Initial atomic coherences and Ramsey frequency pulling in fountain clocks

Vladislav Gerginov,* Nils Nemitz,† and Stefan Weyers
Physikalisch-Technische Bundesanstalt, Bundesallee 100, 38116 Braunschweig, Germany
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In the uncertainty budget of primary atomic cesium fountain clocks, evaluations of frequency-pulling shifts of the hyperfine clock transition caused by unintentional excitation of its nearby transitions (Rabi and Ramsey pulling) have been based so far on an approach developed for cesium beam clocks. We re-evaluate this type of frequency pulling in fountain clocks and pay particular attention to the effect of initial coherent atomic states. We find significantly enhanced frequency shifts caused by Ramsey pulling due to sublevel population imbalance and corresponding coherences within the state-selected hyperfine component of the initial atom ground state. Such shifts are experimentally investigated in an atomic fountain clock and quantitative agreement with the predictions of the model is demonstrated.

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I. INTRODUCTION

Many high-resolution spectroscopy experiments such as coherent population trapping [1] and electromagnetically induced transparency [2] are based on intentionally created initial state coherences. The operating principle of cesium atomic clocks themselves relies on the Ramsey scheme [3], where the phase of an intentionally induced hyperfine state coherence is compared to the phase of the driving microwave radiation field after a free propagation time in the absence of microwave radiation. Here we consider the consequences of creating unintentional initial coherent superpositions between the magnetic sublevels belonging to the same ground-state hyperfine component (Zeeman coherences) before the first Ramsey interaction in cesium fountain clocks [4].

This type of cesium clock provides the most precise realization of the SI unit of time, the second, which is defined by the unperturbed transition frequency between the two hyperfine ground-state components of the cesium atom. The systematic uncertainty of fountain clocks reaches a few parts in $10^{-16}$ [5–8] and cryogenic sapphire oscillators [9] or optically stabilized microwave oscillators [10,11] allow statistical uncertainties also at the level of a few parts in $10^{-16}$ after 1 day of operation. These developments necessitate a progressively careful re-evaluation of effects that previously contributed only marginally to the overall uncertainty budget, such as frequency shifts due to unwanted hyperfine transitions neighboring the clock transition (namely, Rabi and Ramsey pulling).

Ramsey-pulling frequency shifts appear when the microwave field driving the clock transition couples magnetically sensitive states to the clock states via $\Delta m_F = \pm 1$ (or $\sigma$) transitions, which change the projection quantum number $m_F$ of the atom’s total angular momentum $F$ [12]. The shift is qualitatively different from the Rabi-pulling shift, which is due to the microwave field driving neighboring $\Delta m_F = 0$ (or $\pi$) transitions between magnetically sensitive states without coupling to a clock state [13]. Work on Ramsey pulling has been published only for thermal beam clocks [12,14,15]. So far its contribution to the uncertainty budget of fountain clocks has been evaluated based on the theory specifically developed for thermal beam clocks by Cutler et al. [12] and is generally estimated to be below $10^{-16}$ when atomic state selection is employed. However, the underlying theory for this estimate assumes a broad atom velocity distribution, constant amplitude and direction of the magnetic microwave field components during the Ramsey interactions, low microwave amplitudes, and incoherent initial atomic states. In contrast, in atomic fountains a narrow velocity distribution is achieved due to the laser cooling, and the amplitude and direction of the magnetic microwave field components are significantly position dependent in fountain cavities. The state selection is performed by atom interaction with a microwave field, which typically generates atomic coherences. Also, experimental uncertainty evaluations often use frequency shift measurements at elevated microwave amplitudes [5, 16–18].

This work extends the Ramsey-pulling theory for beam clocks presented in Ref. [12], to better represent the case of atomic fountains and to include the effects of initial Zeeman coherences. For beam clocks large frequency shifts have been demonstrated and explained, when initial Zeeman coherences are caused by Majorana transitions and undesired, but unavoidable, $\sigma$ transitions occur in the Ramsey cavity [19]. Independent of this work, Ramsey pulling in the presence of initial coherences caused by optical pumping has been discussed for thermal beam clocks [15]. In fountain clocks initial Zeeman coherences are generally created during the state preparation process itself, before the clock transition takes place. We demonstrate that, as a result, in the case of broken symmetry between the initially coherently populated $m_F = +1$ and $m_F = -1$ states, correspondingly large frequency shifts can occur. This is confirmed by direct experimental observation, in good agreement with the theory. The Rabi-pulling effect [13], causing a frequency shift smaller in magnitude than the Ramsey-pulling shift, is naturally included in the theoretical model. The theory gives an improved uncertainty estimate for the Physikalisch-Technische Bundesanstalt (PTB) fountain clocks CSF1 and CSF2.

Our discussion presents the perspective that frequency shifts due to Majorana transitions before the first Ramsey
interaction [19] should be considered as Ramsey pulling involving initial asymmetric Zeeman coherences. On the other hand, the effect of Majorana transitions occurring after the second Ramsey interaction [20] is to increase the frequency shifting effect of the Ramsey pulling in combination with inhomogeneous atom state detection. We, finally, also note that interfering asymmetric initial Zeeman coherences may be present in optical frequency standards too, depending on the employed state preparation techniques [21].

The physical mechanisms of Rabi and Ramsey pulling and the employed theoretical model are described in Section II. Section III presents basic symmetries and the calculated magnetic field dependence of Ramsey pulling, and Sec. IV the calculated microwave amplitude and population dependences. The measured Ramsey-pulling shifts in the presence of initial Zeeman coherences are described in Sec. V and compared with the results of the calculation. Finally, in Sec. VI the contribution of the combined Rabi- and Ramsey-pulling shifts to the uncertainty budget of the PTB fountain clocks CSF1 and CSF2 is discussed. Appendix A gives the detailed derivation and the explicit form of the total Hamiltonian. Appendix B gives the explicit position dependence of the magnetic microwave field components assumed for the microwave cavities of CSF1 and CSF2. Appendix C describes details of the calculation related to the use of two-dimensional (2D) atom clouds.

