Auto-Balanced Ramsey Spectroscopy

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We devise a perturbation-immune version of Ramsey’s method of separated oscillatory fields. Spectroscopy of an atomic clock transition without compromising the clock’s accuracy is accomplished by actively balancing the spectroscopic responses from phase-congruent Ramsey probe cycles of unequal durations. Our simple and universal approach eliminates a wide variety of interrogation-induced line shifts often encountered in high precision spectroscopy, among them, in particular, light shifts, phase chirps, and transient Zeeman shifts. We experimentally demonstrate auto-balanced Ramsey spectroscopy on the light shift prone $^{171}$Yb$^+$ electric octupole optical clock transition and show that interrogation defects are not turned into clock errors. This opens up frequency accuracy perspectives below the $10^{-18}$ level for the Yb$^+$ system and for other types of optical clocks.

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A measurement of a physical observable in a quantum system profoundly affects the system’s state. Within the limits set by quantum mechanics [1], precision measurements strive to minimize fluctuations and systematic errors due to measurement-induced perturbations. In this regard atomic clocks are a prominent example: With their ever-improving stability and accuracy [2] they have now reached a level of performance that renders previously negligible clock errors introduced with the spectroscopic interrogation highly relevant.

In this Letter we show that a balanced version of Ramsey’s method of separated oscillatory fields [3, 4] is well suited for measuring unperturbed transition frequencies of ultra narrow atomic reference transitions that suffer from significant frequency shifts due to the spectroscopic interrogation with the oscillatory drive pulses. Relying on simple common-mode suppression arguments we devise an auto-balancing scheme which unlike more specialized composite pulse proposals [4, 5] and other modified Ramsey techniques [10–12] provides universal immunity to all kinds of interaction pulse aberrations and associated systematic shifts including drive-induced AC Stark shifts (light shifts) [13, 14], phase chirps and other phase and/or frequency deviations [6], transient Zeeman shifts [15], and other pulse-synchronous shifts [17].

Using the example of the strongly light shift disturbed $^{171}$Yb$^+$ electric octupole (E3) optical clock transition at 467 nm [18] we experimentally demonstrate the advantages of auto-balanced Ramsey spectroscopy and show that no systematic clock errors are incurred for arbitrarily detuned or otherwise defective drive pulses. In addition we present in this context an experimental method addressing spectroscopy-degrading issues related to the motional ion heating [19] typically encountered in ion traps.

Our work is motivated by the prospect of exploiting the full potential of ultra narrow clock transitions without being limited by the detrimental consequences of very weak oscillator strengths and measurement-induced imperfections. In particular for the Yb$^+$ E3 clock this opens up the path to reproducible long-term frequency ratio measurements as discussed in searches for variations of fundamental constants [20–22], violations of Lorentz invariance [23], and ultra-light scalar dark matter [24].

Ramsey spectroscopy conceptually relies on the measurement of the relative phase $\Delta \phi$ accumulated in a superposition state $|g\rangle + |e\rangle e^{-i\phi(t)}$ over a free evolution time $T$ (often called dark time or Ramsey time). $\Delta \phi = EcT/\hbar$ with $\hbar$ being the reduced Planck constant, directly reflects the energy difference $E_c$ between excited state $|e\rangle$ and ground state $|g\rangle$. Ramsey spectroscopy compares this evolving atomic phase $\varphi(t)$ to the phase evolution $\phi_{LO}(t)$ of a local oscillator (LO), e.g., a microwave source or a laser [7]. The fact that the spectroscopically relevant phase information is acquired during an interaction-free period makes Ramsey’s protocol the natural choice when aiming to undisturbingly extract the transition frequency $\omega_{eg} = E_c/\hbar$. Nonetheless, the deterministic preparation of the initial superposition state and the subsequent phase readout which maps phase differences to directly observable population differences require interactions with the oscillatory drive pulses. Hence pulse defects (e.g., frequency deviations) and other interrogation artifacts potentially affect the outcome of the spectroscopic measurement leading to erroneous phase values and corresponding clock errors.

