Cancellation of collisional frequency shifts in optical lattice clocks with Rabi spectroscopy

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Abstract

We analyze both the \(s\)- and \(p\)-wave collision induced frequency shifts and propose an over-\(\pi\) pulse scheme to cancel the shifts in optical lattice clocks interrogated by a Rabi pulse. The collisional frequency shifts are analytically solved as a function of the pulse area and the inhomogeneity of the Rabi frequencies. Experimentally measured collisional frequency shifts in an Yb optical lattice clock are in good agreement with the analytical calculations. Based on our analysis, the over-\(\pi\) pulse scheme for canceling the collision shift in optical lattice clocks using Rabi spectroscopy. We apply our analysis to both Sr and Yb optical lattice clocks by canceling the collisional frequency shift.

1. Introduction

Ultracold atoms trapped in an optical lattice have served as ample frameworks for high-precision atomic clocks [1–4], quantum information processing [5, 6], and for quantum simulators with ultracold quantum gases [7, 8]. Many atoms in the same optical lattice site inevitably lead to collisions among atoms; therefore, efforts to understand and control the collision dynamics are crucial to improve performance levels. For optical lattice clocks, the simultaneous interrogation of many atoms is a key feature in contrast to ion clocks, and a reduction of the collision shift is one of the most important factors to reach relative frequency uncertainty level of \(10^{-18}\). In atomic clocks using cold Fermi atoms, \(s\)-wave collision is first considered as the only origin of the collisional shift due to the inhomogeneity of Rabi frequencies since \(p\)-wave collision was assumed to be suppressed at low temperature [9–13]. However, it has been shown that \(p\)-wave collisions are even dominant in Yb and Sr lattice clocks [14, 15]. To overcome collisional shifts, the suppression of \(s\)-wave collisions by strong atom–atom interactions in a Sr 2D optical lattice [9] and precise measurements of the collision shift per atom number in a Sr 1D optical lattice with Rabi spectroscopy [16] were reported. In terms of cancellation of the frequency shift, the cancellation of \(s\)-wave collisional frequency shifts (SCFSs) by adjusting the second pulse area was demonstrated in Ramsey spectroscopy [17, 18]. Collision-shift cancellation in the strong \(p\)-wave collision case has also been demonstrated with Ramsey spectroscopy [14, 19]. However, in Rabi spectroscopy, which is the most widespread interrogation method in optical lattice clocks, the cancellation of the shift were investigated assuming only \(s\)-wave collisions [10, 12, 13]. Recently, the Rabi line shape distortion in the \(p\)-wave dominant case has been studied by observing many-body interactions [15]. But the cancellation of collision shift under strong \(p\)-wave collision has not been explored.

In this paper, we analyze both \(s\)- and \(p\)-wave cold collisions and obtained an analytic solution for the collision shift, for the first time, under typical clock operation conditions. We also propose an over-\(\pi\) pulse interrogation scheme for canceling the collision shift in optical lattice clocks using Rabi spectroscopy. We apply our analysis to the experimentally measured collisional frequency shift in an Yb optical lattice clock. We find out the \(p\)-wave collisions mainly determine the collisional frequency shift in an Yb optical lattice clock. We find out the \(p\)-wave p-phase shift, which is explained in detail below, should be small enough to cancel the collision shift. Although shift and uncertainty on the \(10^{-17}\) level are reported here due to operation at slightly off of collision-shift-free conditions, it is shown that shift and uncertainty on the \(10^{-18}\) level can be reached under experimentally achievable conditions, such as a small inhomogeneity below 0.1 in the Rabi frequencies of trapped atoms and a
small degree of lock servo-fluctuation (±1%), for both Yb and Sr lattice clocks when using an optimum over-π pulse.

