

## Generalized Autobalanced Ramsey Spectroscopy of Clock Transitions

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 (Received 25 December 2017; revised manuscript received 22 March 2018; published 23 May 2018)

When performing precision measurements, the quantity being measured is often perturbed by the measurement process itself. Such measurements include precision frequency measurements for atomic clock applications carried out with Ramsey spectroscopy. With the aim of eliminating probe-induced perturbations, a method of generalized autobalanced Ramsey spectroscopy (GABRS) is presented and rigorously substantiated. The usual local-oscillator frequency control loop is augmented with a second control loop derived from secondary Ramsey sequences interspersed with the primary sequences and with a different Ramsey period. This second loop feeds back to a secondary clock variable and ultimately compensates for the perturbation of the clock frequency caused by the measurements in the first loop. We show that such a two-loop scheme can lead to perfect compensation for measurement-induced light shifts and does not suffer from the effects of relaxation, time-dependent pulse fluctuations and phase-jump modulation errors that are typical of other hyper-Ramsey schemes. Several variants of GABRS are explored based on different secondary variables including added relative phase shifts between Ramsey pulses, external frequency-step compensation, and variable second-pulse duration. We demonstrate that a universal antisymmetric error signal, and hence perfect compensation at a finite modulation amplitude, is generated only if an additional frequency step applied during both Ramsey pulses is used as the concomitant variable parameter. This universal technique can be applied to the fields of atomic clocks, high-resolution molecular spectroscopy, magnetically induced and two-photon probing schemes, Ramsey-type mass spectrometry, and the field of precision measurements. Some variants of GABRS can also be applied for rf atomic clocks using coherent-population-trapping-based Ramsey spectroscopy of the two-photon dark resonance.

DOI: [10.1103/PhysRevApplied.9.054034](https://doi.org/10.1103/PhysRevApplied.9.054034)

### I. INTRODUCTION

Atomic clocks are based on high-precision spectroscopy of isolated quantum systems and are currently the most precise scientific instruments. Fractional frequency instabilities and accuracies at the level of  $10^{-18}$  have already been achieved, with the goal of  $10^{-19}$  on the horizon [1]. Frequency measurements at such a level could enable previously inaccessible tests of quantum electrodynamics and cosmological models, searches for drifts of fundamental constants, and alternative types of chronometric geodesy [2].

For some of the promising clock systems, a key limitation is the frequency shift of the clock transition due to the excitation pulses themselves (probe-field-induced shift). In particular, for ultranarrow transitions (e.g., electric octupole [3] and two-photon transitions

[4,5]), the off-resonant ac-Stark shift can be so large in some cases that high-accuracy clock performance is not possible. In the case of magnetically induced spectroscopy [6,7], these shifts (quadratic Zeeman and ac-Stark shifts) could ultimately limit the achievable performance. A similar limitation exists for clocks based on direct frequency-comb spectroscopy [8,9] due to off-resonant ac-Stark shifts induced by large numbers of off-resonant laser modes. In addition to optical standards, probe-field-induced shifts can create significant instability for atomic clocks in the microwave range based on coherent population trapping (CPT) [10–15]. In this context, we can also consider compact cold-atom clocks [16,17] and hot-cell devices like the pulsed optical pumping (POP) clock [18,19].

These challenges can be addressed through the use of Ramsey spectroscopy [20], including its various generalizations and modifications. In contrast to continuous-wave spectroscopy, Ramsey spectroscopy has a large number of

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additional degrees of freedom connected with a wide assortment of parameters that can be precisely controlled: the durations of Ramsey pulses  $\tau_1$  and  $\tau_2$ , the time of free evolution (dark time)  $T$ , the phase composition of Ramsey pulses (e.g., the use of composite pulses [21]), a variety of Ramsey sequences (e.g., the use of three and more Ramsey pulses), different variants to build an error signal, etc.

Some modified Ramsey schemes for the suppression of the probe-field-induced shifts in atomic clocks were theoretically described in Ref. [22], which proposed the use of pulses with different durations ( $\tau_1 \neq \tau_2$ ) and the use of composite pulses in place of the standard Ramsey sequence with two equal  $\pi/2$  pulses. This “hyper-Ramsey” scheme has been successfully realized in an ion clock based on an octupole transition in  $\text{Yb}^+$  (see Refs. [23,24]), where a suppression of the light shift by 4 orders of magnitude and an immunity against its fluctuations were demonstrated. Further developments of the hyper-Ramsey approach have used alternative phase variants to build error signals [25–27]. These developments have allowed for significant improvement in the efficiency of suppression of the probe-field-induced shifts in atomic clocks. However, as was shown in Ref. [28], all previous hyper-Ramsey methods [22–25,27,29] are sensitive to decoherence and spontaneous relaxation, which can appreciably impede the achievement of relative instability and inaccuracy at the level of  $10^{-18}$  (or lower) in modern and future atomic clocks, for which the probe-field-induced shift is not negligible. To eliminate this disadvantage, a more complicated construction of the error signal was recently proposed in Ref. [30] which requires four measurements for each frequency point (instead of two measurements for previous methods), with the use of different generalized hyper-Ramsey sequences presented in Ref. [27]. Nevertheless, the method in Ref. [30] is not free from other disadvantages related to technical issues such as time-dependent pulse area fluctuations and/or phase-jump modulation errors during the measurement of the error signal.

The above approaches [22–25,27,29,30] can be referred to as one-loop methods because they use only one feedback loop and one error signal. However, frequency stabilization can also be realized with two feedback loops connected to Ramsey sequences with different dark periods,  $T_1$  and  $T_2$  (see Refs. [28,31,32]). For example, in Ref. [28], a synthetic frequency protocol was proposed which, in combination with the original hyper-Ramsey sequence [22], allows for a substantial reduction in the sensitivity to decoherence and nonidealities of the interrogation procedure. An alternative and effective approach called autobalanced Ramsey spectroscopy was proposed and experimentally demonstrated in Ref. [32], where, in addition to the stabilization of the clock frequency  $\omega$ , a second loop feeding back on a variable phase during the second pulse was employed. Both of these two-loop methods [28,32] strongly suppress probe-induced shifts of the measurement of the clock frequency.

In this paper, we present and rigorously substantiate a method of generalized autobalanced Ramsey spectroscopy (GABRS), of which the intuitive approach realized in Ref. [32] is a particular case. Our method uses a two-loop approach to feed back and to stabilize the clock frequency  $\omega$  as well as a second (concomitant) parameter  $\xi$ , which is an adjustable property of the first and/or second Ramsey pulses  $\tau_1$  and  $\tau_2$ . To determine the error signals, it is necessary to use Ramsey sequences with two different dark times,  $T_1$  and  $T_2$ . The operation of GABRS consists of the correlated stabilization of both variable parameters,  $\omega$  and  $\xi$ . In addition to the suppression of probe-field-induced shifts, the GABRS technique is protected against various processes of decoherence and also technical issues including time-dependent pulse area fluctuations (even more powerful than the common weak pulse area variation from previous schemes) and phase-jump modulation errors needed to generate the error signal. This insensitivity to decoherence and technical noise is in contrast to previous hyper-Ramsey schemes [22,25,27], which can suffer from relaxation, time-dependent pulse fluctuations, and phase-jump modulation errors. We consider several variants of GABRS with the use of different concomitant parameters  $\xi$ . It is found that the most optimal and universal variant is based on the frequency-step technique, where the concomitant parameter  $\xi$  is equal to the varied additional frequency step  $\Delta_{\text{step}}$  during both Ramsey pulses,  $\tau_1$  and  $\tau_2$ . In this case, universal antisymmetrical error signals are realized.