To further analyze this error propagation we adopt the standard 2-by-2 matrix based description of a coherently driven two-level system [1, 4]. A drive field oscillating with $\cos \phi_{LO}(t)$ connects the two energy eigenstates $|g\rangle$ and $|e\rangle$. Applying this field for a time $\tau = t_1 - t_0$ and neglecting counter-rotating terms converts an initial state $|i; t = t_0 = g0|g\rangle + e_0|e\rangle$ into a final state $|f; t = t_1\rangle = g1|g\rangle + e1|e\rangle$ via

$$ e^{i\delta'\tau/2} V[-\phi_{LO}(t_1)] U[\delta', \tau, \Omega_0] \ V[\phi_{LO}(t_0)] \begin{pmatrix} g0 \\ e0 \end{pmatrix}. \quad (1) $$

In this expression the unitary matrices $U$ and $V$ are defined as $U[\delta', \tau, \Omega_0] =$...
\[
\left( \cos \frac{\Omega \tau}{2} - i \frac{\delta}{\Omega} \sin \frac{\Omega \tau}{2} \quad i \frac{\Omega \tau}{2} \sin \frac{\Omega \tau}{2} \quad \cos \frac{\Omega \tau}{2} + i \frac{\delta}{\Omega} \sin \frac{\Omega \tau}{2} \right)
\]

(2)

and \( V[\xi] = \begin{pmatrix} 1 & 0 \\ 0 & e^{i\xi} \end{pmatrix} \) with Rabi frequency \( \Omega_0 \) and generalized Rabi frequency \( \Omega = \sqrt{\Omega_0^2 + \delta^2} \) characterizing the strength of the coupling between drive field and atomic transition. \( \delta = \omega_{\text{LO drive}} - \omega_{eg}' \) denotes the detuning of the oscillatory drive frequency \( \omega_{\text{LO drive}} \) from the instantaneous atomic resonance frequency \( \omega_{eg}' \). Primed variables identify quantities that due to interaction-induced shifts possibly deviate from their unprimed counterparts. For instance, in the case of the \(^{171}\text{Yb}^+\) octupole transition \( \omega_{eg} \) corresponds to the undisturbed true clock transition frequency; during the initialization and read-out Ramsey pulses, however, off-resonant coupling of the high intensity drive light to several dipole transitions outside the two-level system leads to a light-shifted instantaneous clock transition frequency \( \omega_{eg}' \) with an unknown light shift \( \Delta \omega_{eg} = \omega_{eg}' - \omega_{eg} \) easily exceeding \( \Omega_0 \).

In the following we consider a standard optical clock operation scenario where an ultra-stable laser is frequency-locked to the atomic reference transition via successive spectroscopic interrogations. The integrated outcome of the individual excitation attempts provides frequency feedback to the laser source.

Using the Ramsey scheme an ideal interrogation sequence starting from the atomic ground state is then comprised of three phase-continuously connected segments: First a drive pulse at frequency \( \omega_{\text{LO drive}} = \omega_{eg}' \) with \( \Omega_0 \tau = \pi/2 \) (i.e., a \( \pi/2 \)-pulse) initializes the atomic superposition state. Then over a free evolution Ramsey time \( T \) the local oscillator’s phase evolves with a rate \( \omega_{\text{LO}} = \omega_{eg} \) that matches the undisturbed atomic phase accumulation. During this interval the LO phase \( \phi_{\text{LO}}(t) \) is alternately incremented or decremented by \( \phi_{\pm} = \pm \pi/2 \) [29]. Finally another \( \pi/2 \)-pulse with \( \omega_{\text{LO drive}} = \omega_{eg}' \) is applied and subsequently the excited state population \( p_{\pm} \) is determined. The \( \phi_{\pm} \) modulation offsets the normally observed Ramsey fringe \( p(\delta) \) (i.e., the final excited state population as a function of Ramsey detuning \( \delta = \omega_{\text{LO}} - \omega_{eg} \)) by a quarter fringe period. Accordingly, the differential excited state population \( \tilde{p}(\delta) = p_{\pm}(\delta) - p_{-\pm}(\delta) \) can be interpreted as a derivative-like frequency error signal with a zero crossing at the Ramsey fringe center. Regulating \( \omega_{\text{LO}} \) such that \( \tilde{p} = 0 \) closes the experimental feedback loop and results in a locked local oscillator whose stability is ideally only limited by the signal-to-noise ratio of the population measurements.

