Протокол синтетической частоты для рамсеевской спектроскопии в атомных часах.

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Motivation

For some of the promising clock systems, one of the key unsolved problems was the frequency shift of the clock transition due to the excitation pulses themselves:

- Magnetically induced spectroscopy of the transition ${}^{1}S_{0} \rightarrow {}^{3}P_{0}$ (in atoms and ions). Here both AC Stark and quadratic Zeeman shifts take place.
- Two-photon spectroscopy for $S \rightarrow S$ and $S \rightarrow D$ transitions for atoms and ions (AC Stark shift).
- Electric octupole transition (for example, in a single 171 Yb⁺ ion) AC Stark shift.
- Direct frequency-comb spectroscopy

It leads to the significant difficulties to use these transitions in optical clocks (at least for standards with $10^{-17} - 10^{-18}$).

How to help?!

Field shifts in the magnetically induced spectroscopy (MIS).

For MIS we have the following frequency shift:

$$\Delta = \kappa I_p + \beta |\mathbf{B}|^2$$

 I_p is probe field intensity, **B** is static magnetic field.

What to do with this shift?

The first (standard) solution:

The precision experimental measurements of the coefficients k and b, and the high-degree control of the values I_p and **B**.

The main problem for MIS is connected with AC Stark shift and quadratic magnetic shift $\beta |\mathbf{B}|^2 \sim 1-10$ Hz and we can not apply weak magnetic field.

Alternative approach:

Ramsey spectroscopy (field separated in time) of optical clock transitions.

Shift is present only during the excitation pulses. During the free evolution period shifts equal zero. Therefore, the Ramsey like excitations contain, in principle, information about unperturbed clock frequency ω_0 . How to extract and use this information?



The standard Ramsey scheme. Problems.

Maximal contrast of the central resonances (≈ 1) at $\tau \Omega_0 = \pi/2$. The top is shifted as:



Thus, the central resonance is linearly (on Δ) shifted and the use of Ramsey method does not look advantages, because the information about unperturbed clock frequency ω_0 is contained only in asymmetry of the resonance lineshape – it is not suitable for clock operation.



Let us consider more general case with different durations of pulses (τ_1 and τ_2). If at the beginning t=0 atoms are in the lower level $|g\rangle$, then after the action of the two pulses the population n_e of atoms in the excited state $|e\rangle$ can be expressed as Taylor expansion in terms of the dimensionless parameter ($T\delta$)

$$n_{\rm e} = a^{(0)} + a^{(1)}(T\delta) + a^{(2)}(T\delta)^2 + \dots,$$

$$n_{\rm e} = a^{(0)} + a^{(1)}(T\delta) + a^{(2)}(T\delta)^2 + \dots$$

where the coefficients $a^{(j)}$ can be expanded in the powers of Δ'/Ω_0 in the following way:

$$a^{(0)} = \mathcal{A}_0^{(0)} + \mathcal{A}_2^{(0)} (\Delta / \Omega_0)^2 + \mathcal{A}_4^{(0)} (\Delta / \Omega_0)^4 + \cdots,$$

$$a^{(1)} = \mathcal{A}_1^{(1)} (\Delta / \Omega_0) + \mathcal{A}_3^{(1)} (\Delta / \Omega_0)^3 + \cdots,$$

$$a^{(2)} = \mathcal{A}_0^{(2)} + \mathcal{A}_2^{(2)} (\Delta / \Omega_0)^2 + \mathcal{A}_4^{(2)} (\Delta / \Omega_0)^4 + \cdots.$$

The occurrence of terms with even or odd powers only is the direct consequence of the symmetry of n_e that does not change under the simultaneous substitutions $\delta \rightarrow -\delta$ and $\Delta \rightarrow -\Delta$ (it can be shown).

