Autobalanced 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 autobalanced 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 ultranarrow 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 that, unlike more specialized composite pulse proposals [5–9] and other modified Ramsey techniques [10–12], provides universal immunity to all kinds of reproducible interaction pulse aberrations and associated systematic shifts. These include drive-induced ac Stark shifts (light shifts) [13,14], phase chirps and other phase and/or frequency deviations [15], transient Zeeman shifts [16], 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 autobalanced 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 ultranarrow 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 $\Delta\varphi$ accumulated in a superposition state $|g\rangle + |e\rangle e^{-i\varphi(t)}$ over a free evolution time T (often called dark time or Ramsey time). $\Delta\varphi = E_e T/\hbar$ with $\hbar$ being the reduced Planck constant directly reflects the energy difference E_e between the excited state $|e\rangle$ and ground state $|g\rangle$. Ramsey spectroscopy compares this evolving atomic phase $\varphi(t)$ to the phase evolution $\phi_{\mathrm{LO}}(t)$ of a local oscillator (LO), e.g., a microwave source or a laser [25]. 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 $\omega_{eg} = E_e/\hbar$. Nonetheless, the deterministic preparation of the initial superposition state and the subsequent phase read-out, 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 2×2 matrix based description of a coherently driven two-level system [1,6]. A drive field oscillating with $\cos\phi_{\mathrm{LO}}(t)$ connects the two energy eigenstates $|g\rangle$ and $|e\rangle$.

Applying this field for a time $\tau = t_1 - t_0$ and neglecting counterrotating terms converts an initial state $|i; t = t_0\rangle = g_0|g\rangle + e_0|e\rangle$ into a final state $|f; t = t_1\rangle = g_1|g\rangle + e_1|e\rangle$ via $\binom{g_1}{e_1} =$

$$e^{i\delta'\tau/2}\mathcal{V}[-\phi_{\mathrm{LO}}(t_1)]\mathcal{U}[\delta', \tau, \Omega_0]\mathcal{V}[\phi_{\mathrm{LO}}(t_0)]\begin{pmatrix} g_0 \\ e_0 \end{pmatrix}. \quad (1)$$

In this expression the unitary matrices $\mathcal{U}$ and $\mathcal{V}$ are defined as $\mathcal{U}[\delta', \tau, \Omega_0] =$

$$\begin{pmatrix} \cos\frac{\Omega\tau}{2} - i\frac{\delta'}{\Omega}\sin\frac{\Omega\tau}{2} & i\frac{\Omega_0}{\Omega}\sin\frac{\Omega\tau}{2} \\ i\frac{\Omega_0}{\Omega}\sin\frac{\Omega\tau}{2} & \cos\frac{\Omega\tau}{2} + i\frac{\delta'}{\Omega}\sin\frac{\Omega\tau}{2} \end{pmatrix} \quad (2)$$

and $\mathcal{V}[\xi] = \left(\begin{smallmatrix} 1 & 0 \\ 0 & e^{i\xi} \end{smallmatrix}\right)$. The Rabi frequency Ω_0 and the generalized Rabi frequency $\Omega = \sqrt{\Omega_0^2 + \delta'^2}$ characterize the strength of the coupling between the drive field and atomic transition. $\delta' = \omega_{\mathrm{LOdrive}} - \omega'_{eg}$ denotes the detuning of the oscillatory drive frequency $\omega_{\mathrm{LOdrive}}$ from the instantaneous atomic resonance frequency ω'_{eg}. Primed variables identify quantities that, due to interaction-induced shifts, possibly deviate from their unprimed counterparts. For instance, in the case of the $^{171}\mathrm{Yb}^+$ octupole transition, ω_{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 ω'_{eg} with an unknown light shift $\Delta\omega_{eg} = \omega'_{eg} - \omega_{eg}$ easily exceeding Ω_0.