2. Theoretical calculation

In actual 1D optical lattice cases, more than two atoms exist in a disk. Although a many-body Hamiltonian is required to describe the 1D case [13, 15, 20], the simple two-atom four-level Hamiltonian shown in equation (1) with effective collision-interaction frequencies can predict many-atom cases in the weak atom–atom interaction regime by adopting a mean field picture [14]. In order to calculate the collisional frequency shifts in Rabi spectroscopy, we solve the Schrödinger equation with the four-level Hamiltonian on a two-atom basis, in this case singlet (|eg⟩ = −1/√2(|e⟩ − |g⟩), and triplet states (|gg⟩, |ee⟩, and |eg⟩ = 1/√2(|e⟩ + |g⟩)):

\[
\frac{H}{\hbar} = \begin{pmatrix}
\delta + V_{eg} & 0 & \Omega/\sqrt{2} & \Delta \Omega/\sqrt{2} \\
0 & -\delta + V_e & \Omega/\sqrt{2} & -\Delta \Omega/\sqrt{2} \\
\Omega/\sqrt{2} & \Omega/\sqrt{2} & V_{ee} & 0 \\
\Delta \Omega/\sqrt{2} & -\Delta \Omega/\sqrt{2} & 0 & U_{ee}
\end{pmatrix},
\]

(1)

where \(V_{eg}\) and \(V_{ee}\), and \(U_{ee}\) correspond to the frequencies characterizing the p-wave collision shifts of the triplet states and the s-wave collision shifts of the singlet state, respectively. These frequency shifts are determined by the scattering lengths and the atomic density. \(\delta\) denotes the laser detuning with respect to the bare atom resonance frequency, \(\omega_{eg}\). \(\bar{\Omega}\) is the thermally averaged Rabi frequency of atoms in vibrational states in an optical lattice and \(\Delta\Omega\) is the RMS spread in the Rabi frequency, \(\Delta\Omega = \sqrt{\left\langle \Omega_{eg}^2 \right\rangle_T - \bar{\Omega}^2}\), where \(\left\langle \Omega_{eg}^2 \right\rangle_T\) is the thermal average of the square of the Rabi frequencies. We define the inhomogeneity of the Rabi frequency as \(\gamma = \Delta\Omega/\bar{\Omega}\).

The populations of the triplet and singlet states are projected to a single-atom basis, \(|e⟩\) and \(|g⟩\), as shown in figure 1(a), to obtain the e-g transition spectrum which is the expectation value of the projection operator, \(\bar{P}_e = \left\langle e|e⟩ + \frac{1}{2}|eg⟩ + \frac{1}{2}|ge⟩\right\rangle\). The collisional frequency shift of the clock transition is determined by measuring the peak position of the e-g transition Rabi spectrum, as shown in figure 1(b). The peak position is unshifted only at some specific pulse area. In experiments, the slightly detuned lasers from the peak of the Rabi spectrum are used to determine the peak position. The peak of the Rabi spectrum can be estimated by the averaged detuning \(\delta = \delta_{eg} \pm \delta_{col}\) satisfying \(P_e(\delta_0) = P_e(\delta_0)\) where \(\delta_0 = \delta_{col} \mp \delta_{fl}\) and \(\delta_{col} = \delta_{col} \pm \delta_{fl}\) are the detuning of the left and right side of the main peak positioned at \(\delta_{col}\), respectively (i.e. locking method). To achieve the resonance population transfer condition for an accurate determination of the peak position, we use the detuning \(\delta_{fl}\) as small as possible. In practical experiments, detuning for a 90% level is used due to shot to shot lock point variation.

As intuitively expected, the collisional frequency shift depends mainly on the time-averaged population difference (i.e. \(\int_0^T (N_g(t) - N_e(t)) dt/T\)), where \(T\) is the pulse duration of a Rabi pulse). This is shown in figures 1(b) and (c), where the X point (or Y point) with a positive (or negative) shift shows a positive (or negative) time-averaged population difference. Such a simple dependence on the time-averaged population difference of the collisional frequency shifts is under the condition that the homogeneous p-wave collisional shift is dominant and \(V_{eg} \gg V_{ee}, V_{gg}\), and \(U_{ee}\).

