

Frequency shift caused by the line-shape asymmetry of the resonance of coherent population trapping

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(Received 24 May 2023; accepted 27 June 2023; published 10 July 2023)

We theoretically study the frequency shift in atomic clocks caused by the line-shape asymmetry of coherent population trapping (CPT) resonance in a bichromatic laser field. This asymmetry arises due to the inequality of the resonant spectral components and nonzero one-photon detuning. The line-shape-asymmetry-induced shift depends on the intensity of the resonant fields, which leads to a degradation in long-term stability due to fluctuations of the laser field parameters. A frequency stabilization based on the harmonic modulation of two-photon detuning is considered. It is shown that the use of a high-frequency modulation (compared to the CPT resonance width, i.e., the Pound-Drever-Hall regime) makes it possible to significantly suppress this shift (by one to two orders of magnitude).

DOI: [10.1103/PhysRevA.108.013103](https://doi.org/10.1103/PhysRevA.108.013103)

I. INTRODUCTION

The resonance of coherent population trapping (CPT) [1–3] is the physical basis for precision small-size atomic clocks [4–8]. The development of such miniature devices with low power consumption and high metrological characteristics is of fundamental importance for a wide range of applications (navigation, communications, synchronization, metrology, etc.) [9–13].

One of the main negative factors limiting the long-term stability of CPT clocks is the light shifts induced by the probe laser field. In this paper, we investigate in detail the shift of the stabilized frequency caused by the line-shape asymmetry of the CPT resonance [14–18], which arises due to the imbalance in the amplitudes of the resonant light components. The dependence of this shift on the total light intensity and the relationship between the amplitudes of the CPT fields is established. A method for suppressing this shift by using a high modulation frequency (compared to the CPT resonance width) of the two-photon detuning for the generation of the error signal is proposed.

II. THEORETICAL MODEL

As a theoretical model, we consider a three-level Λ system (see Fig. 1) interacting with a bichromatic field,

$$E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + \text{c.c.} \quad (1)$$

The coherent population trapping resonance is excited under the condition that the frequency difference ($\omega_1 - \omega_2$) is scanned near the transition frequency ω_{hfs} between the lower

states of the Λ system. The temporal dynamics of the Λ system is described using the density matrix formalism

$$\hat{\rho}(t) = \sum_{m,n=1,2,3} |m\rangle \rho_{mn}(t) \langle n|. \quad (2)$$

In the rotating-wave approximation, the equations for the density matrix elements ρ_{mn} have the following form,

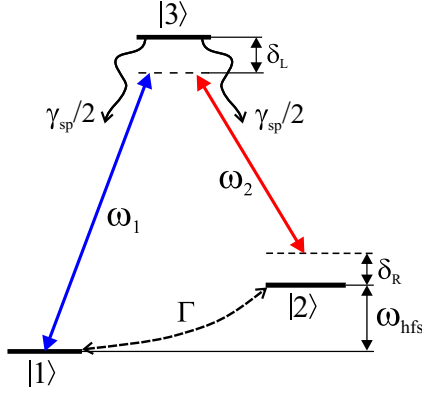
$$\begin{aligned} \partial_t \rho_{11} &= -\Gamma \left(\rho_{11} - \frac{1}{2} \right) + \frac{\gamma_{\text{sp}}}{2} \rho_{33} - i\Omega_1 \rho_{13} + i\Omega_1^* \rho_{31}, \\ \partial_t \rho_{22} &= -\Gamma \left(\rho_{22} - \frac{1}{2} \right) + \frac{\gamma_{\text{sp}}}{2} \rho_{33} - i\Omega_2 \rho_{23} + i\Omega_2^* \rho_{32}, \\ \partial_t \rho_{33} &= -(\gamma_{\text{sp}} + \Gamma) \rho_{33} + i\Omega_1 \rho_{13} - i\Omega_1^* \rho_{31} + i\Omega_2 \rho_{23} \\ &\quad - i\Omega_2^* \rho_{32}, \\ \partial_t \rho_{21} &= (-\Gamma + i\delta_{\text{R}}) \rho_{21} - i\Omega_1 \rho_{23} + i\Omega_2^* \rho_{31}, \\ \partial_t \rho_{31} &= (-\gamma_{\text{opt}} + i\delta_{\text{L}}) \rho_{31} + i\Omega_1 (\rho_{11} - \rho_{33}) + i\Omega_2 \rho_{21}, \\ \partial_t \rho_{32} &= (-\gamma_{\text{opt}} + i\delta_{\text{L}}) \rho_{32} + i\Omega_2 (\rho_{22} - \rho_{33}) + i\Omega_1 \rho_{12}, \\ \rho_{12} &= \rho_{21}^*, \quad \rho_{13} = \rho_{31}^*, \quad \rho_{23} = \rho_{32}^*. \end{aligned} \quad (3)$$