II. THEORETICAL MODELING OF RABI AND RAMSEY PULLING

We focus only on the six most relevant atomic states for the case of cesium clocks, namely, states with quantum numbers $F = 3$, $F = 4$, and $m_F = -1, 0, +1$. The energy diagram of these states together with the clock transition between the $|3, 0\rangle$ and the $|4, 0\rangle$ states is shown in Fig. 1, which also indicates the energy shift given by the Breit-Rabi formula [22]. The depicted off-resonant $\Delta m_F = 0 \pi$ transitions (caused by microwave magnetic field components along the vertical homogeneous magnetic quantization field) with $m_F = \pm 1$ are responsible for the Rabi-pulling frequency shift: An asymmetric $m_F = \pm 1$ state population of incoming $F = 3$ atoms leads to an asymmetric incoherent sum of the transition probabilities for $m_F = \pm 1$ atoms and the transition probability for $m_F = 0$ atoms and, thus, to a frequency shift.

![Energy diagram of the relevant atomic states of the cesium atom. Transitions due to the microwave field component in the direction of the quantization axis ($\pi$ transitions) are shown by solid lines; those due to the microwave field component in the transverse direction ($\sigma$ transitions), by dotted lines. The first-order Zeeman effect is also shown. The $g$ factor of the cesium ground state $F = 4$ component is $g_F \approx 1/4$.](image)

The off-resonant $\Delta m_F = \pm 1 \sigma$ transitions in Fig. 1, responsible for the Ramsey-pulling shift, are of primary interest in this work. Starting from the three $F = 3$ states, this type of transition, caused by transverse microwave magnetic field components (perpendicular to the vertical homogeneous magnetic quantization field) in the Ramsey cavity of cesium clocks, together with the $\pi$ transitions leads to a coherent excitation of the three $F = 4$ states. As a result, measured clock transition frequency shifts occur when there is an asymmetric $m_F = \pm 1$ state population of incoming $F = 3$ atoms or if atoms in $|4, \pm 1\rangle$ states are detected with different efficiencies.

To calculate frequency shifts caused by Rabi and Ramsey pulling in an atomic fountain, we find the time evolution of a cesium atomic ensemble represented by the density matrix $\rho$ by solving the von Neumann equation,

$$i\hbar \frac{d\rho(t)}{dt} = [(H_0 + H_{RF}), \rho(t)],$$

where the total Hamiltonian of the cesium atomic system in the presence of a static magnetic field $\vec{B}$ pointing along the $z$ direction and a microwave magnetic field $\vec{B}_{RF}^s$ is composed of the atomic Hamiltonian $H_0$ describing both the hyperfine and the static magnetic field interactions and the Hamiltonian $H_{RF}$ describing the microwave interaction (see Appendix A for details).

For the modeling of Rabi and Ramsey pulling we consider a fountain with a spatial dependence of the microwave field components inside the state selection and Ramsey cavities given in Appendix B. In the presence of the microwave field in the state selection and Ramsey cavities the von Neumann equation is solved numerically, while during periods without microwave fields it is solved analytically. We calculated the trajectory of each atom ensemble from a given initial position and velocity of that ensemble. Each considered trajectory passes twice through the lower aperture of the state selection cavity. The velocity of the atom ensemble during a cavity passage is assumed constant and is calculated from the duration of the passage and the cavity dimensions. The relevant spatial microwave field components $B_{RF}^s (k = x, y, z)$ and corresponding Rabi frequencies $\nu_k = g_k B_{RF}^s (g_F - g_I)/2\hbar$ (see Appendix A) are calculated from the horizontal positions using Eqs. (B1) and (B2), respectively. The time sequence of the atom-microwave interactions and free propagations is shown in Fig. 2, together with a sketch of the corresponding amplitudes of the magnetic microwave field components.

Before the state selection interaction all elements of the initial density matrix are 0, except for the diagonal elements describing the populations of the $|4, 0\rangle$ and $|4, \pm 1\rangle$ states. This describes an atomic ensemble created at the end of the launch phase through optical pumping by the molasses lasers. After solving Eq. (1) for the state selection microwave interaction for a given trajectory, the atom ensemble is projected on the $F = 3$ ground-state component. This is the equivalent of the experimental state selection process, where (in the ideal case) atoms are selectively transferred to the $|3, 0\rangle$ state and any atoms remaining in the initially populated $F = 4$ ground-state component are removed by radiation pressure from a clearing laser pulse.

The further internal evolution of the atom ensemble is determined from the solution of Eq. (1) for the subsequent
free propagations and Ramsey microwave interactions. For the complete fountain cycle shown in Fig. 2, the final density matrix is used to determine the probability that the atom is in the $F = 4$ ground-state component. To simulate the behavior of a real fountain clock, we define the clock transition probability as $p = \sum_{m_F=-1}^{1} \rho^{F}_{m_F,m_F}$, averaged for all atomic ensembles, each with a different trajectory. Here, $\rho^{F}_{m_F,m_F'}$ is the corresponding element of the $6 \times 6$ density matrix for states belonging to the $F$ ground-state component, obtained by the unitary transformation given in Appendix A. The resonant effective Rabi frequencies $b_0^F$ in the state selection cavity and $b_1$ in the Ramsey cavity (as defined in Appendix B) were numerically adjusted in the simulation to yield a maximum average transition probability after resonant state selection and Ramsey microwave interactions.

Finally, a set of four probabilities $p_0$, $p_\pi$, and $p_{\pm\pi/2}$ is calculated for phase changes of 0, $\pi$, and $\pm\pi/2$ of the resonant microwave field component between the two Ramsey interactions. The probabilities $p_0$ and $p_\pi$ are used to calculate the contrast of the Ramsey fringe. The probabilities $p_{\pm\pi/2}$ then give the frequency shift (assumed to be much smaller than the fringe width of approximately 1 Hz) of the transition frequency from the unperturbed clock transition frequency as $(p_{\pi/2} - p_{-\pi/2})/(2\pi T_R(p_0 - p_\pi))$, with $T_R$ the Ramsey time corresponding to the center of the atom cloud.

A typical clock transition probability as a function of the microwave detuning, measured with CSF2 [18], is shown in Fig. 3 together with the result of a simulation based on the six-level model described here. The contributions of detection cross-talk and hyperfine pumping in the detection zone, estimated through measurements, were taken into account in the simulation by appropriate rescaling of the transition probability.