However, \( \tilde{p} \) is not only a function of its explicit argument \( \delta \); as pointed out before, any aberration from the ideal interrogation sequence, e.g., drive pulse defects (frequency-wise quantified via \( \delta' \neq 0 \)) or pulse-synchronous variations of the magnetic field, also affect the outcome of the population measurements and might shift the error signal zero crossing point away from its undisturbed position. Therefore an apparent \( \tilde{p}(\delta) = 0 \) does not necessarily correspond to a true \( \omega_{\text{LO}} = \omega_{eg} \) but can rather imply an actual clock error \( \delta' \neq 0 \) whose magnitude will then approximately scale inversely with \( T \) [27].

FIG. 1: Ramsey interrogation with shift-inducing drive pulses. Unlike in the ideal case where \( \omega_{\text{LO drive}} = \omega_{eg}' \), the drive pulse frequency in (a) is assumed to be slightly higher than the light-shifted transition frequency \( \omega_{eg} \). Therefore, the LO phase evolves initially faster than the atomic superposition phase \( \phi(t) \). Over the following dark Ramsey time interval of variable duration \( T \) the acquired phase difference between LO field and atomic oscillator will remain unchanged (aside from the \( \phi_{\pm} \) modulation) if the LO phase advances with the unperturbed transition frequency \( \omega_{\text{LO}} = \omega_{eg} \) during \( T \). In this case the phase-to-population conversion performed by the second drive pulse will produce identical outcomes after short and long Ramsey times. (b) Relying on this \( T \)-invariant response an auto-balancing control scheme makes it possible to extract the unperturbed atomic transition frequency even with arbitrarily distorted drive pulses. The servo-controlled phase correction \( \delta' \) balances the short sequence and immunizes the interconnected LO-steering long sequence against drive-induced shifts.

To correctly reproduce \( \omega_{eg} \) it is essential to distinguish deviations of \( \tilde{p} \) due to \( \omega_{\text{LO}} \neq \omega_{eg} \) from misleading deviations caused by a flawed interrogation process. This discrimination is accomplished by exploiting the above mentioned dependance of \( \delta \) and thus \( \tilde{p} \) on \( T \): As shown in Figure 1(a), only for \( \omega_{\text{LO}} = \omega_{eg} \), i.e., only if LO phase and atomic phase evolve at identical pace, one can expect to find the same excited state population when comparing the outcome of the two “isomorphic” interrogation sequences which only differ in their dark times \( T_{\text{short}} \) and \( T_{\text{long}} \) (see also reference [25]).

Based on the simple insight that interrogation-induced \( \tilde{p} \) deviations are common mode for isomorphic Ramsey sequences it is now straightforward to combine \( T_{\text{short}} \) and \( T_{\text{long}} \) Ramsey interrogations yielding \( \tilde{p}_{\text{short}} \) and \( \tilde{p}_{\text{long}} \), respectively, into a defect-immune spectroscopy scheme: By using the differential population \( \tilde{p}_{\text{bal}} = \tilde{p}_{\text{long}} - \tilde{p}_{\text{short}} \) as an error signal for \( \omega_{\text{LO}} \) one obtains a passively balanced frequency feedback with \( \tilde{p}_{\text{bal}} = 0 \) exclusively for \( \omega_{\text{LO}} = \omega_{eg} \).
This approach, however, comes with a major drawback, that is to a lesser extent also encountered in recently proposed coherent composite pulse schemes [9, 10]: For \( \hat{p}_{\text{short}} \neq 0 \) the frequency discriminant \( \tilde{p}_{\text{bal}}(\delta) \) is no longer an odd function around \( \delta = 0 \), which leads to skewed sampling distributions and corresponding clock errors as explained in Figure 2. To avoid this issue we implement an active balancing process with two interconnected control loops as schematically displayed in Figure 1(b). The first feedback loop, acting on the short Ramsey sequence, ensures \( \hat{p}_{\text{short}} = 0 \) by injecting together with \( \phi^+ \) an additional phase correction \( \phi'^- \). Of course, this requires \( \delta' < \Omega_0 \) for sufficient fringe contrast. Depending on the dominant source of error one could also use \( \omega_{\text{LO} \text{drive}} \) as the control variable [29]. During \( T_{\text{short}} \) the local oscillator evolves with \( \omega_{\text{LO}} \) as determined via the second feedback loop which controls the long Ramsey sequence by steering \( \omega_{\text{LO}} \) so that \( \hat{p}_{\text{long}} = 0 \). Vice versa, \( \phi^- \) (or \( \omega_{\text{LO} \text{drive}} \)) as obtained from the short sequence is identically applied in the long Ramsey sequence. In this way a constantly updated common-mode correction autocalibrates the long interrogation whose outcome is then denoted by \( \hat{p}_{\text{auto}} \). As shown in Figure 2(c), auto-balancing prevents skewed sampling responses and leads to an always antisymmetric lock fringe \( \hat{p}_{\text{auto}}(\delta) \) which ensures that deviations \( \hat{p}_{\text{auto}} \neq 0 \) are solely due to \( \omega_{\text{LO}} \neq \omega_{eg} \).