Taking into account the conservation of the total population, we supplement the equations system (3) with a normalization condition

$$\text{Tr}\{\rho\} = \rho_{11} + \rho_{22} + \rho_{33} = 1. \quad (4)$$

In Eqs. (3), we use the following notations: $\Omega_1 = d_{31} E_1 / \hbar$ and $\Omega_2 = d_{32} E_2 / \hbar$ are the Rabi frequencies for the transitions $|1\rangle \leftrightarrow |3\rangle$ and $|2\rangle \leftrightarrow |3\rangle$, respectively (where d_{31} and d_{32} are matrix elements of the electric dipole operator); $\delta_{\text{R}} = \omega_1 - \omega_2 - \omega_{\text{hfs}}$ is a two-photon (Raman) detuning; δ_{L} is a one-photon detuning; γ_{opt} is the damping rate of optical coherences (due to spontaneous decay processes, collisions with

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FIG. 1. The scheme of the three-level Λ system.

buffer gas atoms, etc.); γ_{sp} is the spontaneous decay rate of the excited state $|3\rangle$; the constant Γ describes the relaxation of atoms (for instance, due to transit effects) to an equilibrium isotropic distribution over the lower-energy levels $|1\rangle$ and $|2\rangle$.

As a spectroscopic signal, we will study the absorbed radiation power, which is proportional to the quantity

$$A(t) = \partial_t \rho_{33} + (\gamma_{\text{sp}} + \Gamma)\rho_{33}, \quad (5)$$

in the case of an optically thin medium.

III. LINE-SHAPE ASYMMETRY OF THE CPT RESONANCE

Under steady state conditions [when $\partial_t \rho_{mn} = 0$ in Eqs. (3)], the absorption signal (5), as shown in Ref. [19], is described by the following function of the two-photon detuning δ_R ,

$$\begin{aligned} A^{(\text{st})}(\delta_R) &= (\gamma_{\text{sp}} + \Gamma)\rho_{33} \\ &= C_0 + C_1 \frac{\tilde{\gamma}^2}{(\delta_R - \delta_0)^2 + \tilde{\gamma}^2} \\ &\quad + C_2 \frac{(\delta_R - \delta_0)\tilde{\gamma}}{(\delta_R - \delta_0)^2 + \tilde{\gamma}^2}, \end{aligned} \quad (6)$$

where the quantities C_0 , C_1 , C_2 , $\tilde{\gamma}$, and δ_0 depend on the model parameters (Ω_1 , Ω_2 , δ_L , γ_{sp} , γ_{opt} , Γ). The last term in expression (6) describes the asymmetry of the resonant line shape with respect to δ_R . The coefficient C_2 , which is responsible for the asymmetry of the CPT resonance, is nonzero under following simultaneously conditions [14,15],

$$\Omega_1 \neq \Omega_2, \quad \delta_L \neq 0, \quad (7)$$

i.e., the Rabi frequencies are not equal and the one-photon detuning is nonzero. In the case of small-size CPT clocks, condition (7) is almost always satisfied, because a vertical cavity surface emitting laser (VCSEL) with microwave modulation of the injection current generates an asymmetric spectrum [20–24]. The nonzero one-photon detuning ($\delta_L \neq 0$) is due to the presence of a hyperfine structure of the excited state in alkali metal atoms. For example, in the case of the ^{87}Rb D_1 line, this hyperfine splitting is 814.5 MHz [25]. Moreover, to increase the lifetime of the CPT state, a buffer gas is added to the cell with working atoms, which broadens the optical transition. As a result, the interaction of the CPT