Under $|\Delta/\Omega_0| << 1$ we can use in a parabolic approximation (on $|T\delta| << 1$) to find the leading approximation for the shift of the central

Ramsey fringe

$$\overline{\delta\omega}_0 \approx -\frac{1}{T} \frac{a^{(1)}}{2a^{(2)}}$$



$$\overline{\delta\omega_0} \approx -\frac{1}{T} \frac{a^{(1)}}{2a^{(2)}} \qquad a^{(1)} = \mathcal{A}_1^{(1)} (\Delta/\Omega_0) + \mathcal{A}_3^{(1)} (\Delta/\Omega_0)^3 + \cdots, a^{(2)} = \mathcal{A}_0^{(2)} + \mathcal{A}_2^{(2)} (\Delta/\Omega_0)^2 + \mathcal{A}_4^{(2)} (\Delta/\Omega_0)^4 + \cdots$$

For the usual Ramsey scheme with $\Omega_0 \tau_1 = \Omega_0 \tau_2 = \pi/2$ we find the expected linear dependence

$$\overline{\delta\omega}_0 \approx -\frac{1}{T} \frac{\mathcal{A}_1^{(1)}}{2\mathcal{A}_0^{(2)}} \left(\frac{\Delta}{\Omega_0}\right) = \frac{1}{T} \frac{2\Omega_0 T}{2 + \Omega_0 T} \frac{\Delta}{\Omega_0}$$

For general case with $\tau_1 \neq \tau_2$ we have found that sometimes it is possible to have zero coefficient:

$$\mathcal{A}_1^{(1)} = 0$$
 and $\mathcal{A}_0^{(2)} \neq 0$

In this case the dominating dependence of the shift on $|\Delta'/\Omega_0| <<1$ is now cubic:

$$\overline{\delta\omega}_0 \approx -\frac{1}{T} \frac{\mathcal{A}_3^{(1)}}{2\mathcal{A}_0^{(2)}} \left(\frac{\Delta}{\Omega_0}\right)^3$$

Hyper-Ramsey #1 excitation

Thus, for the condition

$$\Omega_0(\tau_1 + \tau_2) = 2\pi$$

$$\overline{\delta\omega}_0 \approx -rac{1}{T}rac{\mathcal{A}_3^{(1)}}{2\mathcal{A}_0^{(2)}}\left(rac{\Delta}{\Omega_0}
ight)^3$$

the shift of the central Ramsey fringe is much smaller than for usual Ramsey scheme.

It can be shown that the maximal contrast (≈ 1) will be if

$$\tau_2/\tau_1 = 3$$
 (or $\tau_2/\tau_1 = 1/3$) $A_3^{(1)}/2A_0^{(2)} \approx -\pi$



Comparision HR#1 and standard Ramsey scheme (numerical calc.)



We see significant suppression (3-4 orders) of the field shifts in HR#1!

HR#1. Main disadvantge

Main disadvantage is connected with sensitivity to the fluctuations of the Rabi frequency Ω_0 , when we will have

$$\Omega_0(\tau_1 + \tau_2) \neq 2\pi$$

What to do?!



Hyper-Ramsey #2

Ramsey scheme with composite second pulse (including a phase jump of π in the beginning of second pulse):



Here we have cubic dependence at arbitrary Ω_0 and τ :



$$\Omega_0 \tau = \pi/2$$

$$\overline{\delta\omega}_0 \approx -\frac{1}{T} \frac{\mathcal{A}_3^{(1)}}{2\mathcal{A}_0^{(2)}} \left(\frac{\Delta}{\Omega_0}\right)^3$$

Condition for maximal contrast (\approx 1).

Comparison HR#1 vs HR#2 (numerical calc.)



10% variation of the Rabi frequency:

 $\Omega_0 \tau = r \times \pi/2$, r = 0.9; 1.1

The problem: what to do, if shift is very large $|\Delta/\Omega_0|>1$. In this case the cubic dependence is not good. There is a simplest solution:

to use an additional frequency jump Δ_{step} (produced by AOM) during Ramsey pulses only (with fixed Δ_{step}) instead of additional shifting laser.



In this case we will have the effective shift of the

clock transition during Ramsey pulses

whish can be easy controlled by
$$\Delta_{\text{step}}$$
,
i.e. it is easy to have the condition $|\Delta'/\Omega_0| <<1$
and to use the cubic dependence for clock operation.