With the help of the second-order perturbation theory under the condition of \(\bar{\Omega} \gg \delta = \delta_{eg} \pm \delta_{col}\), we obtain the analytical formula of the collisional frequency shift (i.e. peak position) as a function of the pulse area \(A = \bar{\Omega} T\) when evaluated at resonance population transfer, \(\delta_t \to 0\). The collisional frequency shift is represented by the sum of a homogeneous p-wave, an inhomogeneous p-wave, and the SCFSs, \(\delta_{col} = \delta_{col}^{(p)} + \delta_{col}^{(q)} + \delta_{col}^{(h)}\). In a temperature of a few microkelvin in which most optical clocks are operated, p-wave collisions are not yet frozen out and have been found to dominate over s-wave collision [14].

The homogeneous p-wave collisional frequency shift (HPCFS) is the dominant term among the three types of collisional frequency shift. It comes from the g-e population difference due to the time-varying probabilities of the triplet states assisted by \(\bar{\Omega}\). It can be written as

\[
\delta_{col}^{(p)} \approx -\frac{\sin A}{A} (V_{gg} - V_{eg}) \alpha(A) + (V_{ee} - V_{eg}) \beta(A),
\]

(2)

where \(\alpha(A) = \left[-1 + \frac{\sin A}{A}\right]/(2\xi(A)), \beta(A) = \left[-2\sin^2(A/2) + \frac{\sin A}{A}\right]/\frac{\sin^4(A/2)}{A^2} - 3\right] \xi(A),\) and \(\xi(A) = 4\sin^2(A/2) - \frac{\sin A}{A}\). Because the time-averaged population difference is given by \(\int_0^T (N_g(t) - N_e(t)) dt/T \approx \sin A/A\) in the perturbative regime, it is revealed that HPCFS depends on the
time averaged population difference as shown in figure 2(a). In the infinitesimally weak pulse regime that \( T \to 0 \) (i.e. pulse area \( A \) approaches 0), the HPCFS has the finite value, \( \delta^{(p)}_{\text{col}} \to (V_{eg} - V_{gg}) \).

The inhomogeneous \( p \)-wave collisional frequency shift (IPCFS) is determined by the \( g \)–\( e \) population difference resulting from the leakage of the triplet-state population to the singlet state due to \( \Delta \Omega \). The inhomogeneity of Rabi frequencies opens an population transfer channel between the triplet states via the singlet state. This population transfer modifies the \( e \)–\( g \) spectrum. The IPCFS can be written as

\[
\delta^{(p)}_{\text{col}} \approx \gamma^2 \left[ \theta(A)(V_{eg} - V_{gg}) + \eta(A)(V_{ee} - V_{gg}) - \chi(A) V_{gg} \right],
\]

where \( \theta(A) = [-3 + 4A^2 + 2A^2 \cos A + 3 \cos 2A - A^3 \sin A]/(4A^2 \xi(A)) \),

\( \eta(A) = [75 - 6A^2 + 24(-3 + A^2) \cos A - 3 \cos 2A - 60A \sin A + 7A^2 \sin A]/(24A^2 \xi(A)) \), and

\( \chi(A) = [15 - 3A^2 + 2A^2 \cos A + 3 \cos 2A - A^3 \sin A]/(4A^2 \xi(A)) \)
The singlet-state $|eg\rangle$ populated according to the nonzero $\Delta \Omega$ undergoes an s-wave collision which is characterized by the interaction frequency $U_{eg}$. The SCFS is written as

$$\delta_{\text{col}}^{(p)} \approx \gamma^2 \chi(A) U_{eg}. \quad (4)$$

In the infinitesimally weak pulse regime and $N_e \rightarrow 1$, the SCFS is given as $\delta_{\text{col}}^{(p)} = \gamma^2 U_{eg}$. In that regime with the additional perturbation $\Omega \gg \Delta \Omega$, the two atom correlation function $G_{eg}(t)$ becomes $\gamma^2$. Therefore, the SCFS term is equivalent to the mean-field s-wave collision formula in [10] in the limit of infinitesimal excitation fraction. However, the two predictions disagree substantially when the excitation fraction is finite. Additionally, the SCFS given in [13] which shows more correct prediction of the s-wave collisional shifts corresponds to equation (4) up to the second order term $O(A^2)$ in the $\Omega \gg U_{eg}$ regime. Since the $\sin A/A$ term can be factored out of the SCFS, the SCFS is modulated by the time-averaged population difference and vanishes at $A = \pi$ [12, 13]. The SCFS is the smallest term among the three types of collision shifts.