## II. GENERAL THEORY OF GABRS

In this section, we demonstrate the universality and unprecedented robustness of GABRS. We consider a two-level atom with unperturbed frequency  $\omega_0$  of the clock transition  $|g\rangle \leftrightarrow |e\rangle$  (see Fig. 1), which interacts with a Ramsey sequence of two absolutely arbitrary pulses (with durations  $\tau_1$  and  $\tau_2$ ) of the resonant probe field with frequency  $\omega$ :

$$E(t) = \text{Re}\{\mathcal{E}(t)e^{-i\varphi(t)}e^{-i\omega t}\}, \quad (1)$$

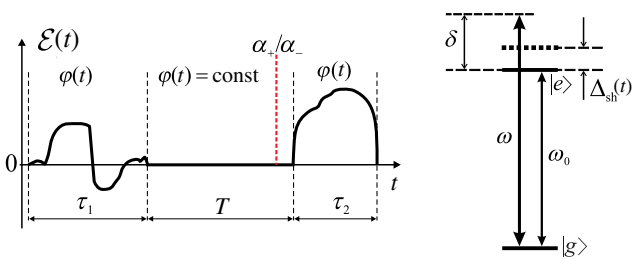


FIG. 1. (Left panel) Schematic illustration of a sequence of two arbitrary Ramsey pulses [with the real amplitude  $\mathcal{E}(t)$  and durations  $\tau_{1,2}$ ] which are separated by the dark time  $T$ . (Right panel) Scheme of the clock transition  $|g\rangle \leftrightarrow |e\rangle$  (with unperturbed frequency  $\omega_0$ ) interacting with the probe field at the frequency  $\omega$ .

which are separated by a free evolution interval (dark time)  $T$ , during which the atom-field interaction is absent (see Fig. 1). We emphasize that the Ramsey pulses with arbitrary durations  $\tau_1$  and  $\tau_2$  can have an arbitrary shape and amplitude [i.e., during  $\tau_1$  and  $\tau_2$ , an amplitude  $\mathcal{E}(t)$  can be an arbitrary real function], and an arbitrary phase function  $\varphi(t)$  (e.g., the Ramsey pulses can be composite pulses). We assume only one restriction: aside from a phase modulation applied to generate the error signal (discussed below), the phase function  $\varphi(t)$  should be constant during the dark time  $T$  (as is usually the case for Ramsey spectroscopy of clock transitions).

Our main goal consists of the development of a universal method, which allows us to stabilize the probe field frequency  $\omega$  at the unperturbed frequency of the clock transition,  $\omega = \omega_0$ , in the presence of decoherence and arbitrary relaxation (including spontaneous relaxation). For this purpose, we use the formalism of density matrix  $\hat{\rho}$ , which has the following form,

$$\hat{\rho}(t) = \sum_{j,k=g,e} |j\rangle \rho_{jk}(t) \langle k|, \quad (2)$$

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

$$\begin{aligned} [\partial_t + \Gamma - i\tilde{\delta}(t)]\rho_{eg} &= i\Omega(t)[\rho_{gg} - \rho_{ee}]/2, \quad \rho_{ge} = \rho_{eg}^*, \\ [\partial_t + \gamma_e]\rho_{ee} - \gamma_{g \rightarrow e}\rho_{gg} &= i[\Omega(t)\rho_{ge} - \rho_{eg}\Omega^*(t)]/2, \\ [\partial_t + \gamma_g]\rho_{gg} - \gamma_{e \rightarrow g}\rho_{ee} &= -i[\Omega(t)\rho_{ge} - \rho_{eg}\Omega^*(t)]/2. \end{aligned} \quad (3)$$

Here, the time dependencies  $\Omega(t)$  and  $\tilde{\delta}(t)$  are determined by the following:  $\Omega(t) = \langle d \rangle \mathcal{E}(t) e^{-i\varphi(t)}$  and  $\tilde{\delta}(t) = \delta - \Delta_{\text{sh}}(t)$  during the action of Ramsey pulses  $\tau_1$  and  $\tau_2$ , but  $\Omega(t) = 0$  and  $\tilde{\delta}(t) = \delta$  during the dark time  $T$ ,  $\langle d \rangle$  is a matrix element of the atomic dipole moment,  $\delta = \omega - \omega_0$  is the detuning of the probe field from the unperturbed atomic frequency  $\omega_0$ , and  $\Delta_{\text{sh}}(t)$  is an actual probe-field-induced shift (see Fig. 1) of the clock transition during the Ramsey pulses (e.g., it can be an ac-Stark shift). Also, Eq. (3) contains five relaxation constants,  $\{\gamma_e, \gamma_{e \rightarrow g}, \gamma_g, \gamma_{g \rightarrow e}, \Gamma\}$   $\gamma_e$  is the decay rate (e.g., a spontaneous one) of the excited state  $|e\rangle$ ;  $\gamma_{e \rightarrow g}$  is the rate of the transmission (e.g., a spontaneous one) to the ground state  $|g\rangle$ ;  $\gamma_g$  is a decay rate of the ground state  $|g\rangle$  (e.g., due to blackbody radiation and/or collisions);  $\gamma_{g \rightarrow e}$  is the rate of the transmission from the ground state  $|g\rangle$  to the excited state  $|e\rangle$ . Note that  $\gamma_{e \rightarrow g} = \gamma_e$  and  $\gamma_{g \rightarrow e} = \gamma_g$  in the case of a closed two-level system, while  $\gamma_{e \rightarrow g} < \gamma_e$  and/or  $\gamma_{g \rightarrow e} < \gamma_g$  in the case of an open system. The constant  $\Gamma = (\gamma_e + \gamma_g)/2 + \tilde{\Gamma}$  describes the total rate of decoherence: spontaneous as well as all other processes, which are included in the parameter  $\tilde{\Gamma}$  (e.g., an influence of the nonzero spectral width and the phase noise of the probe field).

Equation (3) can be rewritten in the vector form:

$$\partial_t \vec{\rho}(t) = \hat{L}(t) \vec{\rho}(t), \quad (4)$$

where  $\hat{L}(t)$  is a  $4 \times 4$  matrix, which is determined by the coefficients of Eq. (3), and  $\vec{\rho}(t)$  is a vector formed by the matrix components  $\rho_{jk}(t)$ :

$$\vec{\rho}(t) = \begin{pmatrix} \rho_{ee}(t) \\ \rho_{eg}(t) \\ \rho_{ge}(t) \\ \rho_{gg}(t) \end{pmatrix}. \quad (5)$$

In this case, a spectroscopic Ramsey signal can be presented in the following general form, which describes Ramsey fringes (as a function of  $\delta$ ):

$$A_{\text{Rams}}(\delta) = (\vec{\rho}_{\text{obs}}, \hat{W}_{\tau_2} \hat{G}_T \hat{W}_{\tau_1} \vec{\rho}_{\text{in}}), \quad (6)$$

where the scalar product is determined in the ordinary way:  $(\vec{x}, \vec{y}) = \sum_m x_m^* y_m$ . Operators  $\hat{W}_{\tau_1}$  and  $\hat{W}_{\tau_2}$  describe an evolution of an atom during the first ( $\tau_1$ ) and second ( $\tau_2$ ) Ramsey pulses, respectively, and the operator  $\hat{G}_T$  describes free evolution during the dark time  $T$ . Vectors  $\vec{\rho}_{\text{in}}$  and  $\vec{\rho}_{\text{obs}}$  are the initial and observed states, respectively. For example, if an atom before the Ramsey sequence is in the ground state  $|g\rangle$  and, after the Ramsey sequence, we detect the atom in the excited state  $|e\rangle$ , then vectors  $\vec{\rho}_{\text{in}}$  and  $\vec{\rho}_{\text{obs}}$  are determined, in accordance with definition (5), as the following:

$$\vec{\rho}_{\text{in}} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 1 \end{pmatrix}, \quad \vec{\rho}_{\text{obs}} = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix}. \quad (7)$$

However, for stabilization of the frequency  $\omega$ , we need to form an error signal (differential signal). In our approach, we use phase jumps  $\alpha_+$  and  $\alpha_-$  of the probe field before the second pulse  $\tau_2$  (see Fig. 1), as was proposed in Ref. [33]. These jumps are described by the operators  $\hat{\Phi}_+$  and  $\hat{\Phi}_-$ , respectively. As a result, the error signal can be presented as a difference:

$$\begin{aligned} S_T^{(\text{err})} &= (\vec{\rho}_{\text{obs}}, \hat{W}_{\tau_2} \hat{\Phi}_+ \hat{G}_T \hat{W}_{\tau_1} \vec{\rho}_{\text{in}}) - (\vec{\rho}_{\text{obs}}, \hat{W}_{\tau_2} \hat{\Phi}_- \hat{G}_T \hat{W}_{\tau_1} \vec{\rho}_{\text{in}}) \\ &= (\vec{\rho}_{\text{obs}}, \hat{W}_{\tau_2} \hat{D}_{\Phi} \hat{G}_T \hat{W}_{\tau_1} \vec{\rho}_{\text{in}}), \end{aligned} \quad (8)$$

with  $\hat{D}_{\Phi} = \hat{\Phi}_+ - \hat{\Phi}_-$ . To maximize the error signal,  $\alpha_{\pm} = \pm\pi/2$  is typically used. However, in real experiments, we can have  $|\alpha_+| \neq |\alpha_-|$  due to various technical reasons (e.g., electronics), which leads to a shift of the stabilized frequency  $\omega$  in the case of standard Ramsey spectroscopy. Therefore, here we consider the general case of arbitrary  $\alpha_+$  and  $\alpha_-$  values to demonstrate the robustness of generalized autobalanced Ramsey spectroscopy, where the condition  $|\alpha_+| \neq |\alpha_-|$  does not lead to a frequency shift in atomic clocks.