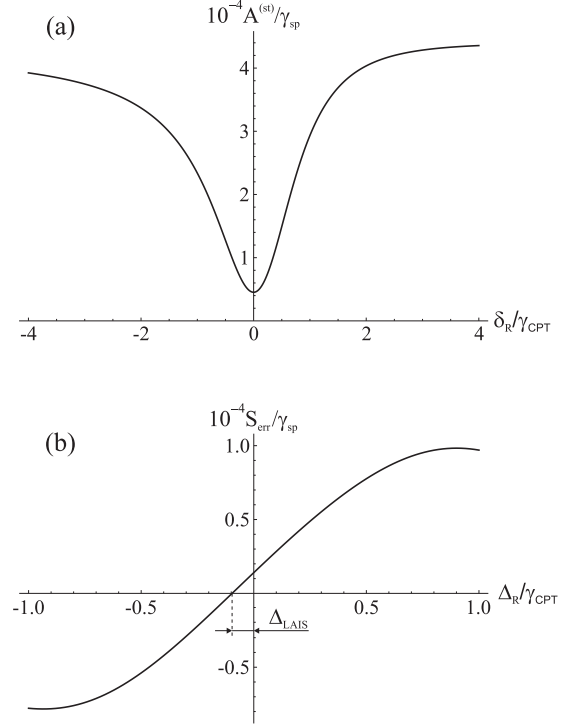


FIG. 2. The CPT resonance line shape under asymmetry conditions (i.e., $\delta_L \neq 0$ and $\Omega_1 \neq \Omega_2$ simultaneously): (a) absorption $A^{(\text{st})}$ under stationary excitation ($\delta_R \equiv \Delta_R$); (b) in-phase error signal ($\phi = 0$, $f = 0.1\gamma_{\text{CPT}}$, $F = \gamma_{\text{CPT}}$). Model parameters: $\Omega_1 = 0.1\gamma_{\text{sp}}$, $\Omega_2 = \sqrt{2}\Omega_1$, $\gamma_{\text{opt}} = 50\gamma_{\text{sp}}$, $\Gamma = 5 \times 10^{-5}\gamma_{\text{sp}}$, $\delta_{1\text{-ph}} = 0.5\gamma_{\text{opt}}$.

driving fields with the neighboring hyperfine energy levels of the excited state leads to the shift of the maximum of the optical transition line shape, which is used to stabilize the optical frequency. Therefore, we have a sufficiently large value of δ_L , for which the asymmetry of the CPT resonance can be significant.

In Fig. 2(a), the steady state line shape of the CPT resonance (6) is shown under asymmetry conditions (7). For convenience, the two-photon detuning is expressed in units of parameter γ_{CPT} ,

$$\gamma_{\text{CPT}} \approx \frac{\Gamma \left(1 + \frac{\Omega_1^2 + \Omega_2^2}{\Gamma\gamma_{\text{opt}}}\right)}{\sqrt{1 + \frac{12\Omega_1^2\Omega_2^2}{\Gamma\gamma_{\text{sp}}\gamma_{\text{opt}}^2} \left(1 + \frac{\Omega_1^2 + \Omega_2^2}{\Gamma\gamma_{\text{opt}}}\right)^{-1}}}, \quad (8)$$

which corresponds to the half width of the symmetric CPT resonance (at $\delta_L = 0$). Note that for the Λ system, the asymmetry of the line shape does not result in a shift of the resonance peak. Despite the fact that in expression (6) the symmetric (Lorentzian) and antisymmetric (dispersion) contours are shifted by $\delta_0 \neq 0$, their superposition leads to a line shape for which the extremum (minimum absorption) is at the point $\delta_R = 0$.

IV. FREQUENCY SHIFT IN ERROR SIGNAL

In practice, the stabilization of local oscillator frequency near the resonant frequency ω_{hfs} is carried out not by the resonance peak, but by the zero position of the error signal. In CPT

clocks, a method based on auxiliary harmonic modulation of the two-photon detuning,

$$\delta_R(t) = \Delta_R + F \cos(ft), \quad (9)$$

is widely used to generate the error signal, where Δ_R is the stabilized component of the two-photon detuning, and f and F are the modulation frequency and depth, respectively. In this case, the spectroscopic signal (5) becomes periodically time dependent $A(t+T) = A(t)$ with period $T = 2\pi/f$. The error signal $S_{\text{err}}(\Delta_R)$ is formed from the transmission signal using the synchronous detection technique, which can be written mathematically as follows,