We normalize the collisional frequency shift to $\delta_{\text{col}} = \Delta \tilde{\Omega} = (V_e - V_g)$ to verify the zero-crossing behavior of the collisional frequency shift independent of $V_e - V_g$. Each normalized collision shift is plotted in figure 2(a) under the condition of $\sigma = \frac{V_e - V_g}{V_e + V_g} = 0.4$. In the over-$\pi$ pulse regime, the normalized HPCFS crosses the zero shift line and becomes a negative value which is opposite to the positive value of the normalized IPCFS. A large inhomogeneity increases the magnitude of the normalized IPCFS, so that the total p-wave collisional frequency shift can not be zero. To realize the cancellation with the negative $\delta_{\text{col}}^{(p)}$ and the positive $\delta_{\text{col}}^{(p)}$, a sufficiently small inhomogeneity is required.

$\delta_{\text{col}}^{(p)}$ can be positive in the entire range of the pulse area in the case of a large $V_e - V_g$ values; hence the cancellation of the collisional frequency shifts is not possible in such a case. To check the necessary condition of $V_e - V_g$ for cancellation, i.e., $\delta_{\text{col}}^{(p)} < 0$, $\delta_{\text{col}}^{(p)}$ is calculated as a function of the pulse area and the ratio between the collision interaction frequencies $\sigma$, as shown in figure 2(b). The small value of $\sigma < 0.63$ is found to be a necessary condition for the cancellation of the collisional frequency shift. Fortunately, the cancellation of the collisional frequency shift is possible in both the Yb ($\sigma \approx 0.1$) and Sr ($\sigma \approx 0.4$) optical lattice clocks [14, 20].

We calculate the total collisional frequency shift as a function of the pulse area and the inhomogeneity in both Yb and Sr atoms using equations (2)–(4). Figures 3(a) and (b) show the existence of the zero-collisional frequency shift in the over-$\pi$ pulse regime. Figures 3(c) and (d) show the normalized uncertainties in the $\tilde{\delta}_{\text{col}}(A, \gamma) = 0$ curves of Sr and Yb. Fluctuations in the atomic density, pulse area, and inhomogeneity are the main origins of the residual collisional frequency shift change. The normalized deviations of the residual collisional frequency shift changes (i.e. normalized uncertainties) are calculated by

$$\tilde{u}_{\text{col}} = \left( \frac{\delta_{\text{col}}(\sigma \rho)}{\rho} \right)^2 + \left( \frac{\partial \delta_{\text{col}}}{\partial \sigma} \delta A \right)^2 + \left( \frac{\partial \delta_{\text{col}}}{\partial \gamma} \delta \gamma \right)^2. \quad (5)$$

For these calculations, it is assumed that the fluctuation of the atomic density $\delta \rho$ is 18%, the fluctuation of the pulse area $\delta A$ is on the 0.1% level, and the fluctuation of the inhomogeneity $\delta \gamma$ is 5%, all of which are typically obtained in experiments without state-of-the-art components. In the collision shift-free $\tilde{\delta}_{\text{col}}(A, \gamma) = 0$ curve, the uncertainty stemming from the fluctuation of
the atomic density, \( \tilde{\rho} \), disappears. Table 1 shows the conditions for \( \tilde{\rho} \) as depicted by the O-R line in figure 3. Under the Hz level where the atomic density is on the order of \( 10^{10} \) cm\(^{-3} \) can provide \( 5 \times 10^{-18} \) uncertainty in this condition. This can be obtained by the large volume optical cavity of an optical lattice without sacrificing the S/N ratio.