Let us consider now the structure of the following operators:  $\hat{G}_T$ ,  $\hat{\Phi}_+$ ,  $\hat{\Phi}_-$ , and  $\hat{D}_\Phi$ . The operator of the free evolution  $\hat{G}_T$  has the following general matrix form:

$$\hat{G}_T = \begin{pmatrix} G_{11}(T) & 0 & 0 & G_{14}(T) \\ 0 & e^{-(\Gamma-i\delta)T} & 0 & 0 \\ 0 & 0 & e^{-(\Gamma+i\delta)T} & 0 \\ G_{41}(T) & 0 & 0 & G_{44}(T) \end{pmatrix}, \quad (9)$$

which corresponds to Eq. (3) if  $\Omega(t) = 0$  and  $\tilde{\delta}(t) = \delta$ . The matrix elements  $G_{11}(T)$ ,  $G_{14}(T)$ ,  $G_{41}(T)$ , and  $G_{44}(T)$  depend on four relaxation constants:  $\{\gamma_e, \gamma_{e \rightarrow g}, \gamma_g, \gamma_{e \rightarrow g}\}$ . In particular, for purely spontaneous relaxation of the excited state  $|e\rangle$ , when  $\gamma_g = \gamma_{g \rightarrow e} = 0$ , we obtain

$$\hat{G}_T = \begin{pmatrix} e^{-\gamma_e T} & 0 & 0 & 0 \\ 0 & e^{-(\Gamma-i\delta)T} & 0 & 0 \\ 0 & 0 & e^{-(\Gamma+i\delta)T} & 0 \\ \frac{\gamma_{e \rightarrow g}}{\gamma_e} (1 - e^{-\gamma_e T}) & 0 & 0 & 1 \end{pmatrix}. \quad (10)$$

Operators for the phase jumps  $\hat{\Phi}_+$  and  $\hat{\Phi}_-$  have the forms

$$\hat{\Phi}_\pm = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & e^{i\alpha_\pm} & 0 & 0 \\ 0 & 0 & e^{-i\alpha_\pm} & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \quad (11)$$

which lead to the following expression for  $\hat{D}_\Phi$ :

$$\hat{D}_\Phi = \hat{\Phi}_+ - \hat{\Phi}_- = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & (e^{i\alpha_+} - e^{i\alpha_-}) & 0 & 0 \\ 0 & 0 & (e^{-i\alpha_+} - e^{-i\alpha_-}) & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}. \quad (12)$$

As a result, taking into account Eq. (9), we obtain a formula for the matrix product  $(\hat{D}_\Phi \hat{G}_T)$ :

$$\hat{D}_\Phi \hat{G}_T = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & e^{-(\Gamma-i\delta)T} (e^{i\alpha_+} - e^{i\alpha_-}) & 0 & 0 \\ 0 & 0 & e^{-(\Gamma+i\delta)T} (e^{-i\alpha_+} - e^{-i\alpha_-}) & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} = e^{-\Gamma T} \hat{\Upsilon}_{\delta T}, \quad (13)$$

where the matrix  $\hat{\Upsilon}_{\delta T}$  is defined as

$$\hat{\Upsilon}_{\delta T} = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & e^{i\delta T} (e^{i\alpha_+} - e^{i\alpha_-}) & 0 & 0 \\ 0 & 0 & e^{-i\delta T} (e^{-i\alpha_+} - e^{-i\alpha_-}) & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}. \quad (14)$$

Note that

$$\hat{\Upsilon}_{\delta T=0} = \hat{D}_\Phi. \quad (15)$$

Thus, the error signal (8) can be rewritten in the following form:

$$S_T^{(\text{err})} = e^{-\Gamma T} (\vec{\rho}_{\text{obs}}, \hat{W}_{\tau_2} \hat{\Upsilon}_{\delta T} \hat{W}_{\tau_1} \vec{\rho}_{\text{in}}). \quad (16)$$

Note that this result is the same if we apply phase jumps  $\alpha_\pm$  at any arbitrary point during the dark interval  $T$ . It is interesting to note that the expression of the error signal in the presence of relaxation is formally different from the

error signal in the absence of relaxation only due to the scalar multiplier  $e^{-\Gamma T}$ , which affects the amplitude, first of all, but not the overall shape of the error signal. This is one of the main specific properties of the phase-jump technique for Ramsey spectroscopy that makes it robust against relaxation. Indeed, for other well-known methods of frequency stabilization, which use a *frequency-jump* technique between alternating total periods of Ramsey interrogation ( $\tau_1 + T + \tau_2$ ), relationship (8) does not exist. In addition, in the ideal case of  $\alpha_+ = -\alpha_- = \alpha$ , the error signal (16) can be expressed as

$$S_T^{(\text{err})} = 2 \sin(\alpha) e^{-\Gamma T} (\vec{\rho}_{\text{obs}}, \hat{W}_{\tau_2} \hat{\Theta}_{\delta T} \hat{W}_{\tau_1} \vec{\rho}_{\text{in}}), \quad (17)$$

where the matrix  $\hat{\Theta}_{\delta T}$ ,

$$\hat{\Theta}_{\delta T} = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & i e^{i\delta T} & 0 & 0 \\ 0 & 0 & -i e^{-i\delta T} & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}, \quad (18)$$

depends only on  $\delta T$ .



The main idea of GABRS is the following. First, apart from  $\delta$  (i.e., frequency  $\omega$ ) for the frequency stabilization procedure, we use an additional (concomitant) variable parameter  $\xi$ , which is related to the first and/or second Ramsey pulses  $\tau_1$  and  $\tau_2$ . For example, the parameter  $\xi$  can be equal to the phase  $\phi^c$  of the second pulse, as was proposed in Ref. [32]. However, as shown below, there are many other variants of the concomitant parameter  $\xi$ . Thus, the error signal in Eq. (8) should be considered as a function of two variable parameters  $S_T^{(\text{err})}(\delta, \xi)$ . Second, we use the Ramsey interrogation of the clock transition for two different, fixed intervals of free evolution,  $T_1$  and  $T_2$ , i.e., we use two error signals,  $S_{T_1}^{(\text{err})}(\delta, \xi)$  and  $S_{T_2}^{(\text{err})}(\delta, \xi)$ .

For GABRS, the procedure for the frequency stabilization is organized as a series of the following cycles. For interrogation with dark time  $T_1$ , the parameter  $\xi$  is fixed, and we stabilize the variable detuning  $\delta$  (i.e., frequency  $\omega$ ) at the zero point of the error signal:  $S_{T_1}^{(\text{err})}(\delta, \xi_{\text{fixed}}) = 0$ . After this procedure, we switch to interrogation with dark time  $T_2$ , where we fix the previously obtained detuning  $\delta$  and stabilize the variable parameter  $\xi$  at the zero point of the second error signal:  $S_{T_2}^{(\text{err})}(\delta_{\text{fixed}}, \xi) = 0$ . If we continue these cycles, then the final result (formally for  $t \rightarrow \infty$ ) consists of the stabilization of both parameters,  $\delta = \bar{\delta}_{\text{clock}}$  and  $\xi = \bar{\xi}$ , which corresponds to the solution of a system of two equations:

$$S_{T_1}^{(\text{err})}(\delta, \xi) = 0, \quad S_{T_2}^{(\text{err})}(\delta, \xi) = 0, \quad (19)$$

in relation to the two unknowns  $\delta$  and  $\xi$ . The value  $\bar{\delta}_{\text{clock}}$  describes the frequency shift in an atomic clock.

Taking into account relationship (16), the system Eq. (19) can be written in the following form:

$$(\vec{\rho}_{\text{obs}}, \hat{W}_{\tau_2} \hat{\Upsilon}_{\delta T_1} \hat{W}_{\tau_1} \vec{\rho}_{\text{in}}) = 0, \quad (\vec{\rho}_{\text{obs}}, \hat{W}_{\tau_2} \hat{\Upsilon}_{\delta T_2} \hat{W}_{\tau_1} \vec{\rho}_{\text{in}}) = 0. \quad (20)$$

Let us show that Eq. (20) always contains the solution  $\delta = 0$ . Indeed, if we apply  $\delta = 0$  for operators  $\hat{\Upsilon}_{\delta T_1}$  and  $\hat{\Upsilon}_{\delta T_2}$ , then, due to Eq. (15), we obtain that the system of two equations (20) is reduced to the following single equation:

$$(\vec{\rho}_{\text{obs}}, \hat{W}_{\tau_2} \hat{D}_{\Phi} \hat{W}_{\tau_1} \vec{\rho}_{\text{in}})|_{\delta=0} = 0, \quad (21)$$

in relation to only one unknown  $\xi$ , which always has a solution under an appropriate choice of the parameter  $\xi$ .

