrogation time of $9 \times T=9$ ms (blue). The red and blue lines are fits to the data, with a slope set to $\tau^{-1/2}$. The dashed black line lies a factor $\sqrt{9}$ below the red curve, and represents the best achievable level if 9 uncorrelated measurements are implemented at each clock cycle. The continuous black line lies a factor 9 below the red curve, and represents the best achievable level if 9 correlated measurements are implemented at each clock cycle. (Bottom) The grey triangles represent the ratio of the Allan deviation for the two clocks; its value is higher than 3 (black dashed line), which is the maximum achievable by realizing at each clock cycle 9 uncorrelated measurements of the phase drift. The black solid line represent the maximum ratio achievable for 9 correlated measurements.

operated on the Bloch sphere must be fast, the measurement induced decoherence limited, and the phase shifter used for the correction accurate. The decoherence related to the repeated interrogations of the relative phase can be strongly reduced by the use of an optical cavity to enhance the probe interaction with the atomic ensemble [33–35]. An optimized clock configuration would consist in a two atomic ensembles using the same LO: the first ensemble provides the information to implement the phase feedback algorithm on the LO; the resulting corrected phase for the LO stays in the inversion region for a much longer period, and this prestabilized LO is used to interrogate the master ensemble with the standard Ramsey sequence. This scheme avoids the requirement of a trade-off between the number of intermediate measurements and the S/N of the final measurement by separating the two problems. The solution promises the same benefits foreseen for the phase reconstruction schemes proposed in [21, 22], but using only a single additional ensemble.

In an atomic clock the phase lock between the LO and the atomic superposition state can reduce the Dick effect [36], *i.e.* the aliasing of the clock oscillator, thanks to the longer interrogation time. However, the most important advantage of the scheme is the reduction of the decoher-

ence related to the local oscillator, which translates to a lower white noise frequency for a fixed detection noise. This can serve to lower the LO stability requirements to the benefit of other parameters, like portability of the experimental setup, or viceversa, to remove the limitation set by the LO to reach ultimate performances. In the latter case, other effects will limit the clock interrogation time. Practically, a first limit is set by the vacuum quality, which can reduce the ensemble coherence via background collisions, but extremely long trapping lifetimes have been already reached for trapped atoms [37]. Ultimately, with the combination of existing techniques and the method presented in this paper, the 1–10 mHz excited states lifetime expected for alkaline earth-like atoms could be reached, which motivates the search for transitions with a lower linewidth [38–40].

Phase locking the LO to the atomic state preserves classical correlations in time against the decoherence by the local oscillator. The technique and the related enhancement factor could thus be combined to spin squeezing, which improves the clock sensitivity by introducing quantum correlations between the particles to go below the standard quantum limit [35, 41]. More generally, increasing the interrogation time using minimally destructive measurements and feedback on the phase could be applied to other atomic interferometers, such as atomic inertial sensors, where for example the phase of the interrogation lasers could be locked to the phase evolution of matter waves.

In conclusion, using a coherence preserving detection we tracked the phase evolution of an atomic collective superposition, and we reproduced it on a classical replica by introducing feedback to implement a PLL. Phase locking the LO to the atomic superposition state can be a key technology to improve the sensitivity of systems where the atomic phase evolution is compared to that of a classical LO, such as in atomic clocks and inertial sensors, whenever the main decoherence source is determined by the classical subsystem. This development may open new directions and possibilities in several technological fields and as well in basic science.

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not constitute an endorsement.

Appendix A: Atomic sample preparation

^{87}Rb atoms laser cooled with a magneto-optical trap are transferred to an optical dipole trap at 1560 nm, that uses a 4 mirror optical resonator to enhance the laser intensity [42]. The atoms are trapped at the crossing of two cavity arms with a waist of 100 μm . The ensemble is evaporatively cooled by decreasing the intensity of the dipole trap till a temperature of 10 μK is reached for 5×10^5 atoms in a cloud with $1/e^2$ radius of 50 μm . In the last operation before starting the Ramsey interrogation the atoms are optically pumped in the $|\downarrow\rangle$ state. The sequence to prepare the ensemble in the initial state, which corresponds to the dead time T_d in the atomic clock sequence, lasts 1.9 s.

Appendix B: Non-destructive dispersive probe

The measurement of J_z is based on the dispersion caused by the trapped atoms on a far off-resonance optical probe. The probe beam has a waist of 47 μm matched to the size of the atomic cloud. It is phase modulated at a frequency of 3.853 GHz and frequency referenced at 3.377 GHz on the red of the the $F=1 \rightarrow F'=2$ transition; these conditions produce a symmetric mixing of the $|\downarrow\rangle$ and $|\uparrow\rangle$ states because of probe induced spontaneous emission, thus avoiding a vertical offset when the Bloch sphere contracts. In this way, each sideband mainly probes the population of one of the two levels, with the same magnitude and opposite sign for the couplings. We cancel the probe induced light shift and the related decoherence by precisely compensating the effect of the carrier with that of the sidebands; this is obtained by setting a modulation depth of 14.8% for the phase modulation. The differential light-shift on the D2 line from the dipole trap at 1560 nm was compensated with light blue detuned from the $5^2P_{3/2} \rightarrow 4^2D_{5/2,3/2}$ transitions at 1529 nm. When the total power of the probe is set to 480 μW , it causes the decay of the atomic coherence with a lifetime of 2.85 μs ; in the experiment here reported the interrogation pulses, obtained using an amplitude electro-optic modulator, have been set to last 60 ns. This determines for each pulse a 2% destructivity of the ensemble coherence, and a S/N of 20 for the J_z measurement on the initial sample of 5×10^5 atoms. The population imbalance read-outs have been normalized to the signal when all atoms were repumped to the state $F=2$.