### 3. Experimental description

To apply our analysis to experiments, we measure the collisional frequency shifts of an \(^{171}\)Yb optical lattice clock as a function of the pulse area in two inhomogeneity cases, \( \gamma = 0.23 \) and \( \gamma = 0.15 \), the latter being the lowest in our current experimental setup. The details of our Yb optical lattice clock are described in the literature \([21, 22]\).

The potential depth of the vertically oriented 1D optical lattice is \( U_{\text{E}} = 1850 \) \( \text{e} \), where \( E_\text{r} \) is the recoil energy of the optical lattice and the trap frequencies are \( \nu_\text{r} \approx 55 \text{ kHz} \) and \( \nu_\text{z} \approx 202 \text{ Hz} \), as measured by the sideband spectra \([23]\). The temperature of the system can also be extracted from the sideband spectra. The S/N ratio of the measured clock spectrum is over 50 (peak excited fraction divided by the background fraction fluctuation) at \( \pi \)-pulse condition and it is reduced by half at 1.4\( \pi \)-pulse case. Nearly 50 000 atoms are trapped in the optical lattice. The atom density here is roughly \( \rho_0 = 4.3 \times 10^{10} \text{ atoms cm}^{-3} \) where \( V_{\text{g},\text{bg}} \) and \( U_{\text{bg}} \) are sub-Hz levels.
measure the collisional frequency shift at the given atom density, the interlacing between the high \( N_d \) and low atom numbers \( 0.57 N_d \) is repeated. During the interlacing, mean frequency differences of the high density to the low density case and the low density to the high density case are recorded to remove the clock laser drift. The final excited fraction is locked at \( 90\% \pm 6\% \) of the maximum excited fraction by feedback to the computer-controlled AOM for the clock laser (a clock laser lock servo). The detuning for a 90\% lock is determined as follows: \( \delta = \Omega \left( \frac{1 - \sin^2(A/2)}{\sin^2(A/2)} - A \frac{\mu_B}{m} \right) \), where \( n = 0.9 \). When \( A = 1.4\pi \), \( \delta \approx 0.20 \Omega \). Therefore, the perturbation \( \Omega \gg \delta \) can be applied to explain the experimental data. The atomic sample is spin–polarized \( (m_F = 1/2 \) or \( -1/2 ) \) by properly detuned 556 nm optical pumping in the presence of an 8.8 Gauss bias magnetic field. The inhomogeneity of our system is 0.23 at a temperature of 4.9(0.4) \( \mu \)K. The inhomogeneity of the Rabi frequencies is derived from the Rabi oscillation. To decrease the inhomogeneity, atoms in higher vibrational states of the optical lattice have to be blown out [24]. We drop the intensity of the lattice laser to 53\% level, at which the potential depth reaches 98\( E_r \). After 20 ms, the intensity of the lattice laser is increased to the original level (the intensity down-up method). The inhomogeneity is lowered to 0.15, where the temperature is 3.9(0.3) \( \mu \)K. The time scale of changes of the lattice depth is under a few \( \mu S \), which is much smaller than the inverse of the trap frequencies, which implies non-adiabatic processes. The pulse area is controlled by the power of the clock laser at a fixed short pulse duration of \( T = 10 \) ms.