Thus, we analytically show here that the GABRS method always leads to zero field-induced shift of the stabilized frequency  $\omega$  in an atomic clock,  $\bar{\delta}_{\text{clock}} = 0$ . This fundamental result does not depend on relaxation constants  $\{\gamma_e, \gamma_{e \rightarrow g}, \gamma_g, \gamma_{g \rightarrow e}, \Gamma\}$ , the values of phase jumps  $\alpha_+$  and  $\alpha_-$  used for error signals, or the parameters [such as amplitude, shape, duration, phase structure  $\varphi(t)$ , shift  $\Delta_{\text{sh}}(t)$ , etc.] of the two Ramsey pulses,  $\tau_1$  and  $\tau_2$ . Such

a robustness is unprecedented for Ramsey spectroscopy. Indeed, all known methods of hyper-Ramsey spectroscopy [22,23,25,27,29,30], which can significantly suppress field-induced shifts, are sensitive (except for Ref. [30]) to relaxation processes and decoherence (see Ref. [28]), and all of these methods require the use of rectangularly shaped Ramsey pulses. Moreover, all previously used Ramsey methods (including the usual Ramsey spectroscopy with two equal  $\pi/2$  pulses) require the condition  $\alpha_- = -\alpha_+$  for phase jumps because any nonideality ( $\alpha_- \neq -\alpha_+$ ) leads to an additional shift which is approximately equal to the value of  $-(\alpha_+ + \alpha_-)/(2T)$ . Summarizing, practically all nonidealities of the interrogation procedure (including field-induced shifts of atomic levels, acousto-optic-modulator-induced phase variations, and so on) and all relaxation processes (including decoherence) influence only the stabilized concomitant parameter  $\bar{\xi}$ , while the stabilized frequency  $\omega$  remains unshifted, with  $\bar{\delta}_{\text{clock}} = 0$ . Note that this result is rigorously valid in the absence of atomic ‘‘phase memory’’ because, in our theory, we assume that the initial state  $\vec{\rho}_{\text{in}}$  [see Eq. (7)] does not depend on field parameters and is the same for all interrogation cycles (with  $T_1$  and  $T_2$ ).

It is interesting to note that the solution of Eqs. (20) and (21) does not formally depend on the values  $T_1$  and  $T_2$  at all. However, from an experimental viewpoint, it is better to use the condition  $T_2 \ll T_1$ . Indeed, because, during the interrogation procedure with dark time  $T_1$ , we stabilize the frequency  $\omega$  using the error signal  $S_{T_1}^{(\text{err})}(\delta, \xi_{\text{fixed}}) = 0$ , we always have  $|\delta| < 1/T_1$ , even during the first cycles of the clock stabilization. On the other hand, nonzero detuning  $\delta \neq 0$  influences the second interrogation procedure with dark time  $T_2$  (to stabilize the concomitant parameter  $\xi$ ), in conformity with the value  $\delta T_2$ , which is contained in the error signal  $S_{T_2}^{(\text{err})}(\delta, \xi)$ . Therefore, if  $T_2 \ll T_1$ , then we obtain an estimation,  $|\delta T_2| < (T_2/T_1) \ll 1$ ; i.e., the results of the stabilization of the concomitant parameter  $\xi$  [using  $S_{T_2}^{(\text{err})}(\delta_{\text{fixed}}, \xi) = 0$ ] weakly depends on the results of the frequency stabilization during an interrogation procedure with dark time  $T_1$ . An additional advantage of the condition  $T_2 \ll T_1$  is connected with the short-term stability of an atomic clock. Indeed, because the second feedback loop (stabilization of  $\xi$ ) increases the total period of each cycle, then it is better to use the shortest possible  $T_2$  value. Formally we can even use  $T_2 = 0$  (with the phase jumps  $\alpha_{\pm}$  in the virtual point between pulses  $\tau_1$  and  $\tau_2$ ). However, owing to technical transient regimes (i.e., in acousto-optic modulators) under the switching off and on of the Ramsey pulses in real experiments, we believe that it is necessary to keep some nonzero dark time,  $T_2 \neq 0$ , which significantly exceeds any various transient times. For example, in the case of magnetically induced spectroscopy [6,7], the transient processes, associated with the switching off and on of the magnetic field, can be relatively slow.

Though the solution  $\bar{\delta}_{\text{clock}} = 0$  does not depend on the amplitude and shape of the Ramsey pulses; nevertheless, to maximize the error signals  $S_{T_1}^{(\text{err})}(\delta, \xi_{\text{fixed}})$  and  $S_{T_2}^{(\text{err})}(\delta_{\text{fixed}}, \xi)$ , we need to use quite specific types of the Ramsey pulses. Some appropriate variants are presented below.

### III. OPTIMAL REGIME OF FREQUENCY STABILIZATION IN THE PRESENCE OF RELAXATION

Let us determine an optimal regime of GABRS for frequency stabilization, which uses the error signal  $S_{T_1}^{(\text{err})}(\delta, \xi = \bar{\xi})$ , where  $T_1 \gg T_2$ . It is well known that the instability of the atomic clock is proportional to the following expression:

$$\frac{\Delta_{\text{width}}}{A_{\text{res}}} N_{\text{noise}} \sqrt{\frac{t_c}{t}}, \quad (22)$$

where  $A_{\text{res}}$  and  $\Delta_{\text{width}}$  are the amplitude and width of the atomic resonance, respectively,  $N_{\text{noise}}$  is the level of noise,  $t_c$  is the time period of one measurement cycle, which includes Ramsey interrogation sequences with both long ( $T_1$ ) and short ( $T_2$ ) dark intervals. Thus, to optimize the stability, we need to minimize the combination:

$$\frac{\Delta_{\text{width}}}{A_{\text{res}}} N_{\text{noise}} \sqrt{t_c}. \quad (23)$$

The amplitude  $A_{\text{res}}$  of the error signal  $S_{T_1}^{(\text{err})}(\delta, \xi = \bar{\xi})$  is proportional to the multiplier  $e^{-\Gamma T_1}$  [see Eq. (16)], whereas the width of the central Ramsey fringe  $\Delta_{\text{width}}$  is proportional to  $1/T_1$ . The noise  $N_{\text{noise}}$  can also depend on  $T_1$  (e.g., due to the Dick effect [34]). However, any reasonable model of noise shows the dependence  $N_{\text{noise}}(T_1)$  as a monotonically increasing function, i.e.,  $\partial N_{\text{noise}}(T_1)/\partial T_1 \geq 0$  for an arbitrary  $T_1$  value. Therefore, a minimization of the expression (23) corresponds to the minimization of the following expression:

$$\frac{e^{\Gamma T_1} N_{\text{noise}}(T_1) \sqrt{t_c}}{T_1}, \quad \left( \frac{\partial N_{\text{noise}}(T_1)}{\partial T_1} \geq 0 \right), \quad (24)$$

in relation to the dark interval  $T_1$ .

Let us consider two limiting cases. At first, we assume that  $t_c$  does not depend on  $T_1$ . In this case, the minimization of Eq. (24) occurs at the point  $T_1 = T_1^{(\text{opt})}$ , which satisfies the following equation (in relation to the unknown  $T_1$  value):

$$T_1 = \frac{1}{\Gamma[1 + (\Gamma N_{\text{noise}})^{-1} \partial N_{\text{noise}} / \partial T_1]}. \quad (25)$$

This equation leads to the condition  $T_1^{(\text{opt})} \leq \Gamma^{-1}$ , where the equality  $T_1^{(\text{opt})} = \Gamma^{-1}$  holds for  $N_{\text{noise}}(T_1) = \text{const}$ . In the opposite case, we assume that  $t_c \propto T_1$ , when a minimization of Eq. (24) occurs at the point  $T_1 = T_1^{(\text{opt})}$ , which satisfies another equation:

$$T_1 = \frac{1}{2\Gamma[1 + (\Gamma N_{\text{noise}})^{-1} \partial N_{\text{noise}} / \partial T_1]}. \quad (26)$$

This equation leads to the condition  $T_1^{(\text{opt})} \leq (2\Gamma)^{-1}$ , where the equality  $T_1^{(\text{opt})} = (2\Gamma)^{-1}$  holds for  $N_{\text{noise}}(T_1) = \text{const}$ .

Thus, we rigorously derive an universal condition for optimization of the frequency stabilization:

$$T_1^{(\text{opt})} \leq \Gamma^{-1}, \quad (27)$$

which is valid for an arbitrary scenario. If the Ramsey pulses are relatively short,  $\tau_{1,2} \ll T_1^{(\text{opt})}$ , then the influence of relaxation during Ramsey pulses (i.e., for the evolution operators  $\hat{W}_{\tau_1}$  and  $\hat{W}_{\tau_2}$ ) is not significant. In this case, from Eq. (16), we see that the relaxation leads primarily to a decrease of the amplitude of the error signal [see the multiplier  $e^{-\Gamma T}$  in Eq. (16)]; however, the line shape of the error signals  $S_{T_1}^{(\text{err})}(\delta, \xi_{\text{fixed}})$  and  $S_{T_2}^{(\text{err})}(\delta_{\text{fixed}}, \xi)$  are not significantly deformed compared to the case with no relaxation ( $\gamma_e = \gamma_{e \rightarrow g} = \gamma_g = \gamma_{g \rightarrow e} = \Gamma = 0$ ).