We now report on an application of auto-balanced Ramsey spectroscopy with an ytterbium single-ion clock where the spectroscopic interrogation causes a particularly large perturbation of the atomic reference. So far, optical clocks operating on the 467 nm \( E3 \) transition in \( ^{171}\text{Yb}^+ \) have employed extrapolation methods [24, 30] or Hyper-Ramsey composite pulse approaches [8] to address the drive light induced AC Stark shift. Our experiments were carried out with the single \( ^{171}\text{Yb}^+ \) ion confined in an endcap-type radio frequency Paul trap [32] whose drive amplitude was adjusted to provide radial secular motion frequencies of \( \omega_r = 2\pi \times 1 \text{ MHz} \) and axial confinement with \( \omega_a = 2\pi \times 2 \text{ MHz} \). Laser cooling is performed on the \( ^2S_{1/2} \leftrightarrow ^2P_{1/2} \) cycling transition at 370 nm with repump lasers at 935 nm and 760 nm preventing population trapping in the metastable \( ^2D_{5/2} \) and \( ^2F_{7/2} \) manifolds [33].

While not actively cooled the trapped ion linearly gains motional energy [19] along each dimension, in our setup at a rate of 300 \( \text{kHz} \) per second. This large heating rate reduces the effective Rabi frequencies for Ramsey pulses applied after long dark times. To compensate for this effect one cannot simply change the laser light intensity because \( \omega_{eg} \) would change accordingly. Intensity-neutral \( \Omega_0 \) equalization is instead achieved by changing the spectral composition of the drive light with an electro-optic modulator that redistributes light from the carrier into sidebands spaced in our case \( \omega_{\text{EOM}} = 2\pi \times 2 \text{ GHz} \) apart. The value for \( \omega_{\text{EOM}} \) can be chosen over a wide range as long as the AC Stark shift’s spectral variation is insignificant and the added sidebands do not interfere with the resonant population transfer. Activating the modulator during the second Ramsey pulse of the short sequence and properly adjusting the modulation depth equalizes the short and long sequences and recovers their isomorphism. A modulation index of 0.6 rad was used to match the thermally averaged 10% reduction of \( \Omega_0 \) due to heating in the long sequence.

Except for heating compensation and interrogation specifics the optical clock is operated following the procedure outlined in reference [30]. To facilitate measurements with well-controlled detunings \( \delta' \) the LO laser (pre-stabilized to a high finesse optical cavity) is referenced to an independent \( \text{Yb}^+ \) \( E3 \) clock setup.

Figure 3(a) shows the results of auto-balanced clock runs with \( \Omega_0 = 2\pi \times 17 \text{ Hz} \) corresponding to a \( \pi/2 \)-pulse duration of 15 ms, \( T_{\text{short}} = 6 \text{ ms} \), and \( T_{\text{long}} = 60 \text{ ms} \). Using about 5 mW of drive laser light focused in a 50 \( \mu \text{m} \) diameter spot causes a large light shift of \( \omega_{eg} - \omega_{eg} = 2\pi \times 660 \text{ Hz} \). Within the statistical uncertainty no clock error is observed for drive detunings \( \delta' \) of up to 2\( \Omega_0 \). Beyond this detuning the response curve stays flat but the data points’ statistical uncertainties eventually increase due to the reduced fringe amplitude. In contrast, operating a Hyper-Ramsey interrogation (REFS) at \( \delta' > \Omega_0 \) yields...