 $\Lambda' = \Lambda - \Lambda$

$$\overline{\delta\omega}_0 \approx -\frac{1}{T} \frac{\mathcal{A}_3^{(1)}}{2\mathcal{A}_0^{(2)}} \left(\frac{\Delta'}{\Omega_0}\right)^3$$

First experimental realization of HR#2 for ¹⁷¹Yb⁺ in PTB



FIG. 4. Frequency offset of the probe laser stabilized at $\Delta_{\rm L} - \Delta_{\rm S}$ relative to the fully compensated case $\Delta_{\rm S} = \Delta_{\rm L}$, for conditions (a) T = 36 ms, $\tau = 9$ ms and (b) T = 144 ms, $\tau = 36$ ms. The solid red line indicates the predicted dependence if the discriminator signal of the stabilization is generated by alternately stepping the phase of the initial pulse by $\pm \pi/2$. The dashed line in (a) shows the position of the central minimum of the HRS spectrum. The inset in (b) is an enlarged view showing the frequency offset in units of the frequency ν_0 of the Yb⁺ octupole transition.



The experiments demonstrate a suppression of the light shift by four orders of magnitude and an immunity against its fluctuations.

Note, we have this huge advantage practically gratis, because we do not need to use additional devices, we should use only special type of the excitation.

Recent best results for ion clock using hyper-Ramsey method

PRL 116, 063001 (2016)

PHYSICAL REVIEW LETTERS

week ending 12 FEBRUARY 2016

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Single-Ion Atomic Clock with 3×10^{-18} Systematic Uncertainty

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(Received 27 November 2015; published 8 February 2016)

We experimentally investigate an optical frequency standard based on the ${}^{2}S_{1/2}(F=0) \rightarrow {}^{2}F_{7/2}(F=3)$ electric octupole (*E*3) transition of a single trapped 171 Yb⁺ ion. For the spectroscopy of this strongly forbidden transition, we utilize a Ramsey-type excitation scheme that provides immunity to probe-induced frequency shifts. The cancellation of these shifts is controlled by interleaved single-pulse Rabi spectroscopy, which reduces the related relative frequency uncertainty to 1.1×10^{-18} . To determine the frequency shift due to thermal radiation emitted by the ion's environment, we measure the static scalar differential polarizability of the *E*3 transition as $0.888(16) \times 10^{-40}$ J m²/V² and a dynamic correction $\eta(300 \text{ K}) = -0.0015(7)$. This reduces the uncertainty due to thermal radiation to 1.8×10^{-18} . The residual motion of the ion yields the largest contribution (2.1×10^{-18}) to the total systematic relative uncertainty of

the clock of 3.2×10^{-18} .







FIG. 1. (a) Error of the Yb⁺ clock frequency ν_{clock} in the realization of the unperturbed transition frequency ν_0 as a function of an error δ_L in the estimate of the light shift for Rabi and for hyper-Ramsey spectroscopy (HRS). Here, Ramsey pulses of 30.5 ms and a free evolution period of 122 ms are assumed according to the experimental conditions. The very different sensitivities of ν_{clock} to δ_L allow one to engage a servo that uses the difference ϵ between ν_{clock} obtained for Rabi spectroscopy and HRS as the discriminator signal. In (b) the instability (Allan deviation) of experimental ϵ data is shown that follows 230 Hz/ τ (s) (dashed line) for $\tau \ge 1000$ s. The green solid line indicates the expected quantum projection noise limited combined instability of the ν_{Rabi} and ν_{HRS} measurements of 7 Hz/ $\sqrt{\tau(s)}$.

TABLE I. Fractional frequency shifts $\delta\nu/\nu_0(10^{-18})$ and related relative uncertainties $u/\nu_0(10^{-18})$ in the realization of the unperturbed ${}^2S_{1/2}(F=0) \rightarrow {}^2F_{7/2}(F=3)$ transition frequency ν_0 of a single trapped 171 Yb⁺ ion.