Note that the results of Sec. III are valid for arbitrary Ramsey spectroscopy (i.e., not only for GABRS), where the error signal is formed by the use of the phase-jump technique during the dark time  $T$ .

### IV. DIFFERENT VARIANTS OF GABRS

In this section, we consider some variants of Ramsey sequences with different choices for the concomitant parameter  $\xi$ . Because arbitrary relaxation and practically all nonidealities of the Ramsey interrogation scheme do not lead to a shift of the stabilized clock frequency,  $\bar{\delta}_{\text{clock}} = 0$ , we focus our attention only on the field-induced shift of the clock transition  $\Delta_{\text{sh}}$  during the Ramsey pulses  $\tau_1$  and  $\tau_2$ . We show how the value  $\Delta_{\text{sh}}$  influences the stabilized concomitant parameter  $\bar{\xi}$  and the error signals  $S_{T_1}^{(\text{err})}(\delta, \xi = \bar{\xi})$  and  $S_{T_2}^{(\text{err})}(\delta = 0, \xi)$ , which contain the main information about the dynamic efficiency of GABRS.

For simplicity, all calculations are done for  $\alpha_{\pm} = \pm\pi/2$  and in the absence of relaxation ( $\gamma_e = \gamma_{e \rightarrow g} = \gamma_g = \gamma_{g \rightarrow e} = \Gamma = 0$ ) because, in the case of  $T_1 \leq T_1^{(\text{opt})}$ , the relaxation leads primarily to a decrease of the amplitude of the error signals, whereas the line shapes of the error signals  $S_{T_1}^{(\text{err})}(\delta, \xi_{\text{fixed}})$  and  $S_{T_2}^{(\text{err})}(\delta_{\text{fixed}}, \xi)$  are not significantly deformed compared to the case with no relaxation

(see Sec. III). Also, for calculations, the initial and observed states  $\vec{\rho}_{\text{in}}$  and  $\vec{\rho}_{\text{obs}}$  correspond to Eq. (7).

### A. Autobalanced Ramsey spectroscopy with an additional phase correction

Here, we describe a detailed theoretical basis for the original autobalanced Ramsey spectroscopy method demonstrated in Ref. [32]. In the context of the general theory developed above, this spectroscopy can be considered as a partial case of GABRS, where the concomitant parameter  $\xi$  is equal to the varied additional phase  $\phi^c$  during the second pulse [Fig. 2(a)]. In this case, we always have  $\bar{\delta}_{\text{clock}} = 0$ , and the stabilized phase  $\bar{\phi}^c$  is determined as the solution of Eq. (21). In the presence of the probe-field-induced shift of the clock transition  $\Delta_{\text{sh}}$  during the Ramsey pulses, the phase  $\bar{\phi}^c$  is a function  $\bar{\phi}^c(\Delta_{\text{sh}})$  of the value  $\Delta_{\text{sh}}$ . These dependencies are presented in Fig. 2(b) for different pulse areas  $\Omega_0\tau$ . In the case of  $(\Delta_{\text{sh}}/\Omega_0) < 1$ , we have the following approximate dependence:  $\bar{\phi}^c(\Delta_{\text{sh}}) \approx 2r\Delta_{\text{sh}}/\Omega_0$ , where the coefficient  $r$  determines the pulse area,  $\Omega_0\tau = r\pi/2$ . Thus, this dependence can be written as  $\bar{\phi}^c(\Delta_{\text{sh}}) \approx 4\Delta_{\text{sh}}\tau/\pi$  (if  $\Delta_{\text{sh}}\tau < 1$ ).

The error signals  $S_{T_1}^{(\text{err})}(\delta, \phi = \bar{\phi}^c)$  and  $S_{T_2}^{(\text{err})}(\delta = 0, \phi)$  for different values  $\Delta_{\text{sh}}$  are presented in Fig. 3. As we see, for the condition  $|\Delta_{\text{sh}}/\Omega_0| > 1$ , the error signal  $S_{T_1}^{(\text{err})}(\delta, \phi = \bar{\phi}^c)$  becomes smaller and distinctly nonantisymmetrical, which can lead to clock errors. Thus, the autobalancing technique of varying the phase only during

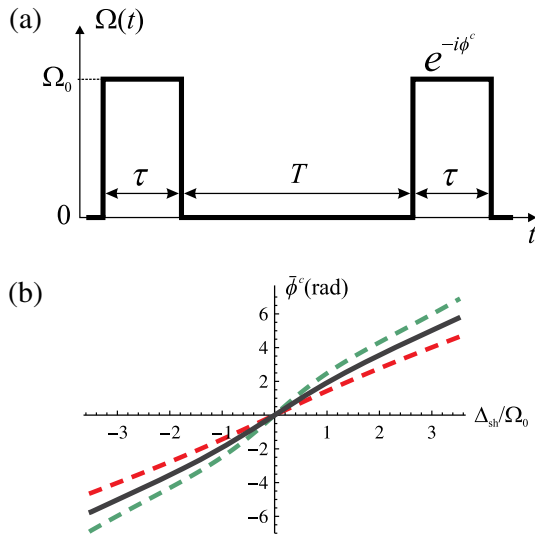


FIG. 2. (a) Schematic illustration of Ramsey pulses (with the same duration  $\tau$ ) for the autobalanced Ramsey spectroscopy technique demonstrated in Ref. [32], where the concomitant parameter  $\xi$  is equal to the additional phase  $\phi^c$  of the second pulse. (b) The dependencies of stabilized phase  $\bar{\phi}^c(\Delta_{\text{sh}})$  for different pulse areas:  $\Omega_0\tau = \pi/2$  (the black solid line),  $\Omega_0\tau = 1.2 \times \pi/2$  (the green dashed line), and  $\Omega_0\tau = 0.8 \times \pi/2$  (the red dashed line).

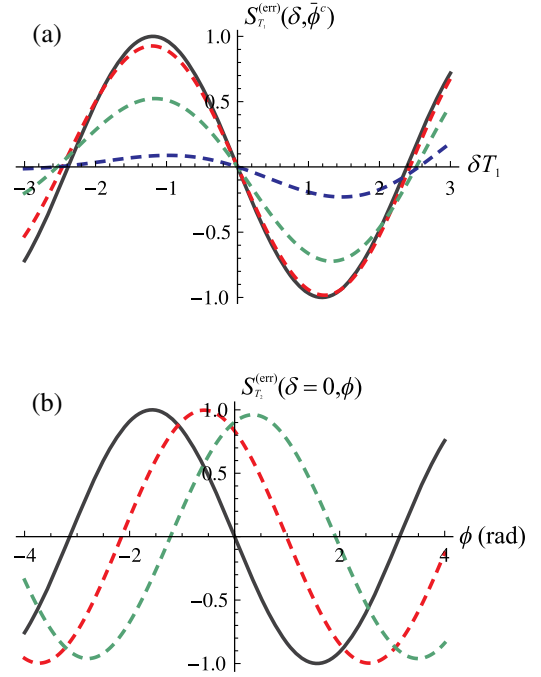


FIG. 3. Error signals under  $\Omega_0 T_1 = 2\pi$  and  $\Omega_0\tau = \pi/2$ , and for different field-induced shifts of the clock transition during Ramsey pulses,  $\Delta_{\text{sh}}$ . (a) Error signal  $S_{T_1}^{(\text{err})}(\delta, \phi = \bar{\phi}^c)$ :  $\Delta_{\text{sh}}/\Omega_0 = 0$  (the black solid line),  $\Delta_{\text{sh}}/\Omega_0 = 1$  (the red dashed line),  $\Delta_{\text{sh}}/\Omega_0 = 2$  (the green dashed line), and  $\Delta_{\text{sh}}/\Omega_0 = 3$  (the blue dashed line). (b) Error signal  $S_{T_2}^{(\text{err})}(\delta = 0, \phi)$ :  $\Delta_{\text{sh}}/\Omega_0 = 0$  (the black solid line),  $\Delta_{\text{sh}}/\Omega_0 = 0.5$  (the red dashed line), and  $\Delta_{\text{sh}}/\Omega_0 = 1.0$  (the green dashed line).

the second Ramsey pulse works well only for  $|\Delta_{\text{sh}}/\Omega_0| < 1$ . Distortions in the error signals arising from this problem can be largely reduced by the use of an additional and well-controllable frequency step  $\Delta_{\text{step}}$  only during the Ramsey pulses  $\tau_1$  and  $\tau_2$  [22,35]. In this case, all dependencies presented in Figs. 2(b) and 3 are the same if we replace  $\Delta_{\text{sh}} \rightarrow \Delta_{\text{eff}} = (\Delta_{\text{sh}} - \Delta_{\text{step}})$ . Thus, we can always apply a frequency step  $\Delta_{\text{step}}$  (e.g., with an acousto-optic modulator) during excitation to achieve the condition  $|\Delta_{\text{eff}}/\Omega_0| \ll 1$  for an effective shift  $\Delta_{\text{eff}}$ , as has been done in experiments [23–25,32].