III. BASIC SYMMETRIES AND MAGNETIC FIELD DEPENDENCE OF RAMSEY PULLING

To better understand the basic dependences of Ramsey pulling and to avoid additional complexity from incorporating the fountain state selection process, first, we performed calculations which included only the two microwave Ramsey interactions and the free propagation between them. Therefore in the calculations in this section and in Sec. IV the initial atomic state of the atom ensemble at the beginning of the first Ramsey interaction was defined by setting the appropriate elements of the density matrix as described below. We consider a hypothetical situation where an atom cloud consists of multiple atom ensembles with positions distributed along a horizontal 2D grid with a radius $r_c$ equal to that of the cavity aperture and identical vertical trajectories. Such a 2D distribution of atom ensembles saves computation time by allowing the use of small numbers of ensembles and allows us to better check the basic symmetries of Ramsey pulling. However, some adjustments of the model are needed to ensure a realistic fringe contrast decrease and the correct dependence of Rabi and Ramsey pulling on the static magnetic field strength (see Appendix C).

Since our calculations confirmed the assumption that normally a symmetry between the $|3, \pm 1\rangle$ states does not lead to a frequency shift in the absence or presence of coherences, we considered asymmetric initial internal atomic states that correspond to 0 population in the $|3, -1\rangle$, 99% in the $|3, 0\rangle$, and 1% in the $|3, +1\rangle$ state, respectively, set through the diagonal elements of the initial density matrix. To explore the coherent effects, we included Zeeman coherences through the relevant off-diagonal matrix elements set according to

$$\rho^{3}_{(1,0)} = e^{i\varphi} \sqrt{\rho^{3}_{(0,0)} \rho^{3}_{(1,1)}},$$

$$\rho^{3}_{(-1,0)} = e^{i(\varphi - \psi)} \sqrt{\rho^{3}_{(0,0)} \rho^{3}_{(-1,-1)}},$$

which describes a maximally coherent initial state with phase $\varphi$.

The Ramsey-pulling shift depends on the direction of the transverse microwave field component driving the $\Delta m_F = \pm 1$ transitions causing the shift and on the phase of the initial

FIG. 2. Time sequence of the fountain clock cycle (not to scale), indicating the time intervals for the state selection cavity traversal $\tau_S$, the free propagation between state selection and the Ramsey cavity $T_{SR}$, the Ramsey cavity traversal $\tau_R$, and the free propagation between the two Ramsey cavity traversals $T_R$. The $B_{x, y, z}^{RF}$ magnetic components of the microwave field are schematically represented by the shaded areas. The $B_{x, y, z}^{RF}$ field component in the considered rectangular state selection cavities is 0 (see Appendix B).

FIG. 3. (Color online) Measured (gray curve) and calculated (red curve) clock transition probability as a function of the microwave frequency detuning for the fountain clock CSF2 in normal operation. The $\Delta m_F = 0$ transitions are labeled “$\pi$”; the $\Delta m_F = \pm 1$ transitions are labeled “$\sigma$.” Due to the large frequency span, the Ramsey fringe structures appear as filled areas.
coherences. We considered two specific spatial phase dependences of the coherences.

In the first case, we assumed that the phase $\varphi$ is the same for all atom ensembles (chosen to be $\varphi = 0$) and the relationship of the density matrix elements is given by Eq. (2). This might occur due to Majorana transitions from the $|3,0\rangle$ state to the $|3, \pm 1\rangle$ states [23] or due to interaction with a homogeneous microwave field on-resonance with the clock transition frequency and with a position-independent direction of the microwave field component. In the latter situation the two magnetically sensitive transitions are symmetrically detuned from resonance, and their corresponding matrix elements have equal magnitude and opposite sign (see Appendix A). In the second case, we chose the phase $\varphi$ in Eq. (2) to correspond to the azimuthal angle of the horizontal position of the atom ensemble. This situation is similar to the result from the state selection process in a cylindrical cavity with the microwave field on-resonance, where the transverse field components have this symmetry.

The cases considered are summarized in Fig. 4, including cases with horizontal cloud offset, obtained by shifting the entire distribution by 2 mm and then discarding the trajectories which are more than $r_c$ away from the fountain axis before the first Ramsey interaction. Also indicated are the calculated maximum magnitudes of the Ramsey-pulling frequency shift for static magnetic field $B_z = B_z^R$ values in and above the Ramsey cavity between 50 and 250 nT. The calculated frequency shifts as a function of $B_z$ are shown in Fig. 5, where experimental parameters listed in Table I were used in the calculations.

Case A is the typically considered Ramsey-pulling shift due to a population difference between the ensembles in the $|3, \pm 1\rangle$ states without initial Zeeman coherences. As shown in Fig. 5 (left scale), the shift is less than $4 \times 10^{-17}$ for magnetic field strengths between 50 and 250 nT and a 1% population imbalance.

When the cloud is horizontally offset from the cavity axis as in case B, the shift is slightly reduced, due to the removal of atom ensembles from the edge regions of the cavity aperture, where the transverse field components have a higher amplitude. It is worth mentioning here that the single-atom Ramsey- and Ramsey-pulling shifts for cases A and B would be above the $10^{-16}$ level for field strengths between 50 and 250 nT.

With a 1% population imbalance in the presence of coherences, but neither spatial dependence of their phase nor atom cloud horizontal offset (case C), the Ramsey-pulling shift is exactly the same as in case A. The potential enhancement of the frequency shift due to the coherences is canceled completely because of the symmetry of the atom ensemble positions: for two ensembles which for both Ramsey interactions have diametrically opposite horizontal positions in the Ramsey cavity with respect to the axis, the shifts due to the coherences have the same magnitudes but opposite signs. This is a result of the phase of the coherence $\rho_{1,0}$ being the same for the

![Diagram of the initial horizontal cloud positions](image1)

**FIG. 4.** Diagram of the initial horizontal cloud positions (shaded area) and atomic states. The circle represents a Ramsey cavity aperture of radius $r_c$. Top labels: the absence (light gray) or presence (dark gray or gray gradient) of coherences and their phase dependence on the azimuthal angle (gray gradient). Bottom labels: maximum magnitudes of the Ramsey-pulling frequency shift for magnetic field $B_z = B_z^R$ values between 50 and 250 nT.