![FIG. 2: Simulated Ramsey lock fringes \( \hat{p}(\delta) \) (top row) and corresponding rectified feedback responses \( R(\delta) = |\hat{p}P_0| \) for a resonant Gaussian sampling frequency distribution \( P_0(\delta) \) (bottom row). Solid, dashed and dotted lines represent fringes with \( \delta'/\Omega_0 = 0, \delta'/\Omega_0 = 0.1 \) and \( \delta'/\Omega_0 = 0.2 \), respectively. All curves are calculated assuming \( \Omega_0 T_{\text{long}} = 2\pi \). (a) Fringes obtained via the standard Ramsey protocol are horizontally shifted by approximately \( \delta = -2\delta'/\Omega_0 T \) for small drive detunings \( \delta' \). Accordingly, an engaged feedback loop will pull the frequency distribution toward lower frequencies. (b) Passive balancing displaces the fringe laterally so that its zero crossing is kept at the origin. However, for \( \delta' \neq 0 \) any finite-width sampling distribution will experience a skewed response that pushes the center frequency away from the \( \delta = 0 \) lock point. (c) Auto-balanced generation of the frequency discriminant ensures antisymmetric frequency feedback for any drive detuning. At large detunings \( \delta' > \Omega_0 \) the fringe loses contrast but maintains all symmetry properties.](image-url)
large clock errors that scale linearly with \( \delta' \); only for \( \delta' \ll \Omega_0 \) the error propagation is suppressed to first order.

Without heating compensation one finds a residual dependence of \( \delta \) on \( \delta' \) as displayed in Figure 3(b). This dependence is rather weak and does not introduce a clock error as long as \( \delta' \) is on average zero; yet it confirms that heating violates the isomorphism of short and long Ramsey sequences. Numerical simulations based on the thermally averaged outcome of the multiplicative of propagation matrices as introduced in equation (1) reproduce the experimental results.

To some extent, auto-balanced Ramsey spectroscopy and Hyper-Ramsey spectroscopy can be interpreted as the incoherent and coherent versions of the same underlying concept of common-mode suppression, however, only the auto-balanced approach can provide universal immunity to arbitrary interrogation defects. A supplementary discussion found in [34] further explains the analogy. In order to illustrate this universality of the auto-balancing approach we measured the \( \text{Yb}^+ \) clock’s response to various intentionally introduced interrogation defects. Figure 4 displays three defect scenarios together with their resulting clock errors. In the first scenario the \( \pi/2 \)-pulses are delivered with 97 percent of the nominal intensity for the last 3 ms of their 15 ms on-time. Considering a total light shift of 660 Hz this intensity defect is equivalent to a temporary \( \omega_{\text{LO}} \delta' \) deviation of more than \( \Omega_0 \) and gives rise to a clock shift of about 1 Hz when using an unbalanced Ramsey sequence. Auto-balancing the acquisition recovers the undisturbed clock frequency to within the statistical uncertainty. Similarly, defect immunity is verified for drive pulses suffering from engineered phase defects with 0.15 \( \pi \) phase excursions during the second half of each atom-light interaction. Finally, a third scenario assumes a phase lag \( \theta = \pi/10 \) after the first \( \pi/2 \)-pulse, i.e., one effectively uses \( \phi^\pm = \pm \pi/2 + \theta \) instead of employing a symmetric phase modulation. In this case the injected servo-controlled phase correction \( \phi^c \) one-to-one compensates the encountered phase lag.

While all these pulse defects are exaggerated for demonstration purposes they represent a wide variety of less pronounced and often unnoticed parasitic interrogation side effects. For instance, many clocks [35] incorporate some form of magnetic field switching for the atomic ground state preparation and certain prohibitively weak clock transitions [36] require magnetic field-induced state admixtures [37] to enable direct optical Ramsey excitation. Due to transient switching effects a presumably dark Ramsey time might get contaminated. Phase excursions (phase chirps) triggered by laser beam shutters or acousto-optic modulators are another prominent source of error. By choosing a proper \( T_{\text{short}} \) that still covers the transient behavior one can preventively address all such issues.

In conclusion, we have introduced a conceptually sim-
ple and powerful spectroscopic technique to remove accuracy constraints imposed by interrogation-induced clock shifts. We have validated this technique using an optical ion clock and applied it to several highly disturbed Ramsey pulse sequences. This technique is directly applicable to other atomic clocks and could be extended in the future to also address quadrupole-mediated frequency errors [38] and related non-scalar shifts.

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In this supplement we present a conceptual comparison of coherent and incoherent pulse defect immunization approaches. Coherent approaches try to implement frequency discrimination and shift suppression within a single phase-coherent interrogation cycle, while incoherent concepts rely on the combined spectroscopic outcome of separate interrogations. We review the prototypical coherent Hyper-Ramsey interrogation and contrast it with the incoherent auto-balanced Ramsey interrogation protocol. Furthermore, we discuss implications of the different immunization schemes for the frequency stability of the atomic clock. Variables are adopted as defined in the main article.