In the following, we consider a standard optical clock operation scenario where an ultrastable 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 composed of three phase-continuously connected segments: First, a drive pulse at frequency $\omega_{\mathrm{LOdrive}} = \omega'_{eg}$ 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 ω_{LO} that matches the undisturbed atomic phase accumulation (Ramsey detuning $\delta = \omega_{\mathrm{LO}} - \omega_{eg} = 0$). During this interval the LO phase $\phi_{\mathrm{LO}}(t)$ is alternately incremented or decremented by $\phi^{\pm} = \pm\pi/2$ [26]. Finally, another $\pi/2$ pulse with $\omega_{\mathrm{LOdrive}} = \omega'_{eg}$ is applied and subsequently the excited state population $p^{\pm}$ is determined. The differential excited state population $\tilde{p}(\delta) = p^{+}(\delta) - p^{-}(\delta)$ can be interpreted as a derivativelike frequency error signal with a zero crossing at the Ramsey fringe center. Regulating ω_{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 δ. As pointed out before, any aberration from the ideal interrogation sequence, e.g., drive pulse defects (for example, $\delta' \neq 0$) 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 $\omega_{\mathrm{LO}} = \omega_{eg}$, but can rather hide an actual clock error $\delta \neq 0$, whose magnitude will then approximately scale inversely with T [27].

To correctly reproduce ω_{eg} it is essential to distinguish deviations of $\tilde{p}$ due to $\omega_{\mathrm{LO}} \neq \omega_{eg}$ from deviations caused by a flawed interrogation process. This discrimination is accomplished by exploiting the above mentioned dependance of $\tilde{p}(\delta)$ on T: as shown in Fig. 1(a), only for

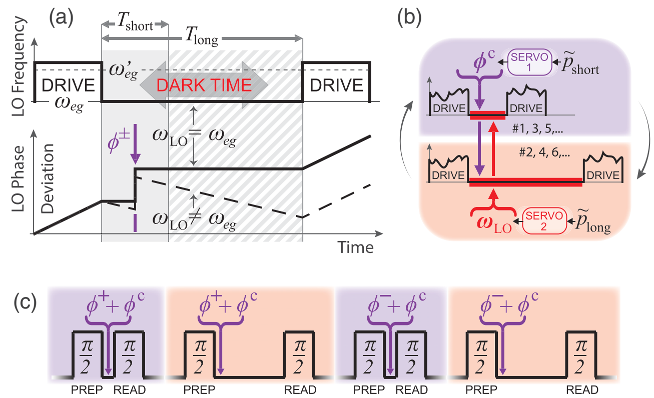

FIG. 1. Ramsey interrogation with shift-inducing drive pulses. The drive pulse frequency $\omega_{\mathrm{LOdrive}}$ in (a) is assumed to be slightly higher than the light-shifted transition frequency ω'_{eg}. This corresponds to an increasing LO phase deviation during the drive period compared to an oscillator that evolves precisely with ω'_{eg} and ω_{eg}. Over the following dark Ramsey time interval of variable duration T, the acquired phase difference between the LO field and atomic oscillator will remain unchanged (aside from the $\phi^{\pm}$ modulation) if the LO phase advances with the unperturbed transition frequency $\omega_{\mathrm{LO}} = \omega_{eg}$ (solid curve). In this case, the phase-to-population conversion performed by the second drive pulse will produce identical outcomes after short and long Ramsey times. Otherwise, for nonzero Ramsey detuning (dashed curve), a T-dependent relative phase is accumulated resulting in differing populations. (b) An auto-balancing control scheme comprises two servo loops acting in parallel on alternately operated short and long Ramsey sequences. Servo 1 equalizes the phase modulated excited state populations p^{+}_{short} and p^{-}_{short}; i.e., it nulls $\tilde{p}_{\mathrm{short}}$ by adjusting ϕ^{c} in both the short and long sequences. Servo 2 nulls $\tilde{p}_{\mathrm{long}}$ via ω_{LO}, also addressing both sequences. The two example plots, displaying LO frequency versus time, represent distorted but isomorphic sequences that only differ in their dark times. (c) One complete probe cycle for a balanced acquisition consists of four Ramsey pulse pairs combining two dark times with two phase hopping directions.

$\omega_{LO} = \omega_{eg}$, i.e., only if the LO phase and atomic phase evolve at an identical pace, can one expect to find the same excited state population when comparing the outcome of two "isomorphic" interrogation sequences that only differ in their dark times T_{short} and T_{long} (see also Refs. [28,29]).