In addition, this variant of GABRS can also be used in atomic clocks based on CPT, where we can use as the concomitant parameter  $\xi$  the varied phase  $\phi^c$  of the second (detecting) pulse in CPT-Ramsey spectroscopy.

### B. Autobalanced Ramsey spectroscopy with an additional frequency step

As an alternative to the previous method with additional varied phase  $\phi^c$  during the second pulse [32], let us describe another variant of GABRS, where the concomitant parameter  $\xi$  is equal to the varied additional frequency step  $\Delta_{\text{step}}$  during both Ramsey pulses  $\tau_1$  and  $\tau_2$  [Fig. 4(a)]. This frequency-step technique was proposed in Refs. [22,35].

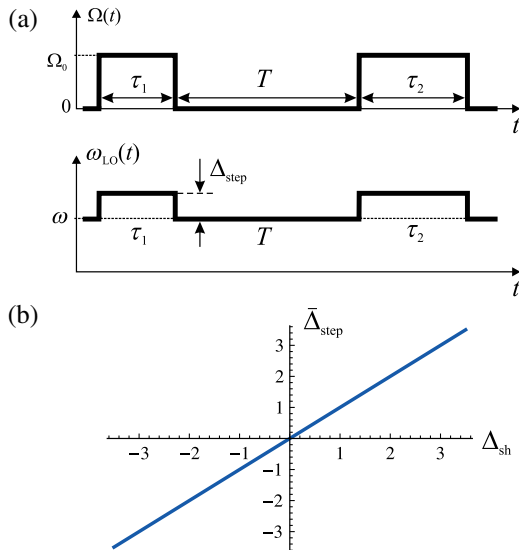


FIG. 4. (a) Schematic illustration of a Ramsey interrogation scheme for GABRS, where the concomitant parameter  $\xi$  is equal to the additional frequency step  $\Delta_{\text{step}}$  during both Ramsey pulses,  $\tau_1$  and  $\tau_2$ . (b) The dependence of the stabilized frequency step  $\bar{\Delta}_{\text{step}}(\Delta_{\text{sh}})$ , which has the universal form  $\bar{\Delta}_{\text{step}} = \Delta_{\text{sh}}$  for arbitrary values of  $\Omega_0$ ,  $\tau_1$ , and  $\tau_2$ .

Excluding the explicit phase jumps  $\alpha_{\pm}$ , the frequency step  $\Delta_{\text{step}}$  can be formally described by a phase function  $\varphi(t)$  in Eq. (1) with the nonzero time derivative  $d\varphi(t)/dt = \Delta_{\text{step}}$  during the pulses ( $\tau_1$  and  $\tau_2$ ) and with the zero time derivative  $d\varphi(t)/dt = 0$  during the dark time  $T$ , and phase continuity is maintained throughout. In this case, we always have  $\bar{\delta}_{\text{clock}} = 0$ , and the stabilized frequency step  $\bar{\Delta}_{\text{step}}$  is determined as the solution of Eq. (21), which has the universal form  $\bar{\Delta}_{\text{step}} = \Delta_{\text{sh}}$  for arbitrary values of  $\Omega_0$ ,  $\tau_1$ , and  $\tau_2$  [Fig. 4(b)]. This universal dependence can be slightly deformed only due to some nonidealities of the interrogation scheme (e.g., if  $\alpha_+ \neq -\alpha_-$ ).

The error signals  $S_{T_1}^{(\text{err})}(\delta, \Delta_{\text{step}} = \bar{\Delta}_{\text{step}})$  and  $S_{T_2}^{(\text{err})}(\delta = 0, \Delta_{\text{step}})$  have universal antisymmetrical forms for different values of  $\Delta_{\text{sh}}$  (Fig. 5). Note that this antisymmetry does not depend on the Rabi frequency  $\Omega_0$ . Thus, we believe that this variant of GABRS is more optimal and robust than the approach used in Ref. [32], where an additional phase  $\phi^c$  was varied during the second pulse. In fact, in the case of  $|\Delta_{\text{sh}}/\Omega_0| \gg 1$ , it is already necessary to use the frequency-step technique of Refs. [22,35] to compensate for the very large actual shift  $\Delta_{\text{sh}}$ . This frequency-step technique was also used in experiments in Ref. [32] with the  $\text{Yb}^+$  ion because, for the octupole clock transition, the condition  $|\Delta_{\text{sh}}/\Omega_0| \gg 1$  is practically always true. But in this case, the use of an additional varied phase  $\phi^c$  in Ref. [32] seems to be an excessive technical complication because we can directly use the frequency-step technique ( $\Delta_{\text{step}}$ ) in GABRS without any additional manipulations.

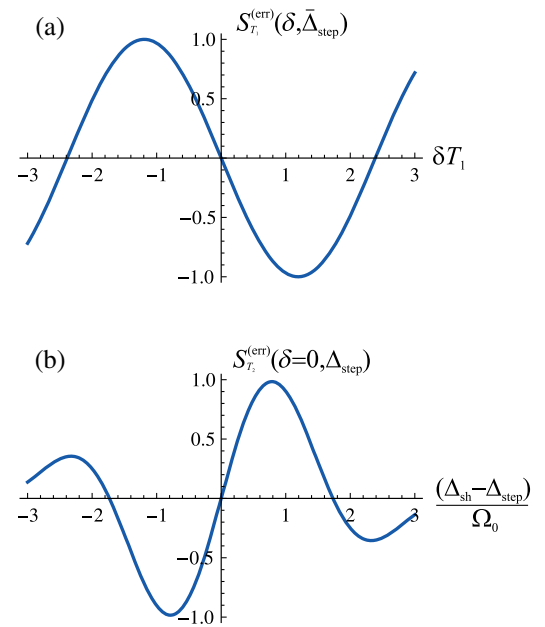


FIG. 5. Error signals under  $\Omega_0 T_1 = 2\pi$ ,  $\Omega_0 \tau = \pi/2$  ( $\tau_1 = \tau_2 = \tau$ ), and for arbitrary field-induced shifts of the clock transition during Ramsey pulses,  $\Delta_{\text{sh}}$ : (a) error signal  $S_{T_1}^{(\text{err})}(\delta, \Delta_{\text{step}} = \bar{\Delta}_{\text{step}})$ , and (b) error signal  $S_{T_2}^{(\text{err})}(\delta = 0, \Delta_{\text{step}})$ .

Note that this variant of GABRS is suitable also for CPT atomic clocks, where we can use as concomitant parameter  $\xi$  the varied frequency step  $\Delta_{\text{step}}$  during both Ramsey pulses in CPT-Ramsey spectroscopy.

### C. Autobalanced Ramsey spectroscopy with a varied pulse duration

For generality, let us describe a variant of GABRS, where the concomitant parameter  $\xi$  is equal to the varied

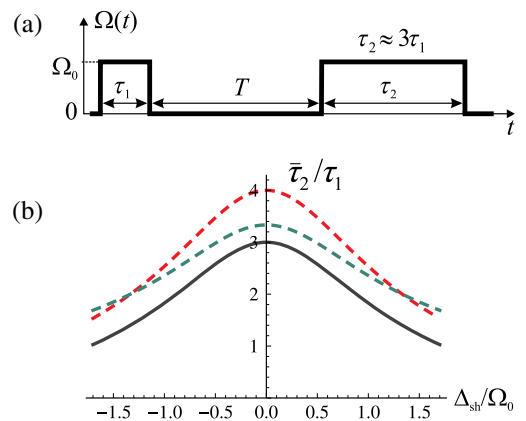


FIG. 6. (a) Schematic illustration of a Ramsey interrogation scheme for GABRS, where the concomitant parameter  $\xi$  is equal to the duration of the second pulse,  $\tau_2$ . (b) The dependence of stabilized pulse duration  $\bar{\tau}_2(\Delta_{\text{sh}})$  for different pulse areas:  $\Omega_0 \tau_1 = \pi/2$  (the black solid line),  $\Omega_0 \tau_1 = 0.8 \times \pi/2$  (the red dashed line), and  $\Omega_0 \tau_1 = 1.2 \times \pi/2$  (the green dashed line).