$$S_{\text{err}}(\Delta_R) = \frac{1}{T} \int_0^T A(t) \cos(ft + \phi) dt, \quad (10)$$

where $\cos(ft + \phi)$ is the reference signal, and ϕ is the phase shift of the reference signal with respect to the harmonic law (9). In the case $\phi = 0, \pi$ the error signal (10) is usually called “in phase,” and for $\phi = \pm\pi/2$ the signal is called “quadrature.” The local oscillator frequency is stabilized near zero of the error signal:

$$S_{\text{err}}(\Delta_{\text{clock}}) = 0. \quad (11)$$

The accuracy and stability of the value Δ_{clock} determine the metrological characteristics of the atomic clocks. In context of the short-term stability, the slope of the linear part of the error signal at the center of the spectral line,

$$K = \left. \frac{\partial S_{\text{err}}}{\partial \delta_R} \right|_{\Delta_R = \Delta_{\text{clock}}}, \quad (12)$$

is an important parameter for the stabilization procedure.

In Fig. 2(b), the error signal (10) is shown under conditions of the line-shape asymmetry (7). Despite the unshifted resonance top, the position of the error signal zero is shifted by Δ_{LAIS} due to the asymmetry of the resonance wings, which we call a line-shape-asymmetry-induced shift (LAIS). The value of Δ_{LAIS} depends on the total light intensity and the ratio between the amplitudes of the spectral components (see Fig. 3). Therefore, random variations in the field amplitudes (for example, due to changes in the laser power or microwave signal power) will lead to fluctuations in the zero position of the error signal, and hence to a degradation in the long-term stability of CPT clock.

V. COMBINATION OF LINE-SHAPE-ASYMMETRY-INDUCED SHIFT AND ac STARK SHIFT

The zero position of the error signal is also affected by the well-known ac Stark shift of the reference transition frequency. Therefore, the total shift consists of two contributions (ac Stark and LAIS), each of which depends on the laser field intensity I and the microwave signal power P_{rf} . In the experiment, it is impossible to separate these shifts from each other, which significantly complicates their study in real conditions.

As is known (e.g., see Refs. [26,27]), the ac Stark shift $\Delta_{\text{ac}} = \eta I$ (where η is the coefficient of proportionality) can be suppressed by selecting the rf modulation index of laser field, for which the light shifts from different spectral components compensate each other (i.e., $\eta = 0$). It is possible to experimentally determine the power of the microwave signal that