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_{short} and T_{long} Ramsey interrogations, yielding $\tilde{p}_{short}$ and $\tilde{p}_{long}$, respectively, into a defect-immune spectroscopy scheme: By using the differential population $\tilde{p}_{bal} = \tilde{p}_{long} - \tilde{p}_{short}$ as an error signal for ω_{LO}, one obtains a passively balanced frequency feedback with $\tilde{p}_{bal} = 0$ exclusively for $\omega_{LO} = \omega_{eg}$. The direct long-short balancing process is passive in the sense that the individual responses of the two isomorphic sequences are not equalized at an actively enforced working point $\tilde{p}_{short} = \tilde{p}_{long} = 0$, but rather at a defect-dependent nonzero value.

This nonzero equalization point, however, comes with a major drawback, which is, to a lesser extent, also encountered in recently proposed coherent composite pulse schemes [7,8]: around $\delta = 0$ the frequency discriminant $\tilde{p}_{bal}(\delta)$ is no longer an odd function, which leads to skewed sampling distributions and corresponding clock errors, as explained in Fig. 2. To avoid this issue, we implement an active balancing process with two interconnected control loops, as schematically displayed in Fig. 1(b). One feedback loop, controlled by the short Ramsey sequence, ensures $\tilde{p}_{short} = 0$ by injecting together with $\phi^{\pm}$ an additional phase correction ϕ^c. Of course, this requires $\delta' < \Omega_0$ for sufficient fringe contrast [30]. The other feedback loop, controlled by the long Ramsey sequence, steers ω_{LO} so that $\tilde{p}_{long} = 0$. During T_{short} the local oscillator evolves with ω_{LO} as determined via the loop that controls the long sequence. Vice versa, ϕ^c as obtained from the short sequence, is identically applied in the long Ramsey sequence. In essence, a common-mode phase correction, continuously extracted via the short interrogation loop, antisymmetrizes the long interrogation, whose ω_{LO} outcome is then also applied during T_{short}. Because the spectroscopic system tunes itself to the proper symmetry-preserving equalization point, it is auto-balancing and we refer to its LO-steering signal as $\tilde{p}_{auto}$. The antisymmetric lock fringe $\tilde{p}_{auto}(\delta)$ ensures that deviations $\tilde{p}_{auto} \neq 0$ are solely due to $\omega_{LO} \neq \omega_{eg}$.

Formally, the antisymmetrizing effect of the phase correction is seen by writing the autobalanced frequency discriminant $\tilde{p}_{auto}(\delta) = \tilde{p}_{long}[\delta, \phi^c, \phi^{\pm}] - \tilde{p}_{short}[\delta, \phi^c, \phi^{\pm}]$ as an explicit function of the arguments listed in the square brackets. Using the formalism introduced with Eq. (1) and not imposing any constraints on the drive pulses, we show in the Supplemental Material [31] that under active ϕ^c feedback $\tilde{p}_{auto}$ can be expressed in the form $\tilde{p}_{auto} = K_{long}\sin(\delta T_{long}) - K_{short}\sin(\delta T_{short})$ with δ-independent