Effect	$\delta \nu / \nu_0 ~(10^{-18})$	$u/\nu_0 \ (10^{-18})$
Second-order Doppler shift	-3.7	2.1
Blackbody radiation shift	-70.5	1.8
Probe light related shift	0	1.1
Second-order Zeeman shift	-40.4	0.6
Quadratic dc Stark shift	-1.2	0.6
Background gas collisions	0	0.5
Servo error	0	0.5
Quadrupole shift	0	0.3
Total	-115.8	3.2

Other modified hyper-Ramsey schemes

PHYSICAL REVIEW A 93, 010501(R) (2016)

Modified hyper-Ramsey methods for the elimination of probe shifts in optical clocks

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We develop a method of modified hyper-Ramsey spectroscopy in optical clocks, achieving complete immunity to the frequency shifts induced by the probing fields themselves. Using particular pulse sequences with tailored phases, frequencies, and durations, we can derive an error signal centered exactly at the unperturbed atomic resonance with a steep discriminant which is robust against variations in the probe shift. We experimentally investigate the scheme using the magnetically induced ${}^{1}S_{0} - {}^{3}P_{0}$ transition in ${}^{88}Sr$, demonstrating automatic suppression of a sizable 2×10^{-13} probe Stark shift to below 1×10^{-16} even with very large errors in shift compensation.



MHR – modified

hyper-Ramsev

scheme

FIG. 1. Pulse patterns (a) and excitation spectra (b) of the various spectroscopy methods considered in this Rapid Communication. In the top half of (a), the blue traces indicate the common intensity and frequency patterns of hyper-Ramsey types A and B, while the dotted green traces show modified Ramsey spectroscopy (MRS). The spectra in (b) are calculated with $T = 4\tau$, $\Omega_0 \tau = \pi/2$, $\Delta_{sh}/2\pi = 1.56/\tau$, and $\Delta_{st} = \Delta_{sh}$.



FIG. 3. (a) Scan over the hyper-Ramsey feature in ⁸⁸Sr for $\tau = 10$ ms, T = 50 ms, and $\Omega_0 \tau \approx \pi/2$, and with $\phi = 0$ in the first and last pulse. The theoretical model is overlaid in gray with no fitting parameters used. (b) Modeled and measured residual Stark shifts for the different spectroscopy methods: The HRA_p and HRA_n "standard" hyper-Ramsey lock (blue) shows good suppression compared with modified Ramsey (dashed green), but the HRA_n and HRB_p modified hyper-Ramsey (red) is better. Inset: Enlarged view showing the residual lock offset in fractional frequency units.



PHYSICAL REVIEW A 93, 042506 (2016)

Probe light-shift elimination in generalized hyper-Ramsey quantum clocks

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We present an interrogation scheme for the next generation of quantum clocks to suppress frequency shifts induced by laser probing fields that are themselves based on generalized hyper-Ramsey resonances. Sequences of composite laser pulses with a specific selection of phases, frequency detunings, and durations are combined to generate a very efficient and robust frequency locking signal with an almost perfect elimination of the light shift from off-resonant states and to decouple the unperturbed frequency measurement from the laser's intensity. The frequency lock point generated from synthesized error signals using either $\pi/4$ or $3\pi/4$ laser phase steps during the intermediate pulse is tightly protected against large laser-pulse area variations and errors in potentially applied frequency shift compensations. Quantum clocks based on weakly allowed or completely forbidden optical transitions in atoms, ions, molecules, and nuclei will benefit from these hyperstable laser frequency stabilization schemes to reach relative accuracies below the 10^{-18} level.

DOI: 10.1103/PhysRevA.93.042506

GHR – generalized hyper-Ramsey scheme



FIG. 5. Sensitivity of the clock frequency shift δv for HR and GHR protocols to some pulse area variations of $\Delta \theta / \theta = \pm 10\%$ during laser pulses vs uncompensated residual offsets. All other conditions as in Fig. 2.

However, both methods MHR and GHR did not take into account the decoherence, which can significantly influence on shift suppression.