**COHERENT DEFECT-IMMUNIZATION**

A coherent pulse defect immunization scheme attempts to replace the standard \( \pi/2 \) Ramsey drive pulses with suitable composite pulse (CP) sequences so that the differential excited state population \( p(\delta = 0) \) is only minimally affected when pulse parameters (e.g., \( \delta' \)) deviate from their intended values. A wide variety of such CP sequences have been employed in NMR spectroscopy [S1] or for high-fidelity ion addressing [S2]. Beyond their specific response plateaus the pulse sequences have to meet additional symmetry criteria and should not compromise spectroscopic sensitivity in order to serve as proper clock drive pulses. In principle, a CP sequence (e.g., as found in [S4]) can be turned into a defect-immune interrogation scheme by inserting a dark Ramsey time \( T \) somewhere in the sequence. Two aspects are emphasized: First, for \( \delta = 0 \) the addition of the Ramsey interval will not affect the measured outcome of the pulse sequence. Second, the measured excited state population is the only parameter relevant for controlling \( \omega_{\text{LO}} \).

Accordingly, one can characterize the performance of a CP sequence employed for Ramsey superposition state preparation and readout by choosing \( T = 0 \) and inspecting how the final excited state populations \( p^+ \) and \( p^- \) change as a function of certain pulse parameter deviations. We illustrate this procedure by applying it to the Hyper-Ramsey CP protocol as introduced in [S4].

**THE HYPER-RAMSEY PROTOCOL**

In its generic form, the Hyper-Ramsey scheme leaves the first pulse \( (\pi/2 \) preparation pulse) unchanged and triples the duration of the second pulse (readout pulse) so that \( \Omega_0 \tau = 3\pi/2 \). With \( T = 0 \) one finds dependances of \( p^\pm \) on the normalized drive pulse detuning \( \varepsilon = \delta'/\Omega_0 \) as displayed in Figure S1(a). Around \( \varepsilon = 0 \) the probabilities \( p^\pm \) stay close to \( 1/2 \) clearly indicating the enhanced immunity of the CP sequence compared to a standard interrogation scenario with two \( \pi/2 \) pulses. This improvement can be understood intuitively by considering the two sequences with omitted \( \phi^\pm \) modulation. In this case the driven system performs simple Rabi oscillations given by

\[
p(\tau) = \frac{1}{1 + \varepsilon^2} \sin^2 \left[ \frac{\Omega_0 \tau}{2} \sqrt{1 + \varepsilon^2} \right].
\]

(S1)

The excited state population \( p \) is probed after a total interaction time \( \tau_{\text{HR}} = 2\pi/\Omega_0 \) (Hyper-Ramsey tripled readout pulse duration) or \( \tau_R = \pi/\Omega_0 \) (Ramsey \( \pi/2 \) readout pulse).

A deviation \( \delta' \neq 0 \) affects \( p \) by means of an amplitude "tilt" factor \( 1/(1 + \varepsilon^2) \) and an angular "speed" factor \( \sqrt{1 + \varepsilon^2} \). Figure S1(b) illustrates that the resulting \( p \) deviation is of fourth order in \( \varepsilon \) for \( \tau_{\text{HR}} \)-long pulses and of second order for \( \tau_R \)-long pulses. This superior robustness of \( 2\pi \) (and integer multiples of \( 2\pi \)) long pulse sequences along with an unchanged spectroscopic sensitivity is preserved when adding the \( \phi^\pm \) modulation.

![FIG. S1: Calculated two-level system excited state populations plotted versus drive detuning \( \varepsilon = \delta'/\Omega_0 \) in the absence of relaxation. (a) Around \( \varepsilon = 0 \) a standard Ramsey-type sequence with a \( \phi^+ \) factor \( \pi/2 (\phi^- = -\pi/2) \) phase hop between the two \( \pi/2 \)-pulses exhibits a linear dependence of the population \( p_R^+ (p_R^-) \) on \( \varepsilon \). For a Hyper-Ramsey-type sequence the population \( p_{\text{HR}}^+ (p_{\text{HR}}^-) \) is to first order unaffected by \( \varepsilon \)-deviations. (b) Without phase hops the corresponding populations \( p_R \) and \( p_{\text{HR}} \) reflect the spectroscopic outcome of simple \( \pi \) and \( 2\pi \) pulses and scale quadratically and quartically with \( \varepsilon \), respectively.]