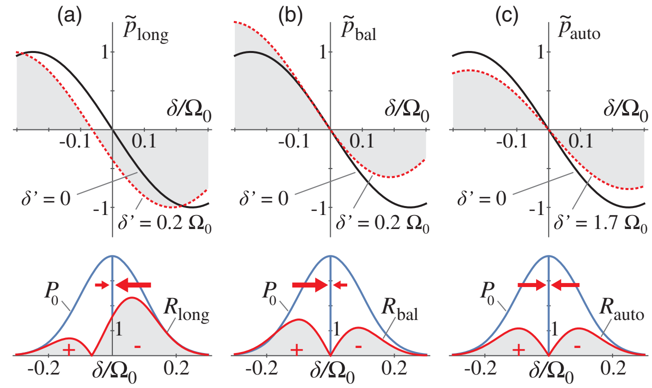

FIG. 2. Simulated Ramsey lock fringes $\tilde{p}(\delta)$ (top row) and rectified feedback responses $R(\delta) = |\tilde{p}P_0|$ for a resonant Gaussian sampling frequency distribution $P_0(\delta)$ (bottom row). $R(\delta)$ is calculated for fringes obtained with nonzero detunings δ' between the drive frequency and the interaction-shifted atomic resonance (dotted red graphs in top row). In agreement with experimental conditions, no coupling is assumed between δ' and the dark time detuning δ. All curves are calculated assuming $\Omega_0\tau = \pi/2$, $\Omega_0 T_{long} = 2\pi$, and $T_{short}/T_{long} = 0.02$. (a) A fringe generated via the standard Ramsey protocol is horizontally shifted by approximately $\delta = -2\delta'/(\Omega_0 T)$ for small drive detunings δ'. Accordingly, an engaged feedback loop will pull the frequency distribution toward lower frequencies. (b) Passive balancing, i.e., subtracting from $\tilde{p}_{long}$ the corresponding response $\tilde{p}_{short}$ of an isomorphic short Ramsey sequence, 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) Autobalanced generation of the frequency discriminant relies on a servo-controlled common-mode correction at the phase level instead of applying direct population corrections. This ensures antisymmetric frequency feedback for any drive detuning. At large detunings $\delta' > \Omega_0$, the fringe loses contrast but maintains all symmetry properties.

amplitudes K_{short} and K_{long}. Therefore, $\tilde{p}_{auto}(\delta)$ is, unlike the passive $\tilde{p}_{bal}(\delta) = \tilde{p}_{long}[\delta, \phi^{\pm}] - \tilde{p}_{short}[\delta, \phi^{\pm}]$, antisymmetric around its zero crossing at $\delta = 0$. For maximum fringe contrast the LO coherence time should exceed the interrogation time scales, which means that δ and δ' become noisewise decoupled. The frequency trajectories of the locked loops will be quantum projection noise [38] dominated and therefore uncorrelated.

We now report on an application of autobalanced 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 $^{171}Yb^+$ have employed extrapolation methods [21,39] or hyper-Ramsey composite pulse approaches [40] to address the drive light induced ac Stark shift. Our experiments were carried out with a single laser-cooled $^{171}Yb^+$ ion confined in an endcap radio-frequency Paul trap [41], with radial and axial trap frequencies of $\omega_r = 2\pi \times 1$ MHz and $\omega_a = 2\pi \times 2$ MHz.

While not actively cooled the trapped ion linearly gains motional energy [19] along each dimension, in our setup at a rate of $300\hbar\omega_r$ 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 ω'_{eg} would change accordingly. We achieve an intensity-neutral Ω_0 equalization by changing the spectral composition of the drive light with an electro-optic modulator that redistributes light from the carrier into inert sidebands spaced in our case $\omega_{\mathrm{EOM}} = 2\pi \times 2$ GHz apart. 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. Except for heating compensation and interrogation specifics, the optical clock is operated following the procedure outlined in Ref. [39]. To facilitate accurate frequency shift measurements with well-controlled detunings δ', the LO laser (prestabilized to a high finesse optical cavity) is also referenced to a second independent Yb^+ $E3$ clock setup. This second clock operates with a hyper-Ramsey protocol and was recently evaluated to be accurate at the millihertz level [42]. Additional details about the experimental implementation, particularly regarding the heating compensation, are contained in the Supplemental Material [31].

Figure 3(a) shows the results of autobalanced clock runs with $\Omega_0 = 2\pi \times 17$ Hz corresponding to a $\pi/2$-pulse duration of 15 ms, $T_{\mathrm{short}} = 6$ ms, and $T_{\mathrm{long}} = 60$ ms. Using about 5 mW of drive laser light focused in a 50 μm diameter spot causes a large light shift of $\omega'_{eg} - \omega_{eg} \approx 2\pi \times$ 660 Hz. Within the statistical uncertainty no clock error is observed for drive detunings δ' 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.

Without heating compensation one finds a residual dependance of δ on δ' as displayed in Fig. 3(b). This dependance is rather weak and does not introduce a clock error as long as δ' is on average zero. Nevertheless, it confirms that heating violates the isomorphism of short and long Ramsey sequences. Numerical simulations based on the thermally averaged outcome of the multiplication of propagation matrices as introduced in Eq. (1) reproduce the experimental results.