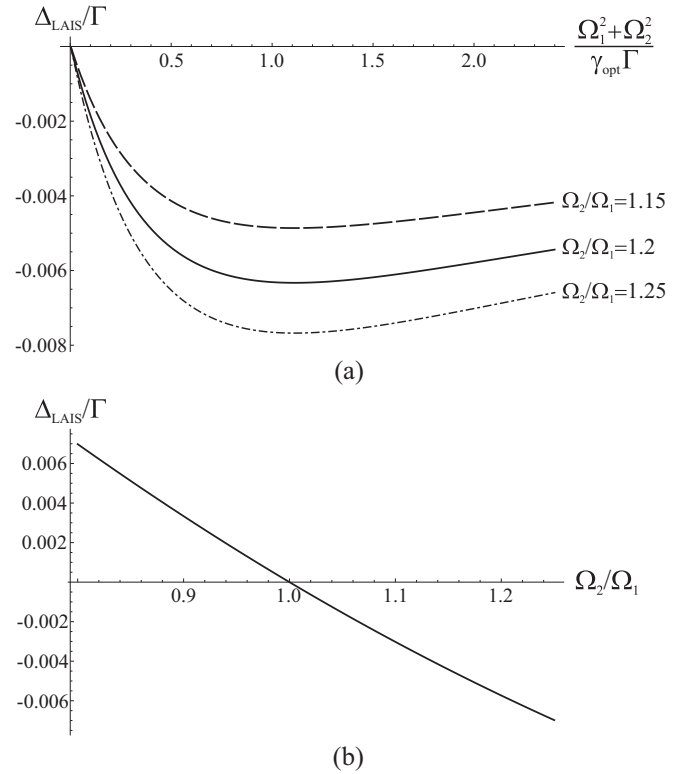


FIG. 3. (a) Dependence of the shift Δ_{LAIS} on the laser field intensity ($\Omega_1^2 + \Omega_2^2 \sim I$) for different ratios between the spectral components: $\Omega_2/\Omega_1 = 1.15$ (dashed line), $\Omega_2/\Omega_1 = 1.2$ (solid line), and $\Omega_2/\Omega_1 = 1.25$ (dashed-dotted line). (b) Dependence of the shift Δ_{LAIS} on the ratio between the spectral components Ω_2/Ω_1 at a fixed laser field intensity $(\Omega_1^2 + \Omega_2^2)/(\gamma_{\text{opt}}\Gamma) = 2$. Model parameters: $\gamma_{\text{opt}} = 50\gamma_{\text{sp}}$, $\Gamma = 5 \times 10^{-5}\gamma_{\text{sp}}$, $\delta_L = 0.2\gamma_{\text{opt}}$, $f = 0.1\Gamma$, $F = \Gamma$.

corresponds to this regime, using the modulation of the laser field intensity [28–30]. For example, in the case of harmonic modulation [28,29],

$$I(t) = I_0 + I_m \sin(\nu t), \quad (13)$$

the ac Stark shift also harmonically varies,

$$\Delta_{\text{ac}}(t) = \eta I(t) = \eta I_0 + \eta I_m \sin(\nu t). \quad (14)$$

Therefore, the value of the microwave signal power, at which there is no response of the stabilized frequency to intensity modulation $\propto \sin(\nu t)$, can be considered as the point of zero light shift, because $\eta = 0$. However, in experiments [29,30], two such points were found that corresponded to different values of the stabilized frequency (the difference exceeded several Hz). This fact seems contradictory, because in the absence of a light shift, the stabilized frequency should have the same value.

This result can be explained if we also take into account the frequency shift Δ_{LAIS} , which is associated with the asymmetry of the CPT resonance (see previous section). Indeed, since Δ_{LAIS} depends nonlinearly on the laser field intensity [see Fig. 3(a)], then in the case of harmonic intensity modulation (13), it can be approximately represented as an expression for the tangent at the point I_0 ,

$$\Delta_{\text{LAIS}}(t) \approx \Delta_{\text{bg}}(I_0) + \xi(I_0)I(t), \quad (15)$$

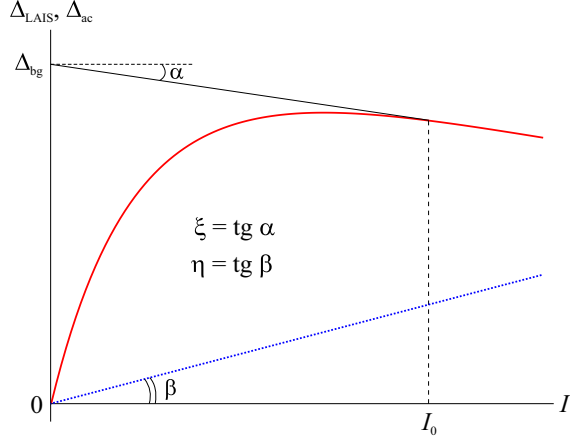


FIG. 4. Schematic view of the nonlinear shift Δ_{LAIS} (solid red line) and linear ac Stark shift Δ_{ac} (dashed blue line) as a function of the laser field intensity I .

if $I_m \ll I_0$. The coefficient $\xi(I_0)$ is the slope of the tangent at the point I_0 , and the quantity $\Delta_{\text{bg}}(I_0)$ corresponds to the intersection of the tangent with the vertical axis (see Fig. 4). Summing up (14) and (15), we get the time dependence for the full shift,

$$\begin{aligned} \Delta_{\text{full}}(t) &= \Delta_{\text{LAIS}}(t) + \Delta_{\text{ac}}(t) \\ &\approx \Delta_{\text{bg}}(I_0) + [\xi(I_0) + \eta]I(t), \end{aligned} \quad (16)$$

where all parameters η , $\xi(I_0)$, and $\Delta_{\text{bg}}(I_0)$ depend on the microwave power P_{rf} . Then, the absence of time variation of the total shift Δ_{full} for some value of the microwave power P_{rf} corresponds to the condition

$$\xi(I_0) + \eta = 0. \quad (17)$$

However, this does not mean that there is no shift at all, because under condition (17) the stationary part (residual stationary shift) remains in Eq. (16),

$$\Delta_{\text{full}} = \Delta_{\text{bg}}(I_0) \neq 0. \quad (18)$$

Then, in the case of several (two or more) points P_{rf} without temporal modulation of the full shift [i.e., condition (17) is satisfied], there are various residual stationary shifts (18), which are observed in the experiments [29,30].