![Graph of Ramsey-pulling shift vs. magnetic field](image2)

**FIG. 5.** (Color online) Ramsey-pulling shift as a function of the static magnetic field $B_z = B_z^S$ in and above the Ramsey cavity for a 1% population imbalance of the $|3, -1\rangle$ and $|3, +1\rangle$ states. The labels of the individual curves represent the initial conditions from Fig. 4: without coherences (cases A and B, left scale) and with coherences (case C, left scale; cases D–F, right scale).

![Table of experimental parameters](image3)

**TABLE I.** Parameters corresponding to the fountain CSF1 at PTB. The values $\tau_S$, $\tau_R$, $T_{SR}$, and $T_R$ correspond to the center of the atom cloud. $B_z^S$, $B_z^{SR}$, $B_z^R$, and $B_z^S$, and $B_z^R$, are the average magnetic field strengths in the state selection cavity, between the state selection and the Ramsey cavity, and in and above the Ramsey cavity. The effective Rabi frequencies $b_0^s$ and $b_0^r$ (defined in Appendix B) correspond to a $\pi$ pulse area (state selection cavity) and a $\pi / 2$ pulse area (Ramsey cavity).

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\tau_S$</td>
<td>7.2 ms</td>
</tr>
<tr>
<td>$T_{SR}$</td>
<td>24.2 ms</td>
</tr>
<tr>
<td>$\tau_R$</td>
<td>10.3 ms</td>
</tr>
<tr>
<td>$T_R$</td>
<td>0.5539 s</td>
</tr>
<tr>
<td>$B_z^S$</td>
<td>96.7 nT</td>
</tr>
<tr>
<td>$B_z^{SR}$</td>
<td>97.2 nT</td>
</tr>
<tr>
<td>$B_z^R$</td>
<td>97.3 nT</td>
</tr>
<tr>
<td>$B_z^S$</td>
<td>99.8 nT</td>
</tr>
<tr>
<td>$b_0^s$</td>
<td>1377 rad/s</td>
</tr>
<tr>
<td>$b_0$</td>
<td>549 rad/s</td>
</tr>
<tr>
<td>$b_0^r$</td>
<td>511 rad/s</td>
</tr>
</tbody>
</table>

*The value is for a uniform 2D cloud.*
two ensembles and the transverse microwave field component pointing in opposite directions.

If a cloud offset from the cavity axis is included in the coherent case, the cancellation of the Ramsey-pulling shift is not complete, as only some of the atom ensembles have a diametrically opposite partner for both Ramsey interactions. The Ramsey-pulling shift in case D can be above the $10^{-15}$ level due to the presence of initial Zeeman coherences. It is shown in Fig. 5 (right scale) and is orders of magnitude stronger than the shift with a cloud offset but without coherences (case B).

The coherent enhancement is also present for the important case E, for which the Ramsey cavity. This leads to a frequency shift that has the same sign and amplitude for all ensembles with the same radial distance from the cavity axis. This prevents the cancellation even for a cloud with no horizontal offset from the cavity axis, and the shift is above the $10^{-15}$ level. The magnitude of the frequency shift depends only slightly on the cloud offset as demonstrated in Fig. 5 (right scale) by comparison of cases E and F, because of the different amplitude of the microwave field components. Due to the removal of ensembles from the edge regions, the shift in case F is reduced compared to case E, similar to cases A and B.

We note that the magnitude of the Ramsey-pulling shift decreases with the inverse square of the value $B_x$ of the static magnetic field without coherences [12] (Fig. 5; shaded area for positive shifts) and with the inverse of $B_x$ with coherences [15] (Fig. 5; shaded area for negative shifts).

A process that changes the coherences phase relations given by Eq. (2) can lead to frequency shifts even when the population in both $|3, \pm 1\rangle$ states is the same. We tested this prediction by calculating the shift for a 1% symmetric population in both $|3, \pm 1\rangle$ states, and an additional phase of $\pi$ added only to the phase of $\rho_{1,0}^3$ and subtracted from the phase of $\rho_{0,1}^3$, while the phases of $\rho_{1,0}^{-1}$ and $\rho_{0,1}^{-1}$ were left as given by Eq. (2). We performed calculations using the symmetries of cases C-F from Fig. 4. For the symmetries of case C and equal populations of $m_F = \pm 1$ states, the coherent enhancement of the shift is canceled for the same reasons as discussed for case C with population imbalance. Because the $|3, \pm 1\rangle$ state populations are the same, the noncoherent part of the shift is even below $10^{-17}$. In contrast, for the symmetries corresponding to cases D-F, the shift has an inverse sign and is approximately twice as large compared to cases D-F.

The significance of coherently enhanced Ramsey-pulling shifts with symmetric populations of the $|3, \pm 1\rangle$ states is that the shifts can be due to the state selection process with a microwave field on-resonance. In practice, the necessary deviation of the phase of the atomic coherences from that given by Eq. (2) could result from the ac Stark shift of the $|3, \pm 1\rangle$ states during the clearing process, when the light frequency detuning and polarization couple the two $|3, \pm 1\rangle$ states to the $6\Delta \pi^2 P_{3/2}$ excited-state components differently. Thus, a coherent enhancement of the Ramsey-pulling shift could be present under normal operation in fountains with resonant microwave state selection. Further investigations are required to determine the magnitude of such light-assisted Ramsey-pulling enhancement.

In summary, when initial Zeeman coherences between the $|3, 0\rangle$ clock state and a $|3, \pm 1\rangle$ state are present, the Ramsey-pulling shift can be above the $10^{-15}$ level when the symmetry of the coherences involving the $|3, -1\rangle$ and $|3, +1\rangle$ states is broken, either by unequal populations in these states or due to a difference in the phase of the corresponding coherences. Fortunately, effects such as Majorana transitions originating from the $|3, 0\rangle$ state will transfer the population to the $|3, -1\rangle$ and $|3, +1\rangle$ states symmetrically, with coherences that fulfill Eq. (2). A broken symmetry can still result from Majorana transitions if an asymmetric population of the $|3, -1\rangle$ and $|3, +1\rangle$ states exists beforehand or if the relative phase of the coherences is altered later by effects such as different ac Stark shifts of the $|3, -1\rangle$ and $|3, +1\rangle$ states with respect to the $|3, 0\rangle$ state. A frequency shift of the state selection microwave field from the clock transition frequency directly leads to a population and coherence asymmetry. Because the phase of the coherences created during a microwave state selection becomes position dependent, a significant shift can result even for a cloud centered on the cavity axis.