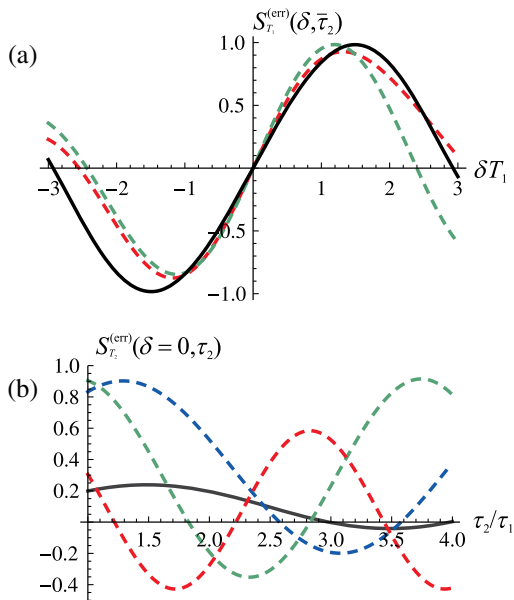


FIG. 7. Error signals under  $\Omega_0 T_1 = 2\pi$ ,  $\Omega_0 \tau_1 = \pi/2$ , and for different field-induced shifts of the clock transition during Ramsey pulses,  $\Delta_{sh}$ . (a) Error signal  $S_{T_1}^{(err)}(\delta, \bar{\tau}_2)$ :  $\Delta_{sh}/\Omega_0 = 0$  (the black solid line),  $\Delta_{sh}/\Omega_0 = 0.5$  (the red dashed line), and  $\Delta_{sh}/\Omega_0 = 1.0$  (the green dashed line). (b) Error signal  $S_{T_2}^{(err)}(\delta = 0, \tau_2)$ :  $\Delta_{sh}/\Omega_0 = 0.1$  (the black solid line),  $\Delta_{sh}/\Omega_0 = 0.5$  (the blue dashed line),  $\Delta_{sh}/\Omega_0 = 1.0$  (the green dashed line), and  $\Delta_{sh}/\Omega_0 = 1.5$  (the red dashed line). The case of  $\Delta_{sh}/\Omega_0 = 0$  is not included here because the error signal  $S_{T_2}^{(err)}(\delta = 0, \tau_2)$  approaches zero for any  $\tau_2$  value in this limit,  $|\Delta_{sh}/\Omega_0| \rightarrow 0$ .

duration of the second (as an example) Ramsey pulse  $\tau_2$ , while the pulse duration of the first Ramsey pulse  $\tau_1$  is fixed. For this method, we use the Ramsey sequence, which was considered in Ref. [22], where  $\tau_2 \approx 3\tau_1$  [Fig. 6(a)]. In this case, we always have  $\bar{\delta}_{clock} = 0$ , and the stabilized pulse duration  $\bar{\tau}_2$  is determined as the solution of Eq. (21). In the presence of the field-induced shift of the clock transition  $\Delta_{sh}$  during the Ramsey pulses, the duration  $\bar{\tau}_2$  is a function  $\bar{\tau}_2(\Delta_{sh})$  of the value  $\Delta_{sh}$ . These dependencies are presented in Fig. 6(b) for different pulse areas of the first Ramsey pulse,  $\Omega_0 \tau_1$ . The error signals  $S_{T_1}^{(err)}(\delta, \tau_2 = \bar{\tau}_2)$  and  $S_{T_2}^{(err)}(\delta = 0, \tau_2)$  for different values  $\Delta_{sh}$  are presented in Fig. 7.

Note that this variant of GABRS is not valid for CPT atomic clocks.

## V. CONCLUSION

In this paper, we develop a method and theoretical basis of GABRS, which allows for the elimination of probe-field-induced shifts in atomic clocks. This universal two-loop method requires the use of a concomitant parameter  $\xi$  in addition to the clock frequency  $\omega$ , which is related to the

first and/or second Ramsey pulses  $\tau_1$  and  $\tau_2$  through the use of interleaved Ramsey sequences with two different dark times,  $T_1$  and  $T_2$ . A correlated stabilization of both variable parameters can be achieved. It is analytically shown that the GABRS method always leads to a zero field-induced shift of the stabilized frequency  $\omega$  in an atomic clock,  $\bar{\delta}_{clock} = 0$ , independent of the relaxation processes (including decoherence) and different imperfections in the interrogation procedure. Such robustness is a direct consequence of the phase-jump technique used to build an error signal in Ramsey spectroscopy. We consider several variants of GABRS with the use of different concomitant parameters  $\xi$ . It is found that the most optimal and universal variant is based on the frequency-step technique, where the concomitant parameter  $\xi$  is a varied additional frequency step  $\Delta_{step}$  during both Ramsey pulses,  $\tau_1$  and  $\tau_2$ . In this case, universal antisymmetrical error signals are generated, which result in a vanishing frequency shift even at a finite modulation amplitude.

Apart from the optical standards, the GABRS technique can be applied in any other atomic clocks using Ramsey spectroscopy for frequency stabilization (i.e., CPT and POP atomic clocks). Some variants of GABRS can also be applied to CPT atomic clocks using CPT-Ramsey spectroscopy of the two-photon dark resonance. Moreover, GABRS is valid for open systems, and therefore this technique can be exploited with more complex schemes, such as molecules for high-resolution molecular spectroscopy [36–38]. It is also possible that more complicated Ramsey pulse sequences (for example, hyper-Ramsey sequence [22]) could also take advantage of the generalized autobalanced techniques described here.

We believe that the implementation of the GABRS technique can lead to a significant improvement of the accuracy and long-term stability for a variety of types of atomic clocks. For example, the experimental results in Ref. [32] can be considered as a first confirmation of the GABRS theory developed in our paper. In addition, the authors of a very recent paper [39] use a two-loop method that is rather closely related to the GABRS technique because, in Ref. [39], the microwave field stabilization is extended to two variables (microwave frequency and amplitude) to minimize instability from the cavity-pulling effect. Also, it will be interesting to investigate the efficiency of the GABRS technique in the presence of probe-laser-intensity fluctuations, as it was recently done theoretically in Ref. [40] for hyper-Ramsey spectroscopy.

## ACKNOWLEDGMENTS

We thank Ch. Sanner, Ch. Tamm, E. Peik, N. Huntemann, K. Beloy, and C. Oates for the useful discussions and comments. This work was supported by the Russian Science Foundation (Grant No. 16-12-10147). A. V. T. was supported by the Russian Foundation for

Basic Research (Grant No. 18-02-00822). M. Yu. B. was supported by the Ministry of Education and Science of the Russian Federation (Grant No. 3.1326.2017/4.6) and the Russian Foundation for Basic Research (Grants No. 17-02-00570 and No. 16-32-60050 mol\_a\_dk). Contributions to this article by workers at NIST, an agency of the U.S. Government, are not subject to U.S. copyright.