Since the residual shift $\Delta_{\text{bg}}(I_0)$ can also depend on various environmental parameters and their fluctuations (temperature, pressure, etc.), the clock operation at the corresponding microwave power P_{rf} will not lead to the significant improvement of the long-term stability for CPT clock. Therefore, the suppression of the shift Δ_{LAIS} , induced by the line-shape asymmetry, is an important problem. Indeed, if $\Delta_{\text{LAIS}} = 0$, then the shift of the CPT resonance is determined only by ac Stark shift Δ_{ac} . In this case, the absence of time modulation of the resonance position corresponds to the condition $\eta = 0$, which in turn means no light shift at all:

$$\Delta_{\text{full}} = \Delta_{\text{ac}} = 0. \quad (19)$$

It is under these conditions that methods [28,29] for determining the appropriate microwave power P_{rf} will be the most

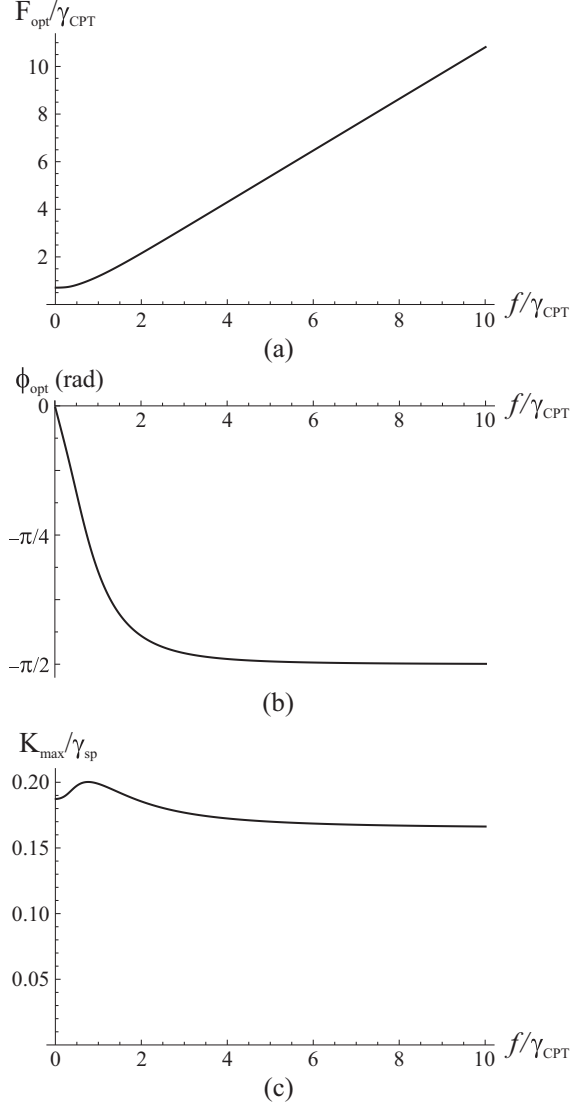


FIG. 5. Dependence of optimal parameters on modulation frequency: (a) frequency modulation depth; (b) phase of the reference signal; (c) corresponding slope of the error signal. Model parameters: $\Omega_1 = 0.05\gamma_{\text{sp}}$, $\Omega_2 = 1.2\Omega_1$, $\gamma_{\text{opt}} = 50\gamma_{\text{sp}}$, $\Gamma = 5 \times 10^{-5}\gamma_{\text{sp}}$, $\delta_{\text{L}} = 0$.

effective for achieving high metrological characteristics of atomic CPT clocks.