To clarify the effects of the tilt- and speed-terms it is insightful to visualize the system’s unitary evolution on the Bloch sphere as shown in Figure S2 for the \( \phi^+ \) case. With the standard conversion [S5] from the complex state vector \( (g(t), e(t)) \) to a real-valued three-dimensional Bloch vector \( (u(t), v(t), w(t)) \) the driven motion of the system corresponds to a rotation of the Bloch vector through an angle \( \Omega_0 \tau \sqrt{1 + \varepsilon^2} \) about the field vector \( \Omega = \Omega_0 e_u + \delta'e_w \).
Therefore a $\delta'$-defect will manifest itself after the initial $\pi/2$-pulse via the tilt-term to first order in an azimuthal displacement of the Bloch vector on the equatorial plane by an angle $-\varepsilon$ (horizontal red line segment). Displacements due to the speed-factor are only of second order in $\varepsilon$.

After the subsequent $\phi^+ = \pi/2$ phase hop (corresponding to a $-\pi/2$ in-plane rotation) the Bloch vector should now ideally be oriented along $\mathbf{e}_w$ but because of the initially acquired azimuthal offset it is further inclined by an angle of $-\varepsilon$ to the median $w$-$u$ plane. From there it will resume its rotation about $\Omega$ as displayed in detail in Figure S2(b). Since the field vector itself is tilted at an angle $\varepsilon$ out of the equatorial plane (vertical red line segment) it is clear that a conventional $\pi/2$ readout pulse will move the Bloch vector to a final position (marked by the first blue dot on the example trajectory) which is $2\varepsilon$ away from the equator where $p^+ = 1/2$. In a sequence with nonzero Ramsey time $T$ such a first-order displacement in $w$-direction (encoding the excited state population) would then erroneously be interpreted to indicate a frequency deviation $\omega_{LO} \neq \omega_{eg}$ eventually leading to a $\delta'$-induced first-order clock error $\delta = -2\varepsilon/T$. For a $3\pi/2$ readout pulse the geometric situation is different: The Bloch vector continues to move another two quarter-turns on its small cone trajectory around $\Omega$ so that it finally returns (ignoring higher-order speed term effects) to the equatorial plane.

![Figure S2: Computed Bloch vector trajectory for a composite pulse sequence with a $\pi/2$ preparation pulse and an up to $3\pi/2$ long readout pulse assuming $\delta = 0$, $\varepsilon = 0.2$, and $\phi^+ = \pi/2$. (a) The system is initialized in the ground state $|g\rangle$ and evolves under a sequence of coherent operations. (b) A magnified projection of the final part of the trajectory onto the $v$-$w$ plane shows how a $3\pi/2$ readout pulse suppresses the mapping of a $\delta'$-induced offset along the $v$-axis to a clock error-inducing offset along the $w$-axis. The $w$ coordinate is related to the excited state population $p$ via $w = 1 - 2p$. All spectroscopic feedback is solely determined by the measured populations, i.e., by the $w$-axis projection of the final Bloch vector.](https://example.com/figure_s2.png)

Altogether, a total atom-light interaction time of $2\pi/\Omega_0$ ensures that a false drive frequency $\omega_{LO\text{drive}} \neq \omega_{eg}'$ does not lead to a first-order change in the measured excited state populations $p^\pm$. The population deviation latent acquired in the first half-time is coherently compensated in the second half-time. Assuming that the false $\omega_{LO\text{drive}}$ is unchanged for all pulse segments this implicit common-mode suppression corresponds to the explicit common-mode suppression realized in an incoherent way with auto-balanced Ramsey spectroscopy. Note that the population sensitivity $p^\pm(\delta)$ to frequency offsets $\delta$ during an inserted dark Ramsey interval is not coupled to the sensitivity of the population $p^\pm(\delta')$ to drive frequency deviations $\delta'$. If the Ramsey interval is added at a proper point during the $2\pi$ long interrogation sequence (when the Bloch vector is crossing the equatorial plane) the frequency error signal $\tilde{p}(\delta)$ will exhibit the same steepness and contrast as with the standard Ramsey protocol for equally long Ramsey times. Reference [S6] discusses sensitivity aspects of various spectroscopy configurations.