Such a short pulse duration gives large Rabi frequency for a fixed pulse area \( A \) and makes the experiment be performed in the perturbation regime that \( \Omega \gg V_{\text{d}} \) \( s \). From a view of the spectral width, the spectral width of the main peak of the Rabi spectrum is proportional to \( \Omega \) for a fixed pulse area. The short pulse guarantees the Rabi spectra from the triplets, the singlet, and non-interacting atoms are well overlapped. When a long Rabi pulse over \( 1 \) s is applied, the interaction frequency \( V_{\text{d}} \) and \( U_{\text{eg}} \) is larger than the spectral width. In such a beyond-perturbation regime, \( \Omega < V_{\text{d}} \), the Rabi spectrum is distorted and the shift starts to stray from the mean-field approach [20]. The shift is not clearly defined in the distorted Rabi line shape. In an atomic clock based on the Rabi spectroscopy, its operation in the perturbative regime is appropriate in order to estimate systematic shifts via a Rabi spectrum. In the \( \Omega \ll V_{\text{d}} \) regime, the multiple peaks at the interaction energies of the singlet, the triplet, and non-interacting atoms can be resolved in a Rabi spectrum. In this regime, our cancellation scheme does not work. Instead of using the over-\( \pi \) cancellation scheme, the zero collisional frequency shift can be achieved by determining the peak position of a resolved Rabi spectrum of the non-interacting atoms.

4. Results and discussion

Figure 4(a) shows the experimental and calculated normalized collisional frequency shifts as a function of the pulse area. We try to extract the interaction energies from fitting the experimental data with the analytical formula by using the nonlinear regression. The fitting parameters are \( V_{\text{d}} \) \( \neq \) \( V_{\text{eg}} \), \( V_{\text{d}} \approx \approx V_{\text{eg}} \), and \( U_{\text{eg}} \approx \approx V_{\text{eg}} \) and are shown in table 2. The negative \( V_{\text{d}} \approx \approx V_{\text{eg}} \) is obtained, which is consistent with the result of Ramsey spectroscopy [14]. The non-zero \( U_{\text{eg}} \) however, is not guaranteed. The analytical solutions show good agreement with the experimental data.

The only s-wave collision shift \( \delta_{\text{loc}} / U_{\text{eg}} \) (the green dotted line) does not explain the measured shift and therefore we confirm that the p-wave collisions are dominant, which is consistent with the result of Ramsey spectroscopy [14]. Although the measured shift does not cross the zero shift mark due to the inhomogeneity limit of 0.15 in our experimental setup, the calculation proves that over-\( \pi \) pulse cancellation is possible at an inhomogeneity level of less than 0.15, as depicted by the blue dashed–dotted line shown in figure 4. The lower inhomogeneity is attainable via an intensity down-up operation of the optical lattice, as described in our experimental description.

Figure 4(b) shows the two Rabi spectra of \( A = \pi \) and \( A = 1.4\pi \). The use of an over-\( \pi \) pulse reduces the height of the main peak of an spectrum. On the other hands, the FWHM of the main peak decreases slightly in the over-\( \pi \) pulse regime. In the experiment, the frequency of the main peak is estimated by the multiply of the excited fraction–frequency slope at the lock point and the excited fraction difference between two lock points. In the over-\( \pi \) pulse regime having the relatively larger slope than the \( \pi \) pulse case, there is a possibility that the measured shift have a larger error which comes from the non-common population deviations at the two lock points. The Rabi spectrum at \( A = 1.4\pi \) shows requirement of the high lock point. At a larger pulse area over \( \pi \), the side structure can be over a half of the main peak. Therefore the lock level under 50\% disturbs the locking at the main peak. A close lock point to a 100\% level enables lock to the main peak and determination of the peak position as an averaged detuning.

The frequency noise of the clock laser and lock-servo error cause the population deviation from the 90\% lock point, therefore induce the unwanted lock point variation which is one of shift sources. In the experiment, the additional uncertainty \( \delta_{\text{lock}} \) from the \( \pm 6\% \) fluctuation of the 90\% lock point leads to the residual frequency shift.
change \( u_{\text{lock}} \) of a \( 10^{-17} \) level. The residual collisional frequency shift change due to lock point variation of \( \pm 6\% \) is written as \( u_{\text{lock}} \). The residual collisional frequency shift change due to lock point variation of \( \pm 6\% \) is written as \( u_{\text{col}} \).

Figure 4(c) shows the Allan deviation of measurement of the collisional frequency shift \( 7.7 \times 10^{-15}/\sqrt{\tau} \). (d) Five independent measurements of the collisional shift at the 1.4\( \pi \) pulse area.