VI. SUPPRESSION OF FREQUENCY SHIFT CAUSED BY LINE-SHAPE ASYMMETRY

On the one hand, the influence of the line-shape asymmetry on the shift of the error signal can be suppressed by decrease the deviation depth F of the two-photon detuning (9), i.e., scanning the CPT resonance only near the top. However, in this case, the slope of the error signal (12) is significantly reduced and, consequently, the signal-to-noise ratio becomes worse, which negatively affects the frequency stability. Therefore, this approach is inefficient. On the other hand, we found that the modulation frequency f of two-photon detuning (9) plays a principal role for the shift Δ_{LAIS} and critically affects the slope of the error signal. As shown in Ref. [31], for each

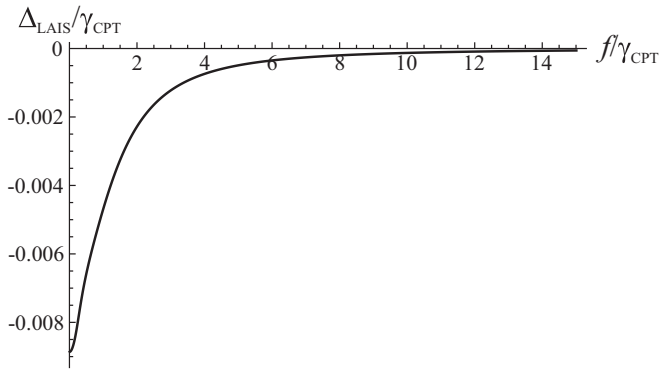


FIG. 6. Dependence of the zero position shift Δ_{LAIS} of the error signal on the modulation frequency f . Model parameters: $\Omega_1 = 0.05\gamma_{\text{sp}}$, $\Omega_2 = 1.2\Omega_1$, $\gamma_{\text{opt}} = 50\gamma_{\text{sp}}$, $\Gamma = 5 \times 10^{-5}\gamma_{\text{sp}}$, $\delta_L = 0.2\gamma_{\text{opt}}$, $F = F_{\text{opt}}$ [see Fig. 5(a)], $\phi = \phi_{\text{opt}}$ [see Fig. 5(b)].

frequency f there are optimal values of the deviation depth F_{opt} and the phase of the reference signal ϕ_{opt} , at which the slope of the error signal will be maximal K_{max} . In Fig. 5, the dependence of these parameters on the modulation frequency is shown.

In Fig. 6, the dependence of Δ_{LAIS} on the modulation frequency f of two-photon detuning is shown. The calculations were carried out for maximal slope of the error signal: $F = F_{\text{opt}}(f)$ and $\phi = \phi_{\text{opt}}(f)$ (see Fig. 5). As it is seen, if the modulation frequency f is much larger than the resonance width ($2\gamma_{\text{CPT}}$), then the shift Δ_{LAIS} is substantially suppressed. To quantitatively estimate the magnitude of this effect, we calculate the ratio of shifts for two values of the modulation frequency:

$$\frac{\Delta_{\text{LAIS}}(f = 15\gamma_{\text{CPT}})}{\Delta_{\text{LAIS}}(f = 0.5\gamma_{\text{CPT}})} \approx 0.009. \quad (20)$$

Thus, the use of high modulation frequency f (compared to the resonance width, i.e., the so-called Pound-Drever-Hall regime) makes it possible to reduce the shift Δ_{LAIS} by one to two orders of magnitude. At the same time, it is necessary to match the optimal modulation depth and phase of the reference signal in order to maximize the signal-to-noise ratio.

VII. CONCLUSION

In the present paper, we have studied the frequency shift in atomic clocks caused by the asymmetry of the CPT resonance line shape in a bichromatic laser field. This asymmetry arises due to inequality of the resonant spectral components and nonzero one-photon detuning. The dependence of the line-shape-asymmetry-induced shift on the laser field intensity and ratio between the amplitudes of the resonant spectral components is established. Note that the total frequency shift is the sum of the ac Stark shift and the line-shape-asymmetry-induced shift. In this case, the absence of a response of the stabilized frequency to the intensity modulation does not really mean the absence of a shift at all, since there is a residual stationary shift due to the line-shape asymmetry. This residual shift depends on various environmental parameters and their fluctuations, which negatively affect the metrological characteristics of atomic clocks. To suppress the shift caused by the line-shape asymmetry, we propose the use of a high modulation frequency (compared to the width of the CPT resonance) of the two-photon detuning for generation of the error signal.

ACKNOWLEDGMENT

This work was supported by the Russian Science Foundation (Grant No. 22-12-00279).

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