IV. MICROWAVE AMPLITUDE AND POPULATION DEPENDENCES OF RAMSEY PULLING

The microwave amplitude dependence is an important feature of Ramsey pulling. Experimentally, elevated microwave amplitudes are used to study distributed cavity phase frequency shifts and microwave leakage effects [24,25], and therefore it might be useful to put an upper limit on the possible Ramsey-pulling contribution to the observed microwave-dependent frequency shifts. As the microwave amplitude increases, the magnitude of the Ramsey-pulling shift also increases, as under normal operation the microwave pulse area for the $\Delta m_F = \pm 1$ transitions is far below $\pi$, and their transition probability increases with the microwave amplitude, while the contrast of the central Ramsey fringe diminishes. In contrast to the approach in Ref. [12], the numerical model allows the calculations to be extended to Rabi frequencies significantly exceeding the frequency splitting between the Zeeman components.

Here we study the microwave amplitude dependence of Ramsey pulling for the cases introduced in the previous section. The microwave amplitude dependence for the case of Ramsey-pulling shifts without initial Zeeman coherences (cases A and B in Fig. 4) as well as with initial Zeeman coherences (cases C to F in Fig. 4) are shown in Fig. 6. Since individual atom ensembles experience microwave amplitudes that vary by up to 15% with position, the Ramsey fringe contrast never goes to 0, and the shift does not show singularities for even-\pi/2 pulse areas. As shown in Fig. 6, the amplitude dependence reaches the $10^{-15}$ level even without initial coherences and, due to its highly nonlinear dependence, can significantly affect frequency evaluations with elevated microwave amplitude levels.

Without coherences, additional calculations show that the shift depends almost linearly on the initial population of one of the $|3, \pm 1\rangle$ states, with the other state not populated. The deviation from linearity is less than 10% for a population
of either of the $|3, \pm 1\rangle$ states reaching 10% of the entire atom ensemble population (with the population of the other state kept 0). The shift value changes by less than 10% when the initial population difference in the $|3, \pm 1\rangle$ states is kept at 1%, and the respective populations of these states are increased to 10% and 9% of the entire atom ensemble population. The shift depends quadratically on the ensemble offset from the cavity axis and is 0 for ensembles on the axis. For smaller atom cloud sizes, the atoms are closer to the cavity axis (where the transverse microwave field components have a lower amplitude) than in the case of a cloud uniformly distributed across the cavity aperture as done in the calculation (with the exception of small clouds with large offsets from the cavity axis). The uniform distribution of the atom cloud across the cavity aperture leads to an overestimation of the calculated Ramsey-pulling shifts by 10%–15% compared to clouds smaller than the cavity aperture and with small offsets from the cavity axis.

In the presence of initial coherences with a magnitude expressed through Eq. (2), the frequency shift with population imbalance deviates from linearity by less than 10% until the relative population of either of the $|3, \pm 1\rangle$ states reaches 10%. Its deviation from linearity is less than 5% when the population of either of the $|3, \pm 1\rangle$ states is kept at 10%, and only the coherence magnitude is changed from 0 to the maximum one given by Eq. (2). The frequency shift dependence on the $|3, 0\rangle \leftrightarrow |3, \pm 1\rangle$ coherence magnitudes given by Eq. (2) deviates from linearity by less than 10% when the population difference of the $|3, \pm 1\rangle$ states is kept at 1%, and the respective populations of these states are increased to 10% and 9% of the entire atom ensemble population. The frequency shift dependence on the ensemble offset from the cavity axis in the presence of coherences is nonlinear, as the coherence phase depends strongly on the amplitude of the transverse microwave components, which varies with the offset from the cavity axis.

V. EXPERIMENTAL OBSERVATION OF RAMSEY-PULLING FREQUENCY SHIFTS

Without Zeeman coherences between the initial atomic states, the calculated Ramsey-pulling shift is below the $10^{-16}$ level for normal fountain operation. The presence of Zeeman coherences makes the shift significant at the $10^{-15}$ level and makes experimental observation possible. To test the predictions of the calculation, we used the state selection process in the fountain PTB CSF1 to create coherent atomic superpositions, which allowed us to observe for the first time a Ramsey-pulling shift in a fountain clock. In CSF1, the state selection cavity is mounted below the Ramsey cavity in the region of the low magnetic C field. As a result, the propagation time between the state selection and the Ramsey cavity is only $T_{SR} = 24.2$ ms (see Fig. 2), and the phases of the individual atom coherences created by the state selection process do not average out. The clearing pulse is also applied during this travel time by a vertical, circularly polarized laser beam.

The state selection microwave field drives $\Delta m_F = 0$ and $\Delta m_F = \pm 1$ transitions simultaneously due to the presence of vertical and transverse field components. Such excitation prepares each atom in a coherent superposition which involves the $|3, 0\rangle$ clock state and the $|3, \pm 1\rangle$ magnetically sensitive states. Under normal operation, with the state selection microwave field on-resonance and its amplitude corresponding to a $\pi$ pulse area, the population of the $|3, \pm 1\rangle$ states is very small due to the presence of the static magnetic field $B_z^3$ in the state selection cavity, which detunes the transitions, and the low amplitude of the transverse microwave field components. To increase the magnitude of the frequency shift, on one hand, the population of the $|3, \pm 1\rangle$ states was increased by operating the state selection cavity at an increased microwave amplitude (corresponding to a $9\pi$ pulse area on-resonance). On the other hand, the Ramsey cavity was also operated at an increased microwave amplitude ($7\pi/2$ pulse area) to enhance the $\Delta m_F = \pm 1$ transition probabilities. The fountain frequency was measured against the frequency average of two hydrogen masers.