Table 2. Interaction energies from fitting the experimental data with the analytical formula, \( \Delta_{\text{col}} \). The values in parenthesis indicate the 95% confidence interval.

| \( \gamma \) | \( V_{\text{ex}} - V_{\text{gg}} \) (Hz) | \( \sigma \) | \( \frac{\Delta_{\text{ex}} - \Delta_{\text{gg}}}{\Delta_{\text{gg}} - \Delta_{\text{ex}}} \) |
|---|---|---|---|
| 0.23 | -0.30 (\( \pm 0.05 \)) | 0.51 (\( \pm 0.29 \)) | -0.36 (\( \pm 0.75 \)) |
| 0.15 | -0.45 (\( \pm 0.04 \)) | 0.51 (\( \pm 0.12 \)) | 0.02 (\( \pm 0.88 \)) |

change \( u_{\text{lock}} \) of a \( 10^{-17} \) level. The residual collisional frequency shift change due to lock point variation of \( \pm 6\% \) is written as \( u_{\text{lock}} \).

Figure 4(c) shows the Allan deviation of measurement of the collisional frequency shift. The measurement instability is \( 7.7 \times 10^{-15}/\sqrt{\tau} \) where \( \tau \) is the averaging time, which is determined by the clock laser stability.

After about 14 000 s, the statistical error goes under 33 mHz. We repeat five independent measurements at the 1.4\( \pi \) pulse area as shown in figure 4(d). The \( A = 1.4\pi \) pulse induces a collisional frequency shift closest to zero at an inhomogeneity level of 0.15. The shifts and uncertainties of the experiment are given in table 3. Comparing \( \Delta_{\text{col}} \) of the \( A = 1.4\pi \) case with that of the \( A = 0.6\pi \) case, we show that the over-\( \pi \) pulse gives the system a lower level of total uncertainty by decreasing the uncertainty from the atomic density fluctuation at the expense of a small \( \Delta_{\text{lock}} \) increment.

A small lock point fluctuation below \( \pm 1\% \) and a small inhomogeneity below 0.1 are expected to provide the total uncertainty \( u_{\text{col}} \) less than \( 5 \times 10^{-18} \) when \( |V_{\text{ex}} - V_{\text{gg}}| \leq 0.6 \text{ Hz} \) using optimized pulse area.
5. Conclusion

In conclusion, we analyzed the collisional frequency shift in an optical lattice clock interrogated by a Rabi pulse. For the perturbative regime in which most optical lattice clocks are operated, the analytical solution was also obtained. It provides convenient expressions to explore the collisional shift and can be used to extract collision interaction energies in Rabi spectroscopy. Based on our analysis, we propose that an over-π pulse combined with a small inhomogeneity enables the cancellation of the total collisional frequency shift. This shows the potential for an optical lattice clock with a 10⁻¹⁸ uncertainty level with Rabi spectroscopy.

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Table 3. Uncertainty budget of the experiment where the interaction energies are given as follows: \( V_{\gamma R} - V_{\gamma G} = -0.45 \text{ Hz}, \) \( V_{\gamma G} - V_{\gamma E} = 0.51 (V_{\gamma R} - V_{\gamma G}) \), and \( V_{\gamma E} - V_{\gamma G} = 0.02 (V_{\gamma R} - V_{\gamma G}) \) which are used to plot figure 4(a).

| \( \Lambda \) | \( \gamma \) | \( \xi_{\text{coll}} \) \((\times 10^{-3})\) | \( \delta_{\text{coll}} \) \((\times 10^{-3})\) | \( \delta_{\text{lock}} \) \((\times 10^{-3})\) | \( \delta_{\text{tot}} \) \((\times 10^{-3})\) | \( \eta_{\text{tot}} \) \((\times 10^{-14})\) |
|---|---|---|---|---|---|---|
| 0.6\( \pi \) | 0.15 | 666 | 120 | 9 | 120 | 104 |
| 1.4\( \pi \) | 0.15 | 78 | 21 | 18 | 28 | 24 |