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- [1] M. Schioppo, R. C. Brown, W. F. McGrew, N. Hinkley, R. J. Fasano, K. Beloy, T. H. Yoon, G. Milani, D. Nicolodi, J. A. Sherman, N. B. Phillips, C. W. Oates, and A. D. Ludlow, Ultrastable optical clock with two cold-atom ensembles, *Nat. Photonics* **11**, 48 (2017).
- [2] A. D. Ludlow, M. M. Boyd, J. Ye, E. Peik, and P. O. Schmidt, Optical atomic clocks, *Rev. Mod. Phys.* **87**, 637 (2015).
- [3] K. Hosaka, S. A. Webster, A. Stannard, B. R. Walton, H. S. Margolis, and P. Gill, Frequency measurement of the  $^2S_{1/2} - ^2F_{7/2}$  electric octupole transition in a single  $^{171}\text{Yb}^+$  ion, *Phys. Rev. A* **79**, 033403 (2009).
- [4] M. Fischer *et al.*, New Limits on the Drift of Fundamental Constants from Laboratory Measurements, *Phys. Rev. Lett.* **92**, 230802 (2004).
- [5] T. Badr, M. D. Plimmer, P. Juncar, M. E. Himbert, Y. Louyer, and D. J. E. Knight, Observation by two-photon laser spectroscopy of the  $4d^{10}5s^2S_{1/2} \rightarrow 4d^95s^2^2D_{5/2}$  clock transition in atomic silver, *Phys. Rev. A* **74**, 062509 (2006).
- [6] A. V. Taichenachev, V. I. Yudin, C. W. Oates, C. W. Hoyt, Z. W. Barber, and L. Hollberg, Magnetic Field-Induced Spectroscopy of Forbidden Optical Transitions with Application to Lattice-Based Optical Atomic Clocks, *Phys. Rev. Lett.* **96**, 083001 (2006).
- [7] Z. W. Barber, C. W. Hoyt, C. W. Oates, L. Hollberg, A. V. Taichenachev, and V. I. Yudin, Direct Excitation of the Forbidden Clock Transition in Neutral  $^{174}\text{Yb}$  Atoms Confined to an Optical Lattice, *Phys. Rev. Lett.* **96**, 083002 (2006).
- [8] T. M. Fortier, Y. Le Coq, J. E. Stalnaker, D. Ortega, S. A. Diddams, C. W. Oates, and L. Hollberg, Kilohertz-Resolution Spectroscopy of Cold Atoms with an Optical Frequency Comb, *Phys. Rev. Lett.* **97**, 163905 (2006).
- [9] M. C. Stowe, M. J. Thorpe, A. Pe'er, J. Ye, J. E. Stalnaker, V. Gerginov, and S. A. Diddams, Direct frequency comb spectroscopy, *Adv. At. Mol. Opt. Phys.* **55**, 1 (2008).
- [10] P. R. Hemmer, M. S. Shahriar, V. D. Natoli, and S. Ezekiel, ac Stark shifts in a two-zone Raman interaction, *J. Opt. Soc. Am. B* **6**, 1519 (1989).
- [11] S. M. Shahriar, P. R. Hemmer, D. P. Katz, A. Lee, and M. G. Prentiss, Dark-state-based three-element vector model for the stimulated Raman interaction, *Phys. Rev. A* **55**, 2272 (1997).
- [12] T. Zanon, S. Guerandel, E. de Clercq, D. Holleville, N. Dimarcq, and A. Clairon, High Contrast Ramsey Fringes with Coherent-Population-Trapping Pulses in a Double Lambda Atomic System, *Phys. Rev. Lett.* **94**, 193002 (2005).
- [13] G. S. Pati, Z. Warren, N. Yu, and M. S. Shahriar, Computational studies of light shift in a Raman-Ramsey interference-based atomic clock, *J. Opt. Soc. Am. B* **32**, 388 (2015).
- [14] M. A. Hafiz, G. Coget, P. Yun, S. Guerandel, E. de Clercq, and R. Boudot, A high-performance Raman-Ramsey Cs vapor cell atomic clock, *J. Appl. Phys.* **121**, 104903 (2017).
- [15] X. Liu, E. Ivanov, V. I. Yudin, J. Kitching, and E. A. Donley, Low-Drift Coherent Population Trapping Clock Based on Laser-Cooled Atoms and High-Coherence Excitation Fields, *Phys. Rev. Applied* **8**, 054001 (2017).
- [16] F.-X. Esnault, D. Holleville, N. Rossetto, S. Guerandel, and N. Dimarcq, High-stability compact atomic clock based on isotropic laser cooling, *Phys. Rev. A* **82**, 033436 (2010).
- [17] Peng Liu, Yanling Meng, Jinyin Wan, Xiumei Wang, Yaning Wang, Ling Xiao, Huadong Cheng, and Liang Liu, Scheme for a compact cold-atom clock based on diffuse laser cooling in a cylindrical cavity, *Phys. Rev. A* **92**, 062101 (2015).
- [18] S. Micalizio, C. E. Calosso, A. Godone, and F. Levi, Metrological characterization of the pulsed Rb clock with optical detection, *Metrologia* **49**, 425 (2012).
- [19] A. Godone, F. Levi, C. E. Calosso, and S. Micalizio, High-performing vapor-cell frequency standards, *Riv. Nuovo Cimento Soc. Ital. Fis.* **38**, 133 (2015).
- [20] N. F. Ramsey, A molecular beam resonance method with separated oscillating fields, *Phys. Rev.* **78**, 695 (1950).
- [21] M. H. Levitt, in *Encyclopedia of Nuclear Magnetic Resonance*, Vol. 2, edited by D. M. Grant and R. K. Harris (John Wiley & Sons, Chichester, England, 1996), p. 1396.
- [22] V. I. Yudin, A. V. Taichenachev, C. W. Oates, Z. W. Barber, N. D. Lemke, A. D. Ludlow, U. Sterr, Ch. Lisdat, and F. Riehle, Hyper-Ramsey spectroscopy of optical clock transitions, *Phys. Rev. A* **82**, 011804(R) (2010).
- [23] N. Huntemann, B. Lipphardt, M. Okhapkin, Chr. Tamm, E. Peik, A. V. Taichenachev, and V. I. Yudin, Generalized Ramsey Excitation Scheme with Suppressed Light Shift, *Phys. Rev. Lett.* **109**, 213002 (2012).
- [24] N. Huntemann, C. Sanner, B. Lipphardt, Chr. Tamm, and E. Peik, Single-Ion Atomic Clock with  $3 \times 10^{-18}$  Systematic Uncertainty, *Phys. Rev. Lett.* **116**, 063001 (2016).
- [25] R. Hobson, W. Bowden, S. A. King, P. E. G. Baird, I. R. Hill, and P. Gill, Modified hyper-Ramsey methods for the elimination of probe shifts in optical clocks, *Phys. Rev. A* **93**, 010501(R) (2016).
- [26] T. Zanon-Willette, S. Almonacil, E. de Clercq, A. D. Ludlow, and E. Arimondo, Quantum engineering of atomic phase shifts in optical clocks, *Phys. Rev. A* **90**, 053427 (2014).
- [27] T. Zanon-Willette, E. de Clercq, and E. Arimondo, Probe light-shift elimination in generalized hyper-Ramsey quantum clocks, *Phys. Rev. A* **93**, 042506 (2016).
- [28] V. I. Yudin, A. V. Taichenachev, M. Yu. Basalae, and T. Zanon-Willette, Synthetic frequency protocol for Ramsey spectroscopy of clock transitions, *Phys. Rev. A* **94**, 052505 (2016).
- [29] T. Zanon-Willette, V. I. Yudin, and A. V. Taichenachev, Generalized hyper-Ramsey resonance with separated oscillating fields, *Phys. Rev. A* **92**, 023416 (2015).
- [30] T. Zanon-Willette, R. Lefevre, A. V. Taichenachev, and V. I. Yudin, Universal interrogation protocol with zero

- probe-field-induced frequency shift for quantum clocks and high-accuracy spectroscopy, *Phys. Rev. A* **96**, 023408 (2017).
- [31] J. Morgenweg, I. Barmes, and K. S. E. Eikema, Ramsey-comb spectroscopy with intense ultrashort laser pulses, *Nat. Phys.* **10**, 30 (2014).
- [32] Ch. Sanner, N. Huntemann, R. Lange, Ch. Tamm, and E. Peik, Autobalanced Ramsey Spectroscopy, *Phys. Rev. Lett.* **120**, 053602 (2018).
- [33] A. Morinaga, F. Riehle, J. Ishikawa, and J. Helmcke, A Ca optical frequency standard: Frequency stabilization by means of nonlinear Ramsey resonances, *Appl. Phys. B* **48**, 165 (1989).
- [34] C. A. Greenhall and G. J. Dick, Local oscillator limited frequency stability for passive atomic frequency standards using square wave frequency modulation, *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* **47**, 1593 (2000).
- [35] A. V. Taichenachev, V. I. Yudin, C. W. Oates, Z. W. Barber, N. D. Lemke, A. D. Ludlow, U. Sterr, Ch. Lisdat, and F. Riehle, Compensation of field-induced frequency shifts in Ramsey spectroscopy of optical clock transitions, *JETP Lett.* **90**, 713 (2009).
- [36] S. Truppe, R. J. Hendricks, S. K. Tokunaga, H. J. Lewandowski, M. G. Kozlov, C. Henkel, E. A. Hinds, and M. R. Tarbutt, A search for varying fundamental constants using hertz-level frequency measurements of cold CH molecules, *Nat. Commun.* **4**, 2600 (2013).
- [37] S. K. Tokunaga, C. Stoeffler, F. Auguste, A. Shelkownikov, C. Daussy, A. Amy-Klein, C. Chardonnet, and B. Darquie, Probing weak force-induced parity violation by high-resolution mid-infrared molecular spectroscopy, *Mol. Phys.* **2013** 2363 ,**111**).
- [38] W. B. Cairncross, D. N. Gresh, M. Grau, K. C. Cossel, T. S. Roussy, Y. Ni, Y. Zhou, J. Ye, and E. A. Cornell, Precision Measurement of the Electron's Electric Dipole Moment Using Trapped Molecular Ions, *Phys. Rev. Lett.* **119**, 153001 (2017).
- [39] M. Gozzelino, S. Micalizio, F. Levi, A. Godone, and C. E. Calosso, Reducing cavity-pulling shift in Ramsey-operated compact clocks, [arXiv:1802.03200](https://arxiv.org/abs/1802.03200) [*IEEE Trans. Ultrason. Ferroelectr. Freq. Control* (to be published)].
- [40] K. Bely, Hyper-Ramsey spectroscopy with probe-laser-intensity fluctuations, *Phys. Rev. A* **97**, 031406(R) (2018).