For zero frequency detuning in the state selection cavity, the Ramsey pulling is expected to be negligible because the $|3, \pm 1\rangle$ states will be symmetrically populated, and the phase relations of Eq. (2) are valid. To cause a significant frequency shift, we used a detuning of the state selection microwave field from the clock transition frequency in the range of $\pm 200$ Hz, which creates asymmetry in the populations of the $|3, \pm 1\rangle$ states and, correspondingly, in the coherent state superpositions $|3, 0\rangle \leftrightarrow |3, \pm 1\rangle$. The shift in the fountain frequency as a function of the state selection microwave frequency is shown in Fig. 7 (symbols). The small state selection field detuning preserved a high population of the $|3, 0\rangle$ clock state and allowed us to measure the fountain frequency without significant degradation of the frequency stability. For detunings of the order of $\pm 100$ Hz and for the chosen microwave amplitude, calculations show that after the state selection process, roughly 2% of the atomic population in the $F = 3$ component can be in either of the two state superpositions $|3, 0\rangle \leftrightarrow |3, \pm 1\rangle$ (depending on the detuning sign), while around 8% of the atomic population in the $F = 3$
component is due to the transfer from the $|4, \pm 1\rangle$ to the $|3, \pm 1\rangle$ states by the $m_F = \pm 1, \Delta m_F = 0$ transitions.

The frequency shift due to the detuned field in the state selection cavity was calculated using the model introduced in Sec. II. The atom cloud was approximated by a cloud with a Gaussian radius $\sigma = 4.0 \times 10^{-4}$ m and temperature $T_m = 1.6 \times 10^{-6}$ K, determined from measurements. The resonant microwave amplitudes in the simulation were adjusted numerically (for the maximum average transition probability) to correspond to $9\pi$ (state selection cavity) and $7\pi/2$ (Ramsey cavity) pulse areas. The magnetic fields were chosen to match the measured values as listed in Table I. An additional magnetic field offset $B_{z}^{\text{Stark}} = -2.6$ nT accounts for the ac Stark shift resulting from the clearing laser pulse, as discussed below. The calculation results are also shown in Fig. 7. The dashed curve corresponds to the measured magnetic field value $B_{z}^{SR}$ between the state selection and the Ramsey cavities as listed in Table I. The solid curve corresponds to $B_{z}^{SR}$ with the additional offset of $B_{z}^{\text{Stark}} = -2.6$ nT, and the shaded area to $B_{z}^{SR}$ with an offset range of $B_{z}^{\text{Stark}} \pm 0.5$ nT, representative of the magnetic field uncertainties.

Our model shows that the Ramsey-pulling shift is determined by both the magnitude and the phase of the Zeeman coherences with which the atoms arrive in the Ramsey cavity. First, these parameters depend on the static magnetic field $B_z^S$ and the microwave field parameters during the microwave interaction in the state selection cavity. Second, during the time the atoms spend between the two cavities, the magnitude of the Zeeman coherences does not change, but their phase evolves due to the effective static magnetic field $B_{z}^{SR}$ and due to the occurrence of the clearing laser pulse (through the ac Stark shift effect). The measured and calculated dependence of the Ramsey-pulling frequency shift on the value of the static magnetic field $B_{z}^{SR}$ is shown in Fig. 8. The detuning of the state selection microwave frequency was fixed to $+110$ Hz [(red) circles] or $-110$ Hz [(blue) squares]. The value of the field $B_{z}^{SR}$ was changed by varying the current in a compensation coil positioned at the bottom of the innermost magnetic shield of the fountain with its axis in the vertical direction. The variation of the coil current leads to different values for the field $B_{z}^{SR}$, the fields $B_{z}^{S}$ in the state selection and $B_{z}^{R}$ in the Ramsey cavities, and the effective field $B_{z}$ above the Ramsey cavity. These magnetic field values were measured using the atoms as a probe for several values of the compensation coil current. The measured values of $B_{z}^{S}$, $B_{z}^{SR}$, $B_{z}^{R}$, and $B_{z}$ were used in the calculation. We note that, as a result, the depicted frequency shifts are caused by the combination of effects due to the actual values of $B_{z}^{S}$, $B_{z}^{SR}$, $B_{z}^{R}$, and $B_{z}$, and not just by the effect of the actual value of $B_{z}^{SR}$, as Fig. 8 might suggest. The dashed curves correspond to $+110$ Hz [curve (a)] and $-110$ Hz [curve (b)]. The clearing pulse ac Stark shift was again included through the $B_{z}^{SR}$ magnetic field offset of $B_{z}^{\text{Stark}} = -2.6$ nT and is shown by the solid lines $[+110\text{ Hz, curve (c)}]$ and $-[110\text{ Hz, curve (d)}]$. The shaded areas correspond to a $B_{z}^{SR}$ offset range of $B_{z}^{\text{Stark}} \pm 0.5$ nT.

A disagreement between measurements and simulation exists, as indicated by the use of two vertical scales. A reason could be the use of interpolation to map the magnetic field along the atom ensemble trajectories from a limited number of measurements at different compensation coil currents and atom cloud launch heights. Other possible reasons are discussed at the end of this section.

The Ramsey-pulling shift magnitude also shows a dependence on the duration of the clearing laser pulse exciting the $5S_{1/2}(F = 4) - 2F_{3/2}(F' = 5)$ optical transition, which is applied by a circularly polarized vertical laser beam in CSF1.
The laser frequency is detuned from all transitions involving the $F = 3$ ground-state component, by the cesium hyperfine component splitting of 9.2 GHz. The states of the $F = 3$ component experience an ac Stark shift due to the presence of the off-resonant laser light. The ac Stark shift is different for the $|3,0\rangle$ and the $|3,\pm1\rangle$ states, which leads to a phase change of the Zeeman coherences. The phase change is proportional to the ac Stark shift frequency differences for the $|3,0\rangle$ and the $|3,\pm1\rangle$ states, the laser intensity, and the clearing pulse duration.

In the fountain CSF1 we have changed the clearing pulse duration from 0.5 to 12 ms, and the corresponding change of the Ramsey-pulling shift for a fixed state selection microwave field detuning of $\pm120$ Hz is shown in Fig. 9 [(red) circles and (blue) squares]. From the sinusoidal fits the absolute value of the Ramsey-pulling shift after the propagation time $T = 24.2$ ms. Due to its clear signature, the frequency shift dependence on the light pulse duration can be used as a simple test to reveal the presence of Zeeman coherences in the initial atomic state before the Ramsey interactions.