Next development: Synthetic Frequency Protocol in the Ramsey Spectroscopy of Clock Transitions

We develop an universal method to significantly suppress probe-induced shifts in any types of atomic clocks using the Ramsey spectroscopy. Our approach is based on adaptation of the synthetic frequency concept [V. I. Yudin, *et al.*, Phys. Rev. Lett. **107**, 030801 (2011)] (previously developed for BBR shift suppression) to the Ramsey spectroscopy with the use of interrogations for different dark time intervals. Most significant suppression of the shift is obtained in combination with so-called hyper-Ramsey spectroscopy [V. I. Yudin, *et al.*, Phys. Rev. A **82**, 011804(R) (2010)]. In the latter case, the probe-induced frequency shifts can be suppressed considerably below a fractional level of 10^{-18} practically for any optical atomic clocks, where this shift previously was metrologically significant. The main advantage of our method in comparison with other hyper-Ramsey approaches [R. Hobson, *et al.*, Phys. Rev. A **93**, 010501(R) (2016); T. Zanon-Willette, *et al.*, Phys. Rev. A **93**, 042506 (2016)] consists in much greater efficiency and resistibility in the presence of decoherence.

General idea of synthetic frequency

Previously [V.I.Yudin et al., *Phys. Rev. Lett.* 107, 030801 (2011)] the so-called synthetic frequency method that allowed the suppression of the thermal (BBR) shift in an atomic clock was proposed. However, the idea of this method can be easy extended to the cancellation of arbitrary systematic shifts (e.g., ac Stark shift, Zeeman shift, quadruple shift, and so on). Indeed, let us consider two clock frequencies $\omega^{(0)}_1$ and $\omega^{(0)}_2$ (different in the general case). Assume that due to a certain physical cause we have the stabilized frequencies $\omega^{(0)}_1$ and $\omega^{(0)}_2$, which are shifted relative to the unperturbed frequencies by the values Δ_1 and Δ_2 :

General idea of synthetic frequency

A key advantage of this concept is that to construct the shift-free frequency ω_{syn} , we do not need to know the actual values of shifts Δ_1 and Δ_2 — we only need to know their ratio ε_{12} , which can be exactly calculated or measured for many cases.





The position of the central Ramsey fringe is determined as

$$\omega_T = \omega_0 + \bar{\delta}_T$$

Where the probe-induced shift satisfies the following general dependence on free evolution interval T

$$\bar{\delta}_T = \frac{A_1}{T} + \frac{A_2}{T^2} + \ldots + \frac{A_n}{T^n} + \ldots$$

It allows us to use the certain superposition of two stabilized frequencies ω_{T_1} and ω_{T_2} for two different dark time intervals T_1 and T_2 , which zeros the main shift contribution A_1/T :

$$\omega_{\rm syn}^{(1)} = \frac{\omega_{T_1} - (T_2/T_1)\,\omega_{T_2}}{1 - (T_2/T_1)}$$

This frequency ω_{syn} we will name as "synthetic frequency"



$$\omega_{\rm syn}^{(1)} = \frac{\omega_{T_1} - (T_2/T_1)\,\omega_{T_2}}{1 - (T_2/T_1)}$$

For determinacy, below we will investigate in detail the particular case of $T_1=T$ and $T_2=T/2$:

$$\begin{split} \omega_{\rm syn}^{(1)} &= 2\omega_T - \omega_{T/2} \,,\\ \bar{\delta}_{\rm syn}^{(1)} &= \omega_{\rm syn}^{(1)} - \omega_0 = 2\bar{\delta}_T - \bar{\delta}_{T/2} \end{split}$$

Moreover, we can go further to define other synthetic frequency $\omega_{\rm syn}^{(2)}$, for which both contributions A_1/T and A_2/T^2 will be simultaneously canceled. Here we need to use three different time intervals (T_1, T_2, T_3) with the corresponding stabilized frequencies ($\omega_{T_1}, \omega_{T_2}, \omega_{T_3}$). In particular, we will consider the case of $T_1=T$, $T_2=T/2$ and $T_3 = T/3$, for which the required superposition takes the form:

$$\begin{split} &\omega_{\rm syn}^{(2)} = 3\omega_T - 3\omega_{T/2} + \omega_{T/3} \,, \\ &\bar{\delta}_{\rm syn}^{(2)} = \omega_{\rm syn}^{(2)} - \omega_0 = 3\bar{\delta}_T - 3\bar{\delta}_{T/2} + \bar{\delta}_{T/3} \end{split}$$

Combination of synthetic frequency protocol with original hyper-Ramsey scheme



Thus, we see significant shift suppression due to the use of synthetic frequency protocol



Under $|\Delta/\Omega_0| <<1$ our calculations show the following general character of the dominating nonlinear dependencies on Δ/Ω_0 :