**UNIVERSAL IMMUNITY VIA INCOHERENT BALANCING**

Coherent balancing as discussed above suppresses the linear error propagation from $\delta'$ to $\delta$. Since symmetry arguments forbid second-order contributions to $\delta(\delta')$, one expects a cubic scaling of the induced clock error $\delta$ with the drive detuning $\delta'$ as described and observed in S4 [S8]. Auto-balanced Ramsey spectroscopy eliminates any residual dependance of $\delta$ on $\delta'$, but more importantly, because of its nonspecific nature it simultaneously addresses all interrogation-induced clock shifts.

While some recently proposed coherent Hyper-Ramsey derivatives [S9] [S10] can also completely decouple $\delta$ from $\delta'$, they are specifically tailored to minimize light shift induced clock errors, do not preserve fringe symmetry, and do not cover other interrogation defects. Reference [S11] suggests the construction of "synthetic" clock frequencies by exploiting the functional dependance of $\delta$ on the Ramsey time $T$ for a given detuning $\delta'$. Depending on the number of different Ramsey times these synthetic frequencies develop a corresponding higher order immunity.

Note that all spectroscopy scenarios considered here are strictly speaking hybrid concepts that combine spectroscopic information not exclusively in a coherent or incoherent way, e.g., $\tilde{p}$ is always incoherently derived from $p^+$ and $p^-$. The classification is merely based on the intrinsic balancing or shift suppression mechanism.

**CLOCK STABILITY IMPLICATIONS**

Whether the shift suppression is implemented in a coherent or incoherent way has distinct consequences for the stability of the atomic clock. First consider a standard Ramsey clock operation with dark time $T$. Ramsey
detuning $\delta$, and independent drive detuning $\delta'$. Even under ideal conditions, i.e., with constant $\delta' = 0$, zero mean detuning $\langle \delta \rangle = 0$, and negligible overhead time, the feedback-controlled LO frequency $\omega_{LO}$ exhibits residual frequency fluctuations $\Delta \omega_{LO}$ caused by the finite signal-to-noise ratio of each population measurement. Besides technical issues, quantum projection noise [S12] fundamentally limits the attainable signal-to-noise ratio. Given that projection measurement noise is unchanged for different dark times it is convenient to introduce a $T$-independent Ramsey phase noise $\Delta \phi_{LO}$ and express the observed LO frequency fluctuations as $\Delta \omega_{LO} = \Delta \phi_{LO} / T$. In the context of an auto-balanced Ramsey interrogation this $T$-independence indicates that the injected phase correction $\phi^c$ (itself derived from an uncorrelated equally noisy short Ramsey sequence) effectively doubles the frequency noise variance observed in the long Ramsey sequence. In other words, due to phase noise on $\phi^c$ carried over from the short into the long Ramsey sequence one expects the $\omega_{LO}$-deviations in the auto-balanced long Ramsey sequence to be a factor of $\sqrt{2}$ larger than those observed with an equally long injection-free sequence: Combining two not coherently connected interrogation segments is stability-wise inferior to merging the segments in a coherent way so that projection noise is not encountered twice.

On the other hand, to minimize interrogation-induced shifts it is advisable to only employ low-power drive pulses, which implies longer pulse durations. With additional constraints (limited laser coherence time) for the total length of the interrogation sequence one typically has to compromise between pulse duration and Ramsey time. For example, the Yb$^+$ E3 clock described in [S8] relies on a Hyper-Ramsey interrogation scheme and is operated with a $\pi/2$-pulse duration to Ramsey time ratio of one-to-four. By replacing the $3\pi/2$ long composite read-out pulse with a simple $\pi/2$-pulse the available Ramsey time $T$ is increased by 50 percent. The longer $T$-interval reduces the clock instability by a corresponding factor of 3/2, i.e., the $\sqrt{2}$ stability drop due to auto-balancing is outweighed by the gained Ramsey time.

The above example demonstrates that using an auto-balanced version of Ramsey spectroscopy can be advantageous not only in terms of accuracy but also in terms of clock stability. In particular when considering non-ideal scenarios (with significant initialization times, intrinsically unstable local oscillators, and additional technical constraints) the stability performance of the various interrogation schemes will depend on specific details of the investigated system.

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