In summary, we find an antisymmetric dependence on the state selection microwave field detuning, with no shift predicted for a state selection field resonant with the clock transition (Fig. 7). In the presence of a state selection detuning, we observe a clear dependence of the resulting shift on the static magnetic field between the microwave cavities (Fig. 8) and on the duration of the clearing laser pulse duration (Fig. 9), both of which affect the phases of the Zeeman coherences.

The agreement between measurements and the calculations shown in Figs. 7 and 8 is not perfect. It is important to mention that the calculations do not include free parameters. One expects a partial disagreement between measurements and calculations because of the uncertainty in the determination of the different static magnetic field values and the assumption of vertical magnetic field direction. Simulations show that an angle of only a few degrees between the field direction and the cavity axis changes the value of the Ramsey-pulling shift appreciably. A magnetic field tilt of several degrees is observed in CSF1 as in the fountain CSF2 [26]. Discrepancy is also expected due to the difference in the actual profile of the state selection microwave field and the TE$_{201}$ mode used by the simulation. The simulation shows that the phase of the initial coherences depends strongly on the state selection microwave pulse area and, thus, on the accuracy of the pulse area determination. The clearing laser pulse spatial profile is not homogeneous, which leads to a dephasing of the atomic coherences. Finally, the calculations assumed that the cloud had a Gaussian shape, with a center of mass on the axis for both the state selection and the Ramsey cavities. These assumptions are only approximately correct for CSF1.

VI. CONTRIBUTION OF THE RAMSEY-PULLING SHIFT TO THE UNCERTAINTY BUDGET OF CSF1 AND CSF2

For CSF1, the low static magnetic field in the state selection cavity leads to coherent initial state superpositions even in the normal mode of operation. To prevent shifts, it is important to keep the excitation of the $|3,\pm1\rangle$ states low and symmetric. This can be accomplished by ensuring a zero detuning of the microwave field in the state selection cavity. As the rapid adiabatic passage method for collisional shift measurements [27] is not applicable to the CSF1 fountain due to the low static magnetic field in the state selection cavity, the method of detuned state selection [28] has been considered for operation with low atomic densities to reduce systematic collisional shift measurement problem can be diagnosed and corrected for by alternating shots with negative and positive shift due to Zeeman coherences is not expected to play a role. On the one hand, the employed rapid adiabatic passage method increases the probability of magnetically sensitive $\pi$ and $\sigma$ transitions due to the high microwave amplitude and pulse spectral width, and in the case of the interrupted pulse used in low-atom-number operation, excites the $|3,\pm1\rangle$ states asymmetrically [30]. On the other hand, however, the state selection cavity of the fountain is outside the magnetic shields, with a Zeeman component splitting of more than 50 kHz. For such detunings, the $\pi$ and $\sigma$ transitions are driven with a very low probability even at high microwave amplitudes. More importantly, the free propagation of the atoms between the state selection and the Ramsey cavity occurs in a static magnetic field above 10 $\mu$T that is strongly
inhomogeneous near the magnetic shield apertures, which enables fast averaging of the phase of the Zeeman coherences. The difference in the individual atom ensembles’ propagation times between the cavities is considerably larger compared to that of CSF1, leading to a significant phase decoherence. No Ramsey-pulling shifts were observed in CSF2 at the $10^{-15}$ level in experiments with operational conditions similar (with detuning and increased microwave amplitudes of the state selection microwave field) to those used with CSF1 to generate coherent Ramsey-pulling shifts.

Assuming that shift enhancement due to initial coherences does not play a role in either fountain, we calculated the Ramsey-pulling shift without coherences for both CSF1 and CSF2 fountains for normal operation. For both fountains, the current value of the static magnetic field $B_z$ inside the magnetic shielding is approximately 150 nT, giving a maximum Ramsey-pulling shift of $5 \times 10^{-18}$ for 1% population imbalance, as shown in Fig. 5 (cases A and B). In both CSF1 and CSF2, the measured population imbalance is 0.25%, resulting in a maximum Ramsey-pulling shift of $1.25 \times 10^{-18}$. This value is used from now on for the uncertainty budget contribution of the combined Rabi- and Ramsey-pulling frequency shift in CSF1 and CSF2. This value naturally includes the Rabi-pulling shifts previously investigated independently.

VII. CONCLUSIONS

Ramsey-pulling frequency shifts were calculated extending the theory of Ramsey pulling presented by Cutler et al. [12] to the case of atomic fountains by taking into account coherent initial atomic states, a narrow atomic velocity distribution, and a varying amplitude and direction of the magnetic microwave field components during Ramsey interactions. The calculations provide an improved estimate of the maximum Rabi- and Ramsey-pulling shift.

It is shown that initial asymmetric coherences between magnetic sublevels of the same hyperfine ground-state component can lead to a dramatic enhancement of the Ramsey-pulling shift, by more than two orders of magnitude, and to a considerable shift under normal fountain clock operation. Initial asymmetric coherences can be due to detuning of the state selection microwave field, Majorana transitions, or can be created during or after the state selection process by atom interaction with the microwave or optical fields. While coherences do not lead to frequency shifts as long as their phases fulfill the symmetry requirements discussed in Sec. III, this symmetry can easily be broken by effects such as the ac Stark shift resulting from the clearing pulse.

Our approach also enabled an investigation of Ramsey pulling at elevated microwave amplitudes in the Ramsey cavity. In this case the magnitude of the Ramsey-pulling shift increases and can reach the $10^{-15}$ level even without initial Zeeman coherences, which can affect the results of experimental investigations at increased amplitudes, such as those used to characterize distributed cavity phase effects.

The properly applied state selection process in fountain clocks excludes frequency shifts caused by asymmetric coherences from Majorana transitions at the early stages of the fountain cycle (before the clock transition takes place). However, potential Majorana transitions and resulting Zeeman coherences generated at the end of the clock cycle (before the atomic state detection) may enhance the Ramsey frequency pulling when the detection is inhomogeneous with regard to the $\pm f$ states [20].