Appendix C: Frequency chain

The 6.835 GHz frequency used to coherently manipulate the atomic spin is generated by a frequency chain

based on a Spectra Dynamics DLR-100 system as a frequency reference. The DLR-100 relies on an ultra-low noise 100 MHz quartz, locked at low frequency to the 10th harmonic of a frequency doubled 5 MHz quartz to further improve the phase noise. The 100 MHz signal is multiplied to 7 GHz and then mixed with a tunable synthesizer at 165 MHz to obtain the signal resonant with the transition between the $|\downarrow\rangle$ and $|\uparrow\rangle$ state. Frequency noise is added to the LO signal using a frequency modulation port on the synthesizer, with a conversion factor set to 200 Hz/V_{rms}. The noise signal for the demonstration of the clock using the PLL sequence is generated with a signal generator, which produces white frequency noise with a spectral density of 2.7×10^{-2} Hz²/Hz; this signal is low pass filtered at 1.85 kHz before being added to the LO. This results in a rms phase drift of 430 mrad over 10 ms for the LO.

Appendix D: Feedback controller

The atomic populations on the $|\downarrow\rangle$ and $|\uparrow\rangle$ states determine a differential phase shift of the probe sidebands. This results in an amplitude modulation of the beam, which is detected by a photodiode (1591NF, New Focus), amplified (two HMC716LP3E, Hittite) and demodulated (ZX05-73C-C+, Minicircuits). The electronic integration of this signal during the 60 ns interrogation pulse produces a voltage proportional to the average atomic population difference during the probing. Such a voltage is digitized by a 14 bit resolution analog-to-digital converter embedded in the microprocessor (MCU) used to control the feedback loop (ADuC841, Analog Devices). In the experiments implementing the phase lock between the LO and the atomic superposition the MCU controls the phase actuator, a 6 bits step phase shifter with a range of 2π (RFPSHT0204N6, RF-Lambda). The total delay for the feedback is approximately 150 μ s depending on the calculation time of the MCU. When running the clock based on the PLL technique, the MCU acts as well on a frequency actuator, which is the frequency modulation input on the 165 MHz synthesizer.

The feedback controller we propose for the clock exploiting the phase lock between the LO and the atomic superposition state consists of a cycle divided in three main steps: in the first one, successive correlated interrogations with probe interval T are realized on the same coherent atomic ensemble, and feedback on the LO phase

is applied. The control law is

$$\begin{aligned}\varphi_{\text{LO}}^{(i)} &= \varphi_{\text{LO}}^{(i-1)} + \varphi_{\text{FB}}^{(i)} \\ &= \varphi_{\text{LO}}^{(i-1)} + g_{\varphi}\varphi^{(i)}\end{aligned}$$

where $\varphi^{(i)}$ is the estimated phase difference between the LO and the atoms at the i -th cycle and $\varphi_{\text{LO}}^{(i)}$ is the phase of the LO only. The values of $\varphi_{\text{FB}}^{(i)} = g_{\varphi}\varphi^{(i)}$ are saved in the feedback controller and typically a gain $g_{\varphi} = -1$ is chosen.

The second step consists in the final phase readout: at the end of the N -th interrogation interval of duration T, a destructive measurement φ_{f} of the phase is performed, with the highest possible precision. The saved phase shifts on the LO and φ_{f} are used to reconstruct the full phase drift between the LO and the atoms in the total interrogation time, equal to $T_{\text{tot}} = N \times T$.

In the third step, one then performs feedback on the frequency as in a conventional atomic clock

$$\begin{aligned}\omega_{\text{LO}}^{(n)} &= \omega_{\text{LO}}^{(n-1)} + \omega_{\text{FB}}^{(n)} \\ &= \omega_{\text{LO}}^{(n-1)} + g_{\omega}\varphi_{\text{tot}}^{(n)}/T_{\text{tot}}\end{aligned}$$

where

$$\begin{aligned}\varphi_{\text{tot}}^{(n)} &= \varphi_{\text{f}}^{(n)} - \sum_{i \in n\text{-th cycle}} \varphi_{\text{FB}}^{(i)} \\ &= \varphi_{\text{f}}^{(n)} - \sum_{i \in n\text{-th cycle}} g_{\varphi}\varphi^{(i)}\end{aligned}$$

and n is the clock cycle. In addition, the phase shift set by the feedback controller on the LO is reset to zero

$$\varphi_{\text{LO}}^{(n)} = \varphi_{\text{LO}}^{(0)} = 0.$$

The cycle then repeats. The important feature of the feedback controller is that the feedback actions on the LO oscillator phase during the interrogation time are saved. They are then used with the output of the final precise measurement to determine the total phase drift. There is no drawback from the uncertainty of the weak measurements, since any feedback errors are detected with the precise final measurement at the end. As already remarked, in the experimental demonstration of the clock operation, we adopted the same probe for the intermediate and the final measurements.

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