To some extent, autobalanced Ramsey spectroscopy and hyper-Ramsey spectroscopy [6] can be interpreted as the incoherent and coherent versions of the same underlying concept of common-mode suppression. However, only the autobalanced approach can provide universal immunity to arbitrary interrogation defects. A supplementary discussion found in Ref. [31] further explains the analogy. In order to illustrate this universality of the auto-balancing approach, we measured the 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% of the nominal intensity for the last 3 ms of their 15 ms on time. Since the total light shift is $\Delta\omega_{eg} \approx 2\pi \times 660$ Hz, this intensity defect is equivalent to a temporary $\omega_{\mathrm{LOdrive}}$ deviation of more than Ω_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 0.15π phase excursions during the second half of each

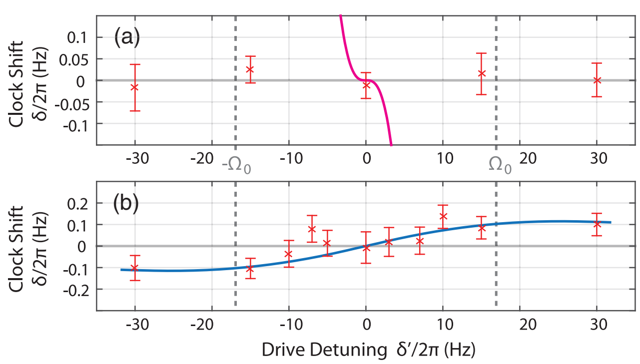

FIG. 3. Clock shifts on the $^{171}\mathrm{Yb}^+$ $E3$ transition frequency as obtained through autobalanced Ramsey spectroscopy when operated with intentionally detuned drive pulses. (a) For isomorphic short and long interrogation sequences, the autobalanced clock is fully immune against drive frequency deviations and the measured data points (1σ error bars shown) line up on the zero clock shift axis. The magenta curve illustrates the δ'-to-δ error propagation expected with the original hyper-Ramsey protocol. (b) Even without heating compensation, the coupling between the clock shift and drive detuning is strongly suppressed. Within statistical uncertainty, the measured clock deviations reproduce the numerically simulated dependance (blue curve).

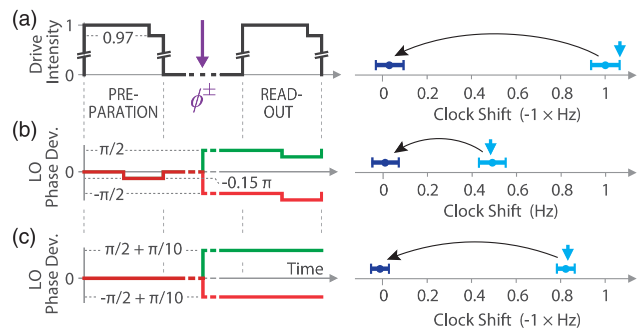

FIG. 4. Defective drive sequences and resulting clock shifts for the case of standard Ramsey spectroscopy (light blue data points with vertical arrows indicating the theoretically expected shift values) and for autobalanced Ramsey spectroscopy (dark blue data points). The applied intensity defect (a), phase excursion (b), and phase lag (c) lead to large clock offsets that are eliminated in the autobalanced Ramsey mode. Both phase deviation plots display ϕ^- (red) and ϕ^+ (green) traces simultaneously.

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 ϕ^c one to one compensates the imposed phase lag.

While all these pulse defects are exaggerated for demonstration purposes, they represent a wide variety of spurious side effects often encountered in Ramsey spectroscopy. For instance, many clocks [43] incorporate some form of magnetic field switching for the atomic ground state preparation. Certain prohibitively weak but metrologically very promising clock transitions [44] also require magnetic field–induced state admixtures to enable direct optical Ramsey excitation [45]. Because of transients associated with the switching of the field, a presumably dark Ramsey time gets contaminated, which has in the past limited the accuracy of such clocks. Phase excursions (phase chirps) triggered by laser beam shutters or acousto-optic modulators are another prominent source of error. By choosing a proper T_{short} that still registers the transient perturbations, one can preventively address all such issues.

Being focused on accuracy improvements, we have not considered the implications of autobalanced Ramsey spectroscopy for the stability of atomic clocks. A detailed stability discussion can be found in the Supplemental Material [31]. In conclusion, we have introduced a conceptually simple 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.

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Note added.—Detailed theoretical results for various autobalanced Ramsey spectroscopy configurations are reported in Ref. [46].

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does not match ω'_{eg} anymore, but as long as $\tilde{p}_{\text{short}} = 0$ is maintained, a common-mode $\delta' \neq 0$ will only cause a loss in fringe contrast and will not lead to any clock errors.

[31] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.120.053602 for additional experimental and conceptual details, which includes Refs. [32–37].

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