Influence of decoherence

To describe the Ramsey spectroscopy in the presence of decoherence, we will use the formalism of density matrix $\hat{\rho}$, which has the form

$$\hat{\rho}(t) = \sum_{j,k=g,e} |j\rangle \rho_{jk}(t) \langle k|; \ \ \rho_{gg} \equiv n^{(g)}; \ \ \rho_{ee} \equiv n^{(e)}$$

in the basis of states $|g\rangle$ and $|e\rangle$. In our case, the density matrix components $\rho_{jk}(t)$ satisfy the following differential equations:

$$\begin{aligned} &[\partial_t + \Gamma - i\tilde{\delta}(t)]\rho_{eg} = i\Omega(t)[n^{(g)} - n^{(e)}]/2; \quad \rho_{ge} = \rho_{eg}^*; \\ &\partial_t n^{(e)} = i[\Omega(t)\rho_{ge} - \rho_{eg}\Omega^*(t)]/2; \quad n^{(g)} + n^{(e)} = 1. \end{aligned}$$
(19)

The main difference of these equations from the Schrodinger equation consists in the presence of the relaxation constant Γ >0 (for off-diagonal matrix elements ρ_{eg} and ρ_{ge}), which describes the decoherence. In particular, such simple model allows us to estimate the influence of nonzero spectral width of the probe field. To achieve this goal, we can assume the order-of-magnitude agreement between the value Γ and spectral width of the probe field. Similar estimations are very important, because even best modern lasers, used in atomic clocks, have the spectral width at the level of 0.1 Hz. Moreover, there are other possible causes of decoherence, which are connected with an action of environment, collisions, an influence of regimes of traps (or lattices), etc.

Comparison between different hyper-Ramsey schemes in the presence of decoherence ($\Gamma \neq 0$)



Synthetic frequency protocol in combination with original hyper-Ramsey scheme



1. Under decoherence both methods MHR and GHR have nonzero residual shifts already.

2. Synthetic frequency protocol in combination with original hyper-Ramsey scheme gives much robust shift suppression than MHR and GHR.

Main advantages of synthetic frequency protocol

1. Our calculations show that for Γ >0.01- 0.001 Hz the synthetic frequency protocol in combination with original hyper-Ramsey scheme produces much more robust suppression of the probe-induced shifts to the fractional level of 10^{-18} - 10^{-19} in comparison with methods MHR and GHR. Moreover, our approach allows us to achieve this level even for Γ >1 Hz.

2. Apart from the combination with Ramsey and hyper-Ramsey spectroscopy for two-level systems, we have applied the synthetic frequency protocol to the Ramsey spectroscopy of the coherent population trapping (CPT) resonances. Our calculations show significant suppression of the light shift for CPT-Ramsey clocks with the use of synthetic frequency protocol. The same approach can be also applied for so-called pulsed optical pumping (POP) clocks. All these examples demonstrate an universality and efficiency of synthetic frequency protocol, which can be used in any type of clocks based on Ramsey spectroscopy.

Благодарю за внимание

Acknowledgments

Отметь Максима и Занона

Thank you very much for your attention!

Qualitative interpretation

$$H = \begin{bmatrix} -\delta_p/2 & \Omega_0/2 \\ \Omega_0/2 & \delta_p/2 \end{bmatrix} = \delta_p \sigma_z + \Omega_0 \sigma_x \qquad \delta_p = \delta + \Delta$$



Ω

Rabi spectroscopy with π -pulse

$$\langle fin|e \rangle = \sin \frac{\pi - 2\phi}{2} = \cos \phi \sim 1 - \frac{\phi^2}{2}$$
$$n_e \sim 1 - \frac{\delta_p^2}{\Omega_0^2}$$



Standard Ramsey scheme



$$\delta_m = -\frac{\Delta}{\Omega_0} \frac{2}{T} \ll \Delta \ll \Omega_0$$

Hyper-Ramsey #1

87/ 1 +in> (HR#1) (m) 1

 $\phi = \frac{\Delta}{\Omega_0} \ll 1$

 $n_e \sim \left(\frac{\delta T}{2}\right)^2$