The theoretical results are supported by experiments with the PTB fountain CSF1, by changing the frequency of the state selection microwave field at increased microwave amplitude in both the state selection and the Ramsey cavities. The Ramsey-pulling shift is shown to depend on the state selection microwave field detuning, on the magnetic field between the state selection and the Ramsey cavities, and on the presence of light interacting with the cesium atoms at the end of the state selection process.

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APPENDIX A: DERIVATION AND FORM OF THE TOTAL HAMILTONIAN

The hyperfine interaction couples the nuclear spin $I = 7/2$ and the electronic angular momentum $J = 1/2$ of the cesium ground state. In the absence of external fields, the atomic Hamiltonian $H_0$ takes the form $H_0 = A(I \cdot J)$, with $A$ the hyperfine structure constant of the $6s^2S_{1/2}$ cesium ground state and $I$ and $J$ the nuclear spin and the electronic angular momentum operators. The eigenstates of the atomic system can be written in the basis of the total angular momentum $F = I + J$, with values for the cesium ground state $F = 3$ or 4 and projection quantum number $m_F = -F, \ldots, F$.

In the presence of an external static magnetic field $\vec{B} = (0,0,B_z)$ pointing along the $z$ direction, $H_0$ is given by

$$H_0 = A(I \cdot J) + \mu_B B_z(I_z + g_I J_z),$$

(A1)

where $\mu_B$ is Bohr’s magneton, $g_I = -0.398 853 9(5) \times 10^{-3}$, and $g_J = 2.002 540 3(2)$ [31,32] are the nuclear and the electronic $g$ factors and $I_z$ and $J_z$ are the $z$ components of the nuclear spin and the electronic angular momentum operators.

The matrix elements $\langle F’, m_F | H_0 | F, m_F \rangle$ of $H_0$ are calculated by transforming the $|F, m_F\rangle$ states into the product $|I, m_I\rangle \otimes |J, m_J\rangle$ basis, acting on the resulting states with the operators $I$ and $J$ in $H_0$, and transforming the result back into the $|F, m_F\rangle$ basis [33]. The resulting eigenvalues (or energies) of the matrix $\langle F’, m_F | H_0 | F, m_F \rangle$ are described by the Breit-Rabi formula,

$$E_{F,m_F} = -\frac{2\pi\hbar v_0}{2(2I + 1)} \pm \frac{\pi\hbar v_0}{2} \sqrt{\left(1 + \frac{4m_F\mu_B B_z}{2I + 1} + x^2 B_z^2\right)},$$

(A2)

where $\hbar$ is Planck’s constant divided by $2\pi$, $v_0 = 9192 631 770$ Hz is the unperturbed hyperfine splitting frequency of the components $F = 3$ and $F = 4$, the (+) sign corresponds to the $F = 4$ and the (−) sign to the $F = 3$ state component, and $x_B = \mu_B B_z (g_I - g_J)/(2\pi\hbar v_0)$. In the low-magnetic-field approximation, the eigenstates of $\langle F’, m_F | H_0 | F, m_F \rangle$ can be replaced by the states $|F, m_F\rangle$. In
this approximation, the matrix \( \langle F', m_F | H_0 | F, m_F \rangle \) is diagonal, with matrix elements given by Eq. (A2).

In the presence of a microwave field with frequency \( \nu_{RF} = \nu_0 + \Delta \nu \), with frequency detuning \( \Delta \nu \) from \( \nu_0 \) and an amplitude of the magnetic field vector component \( B_{RF} \), the total atomic Hamiltonian becomes \( H = H_0 + H_{RF} \), where the microwave interaction Hamiltonian \( H_{RF} \) is given by

\[
H_{RF} = \sum_{k=x,y,z} \mu_B B_{RF} (g_I I_k + g_J J_k) (e^{i2\pi(\nu_0+\Delta \nu) t} + c.c.)/2.
\]  

(A3)

For the cesium ground state, the matrix elements of the nuclear spin operator \( I \) have the same magnitude (except for the corresponding \( g \) factors) and the opposite sign to the matrix elements due to the electronic angular momentum operator \( J \). Thus, we can replace the product \( g_I I_k + g_J J_k \) in Eq. (A3).

The corresponding matrix elements of \( H_{RF} \) are calculated in the same way as those of \( H_0 \). It is now convenient to use a unitary transformation which allows us to set to 0 the matrix elements of \( H \) oscillating at the hyperfine frequency \( \nu_0 \). The matrix elements \( \langle F', m_F | U | F, m_F \rangle \) of the unitary transformation \( U \) are

\[
U_{F'Fm_F'm_F} = \begin{cases} 
\delta_{F'F}\delta_{m_F'm_F} e^{-i(\nu_0+\Delta \nu)t} & \text{(for } F = 4), \\
\delta_{F'F}\delta_{m_F'm_F} e^{i(\nu_0+\Delta \nu)t} & \text{(for } F = 3), 
\end{cases}
\]

(A5)

where \( \delta \) is the Kronecker \( \delta \). The transformation rule for the total Hamiltonian \( H' = H_0 + H_{RF} \) is

\[
H' = \mathcal{U} (H_0 + H_{RF}) \mathcal{U}^{\dagger} - i \hbar \mathcal{U} \frac{d\rho}{dt},
\]

(A6)

where \( \mathcal{U} \) is the Hermitian adjoint operator of \( U \). The transformation rule for the density matrix is \( \rho' = U \rho U \).

The unitary transformation \( U \) causes terms to appear in \( H' \) which oscillate at frequencies close to \( 2 \nu_0 \), which we will neglect (rotating-wave approximation). Also, the matrix elements corresponding to transitions between Zeeman sublevels of the same ground-state component \( F \) have a time dependence oscillating at frequencies close to \( \nu_0 \). These terms are also neglected in the rotating-wave approximation, as they correspond to transitions well detuned for weak magnetic fields \( B \).

The atomic state is represented by a density matrix \( \rho \) constructed from the six relevant states in the following order: \((4, +1), (4, 0), (4, -1), (3, +1), (3, 0), (3, -1)\). The total Hamiltonian \( H' \) of the six-level atomic system, after application of the unitary transformation \( U(t) \) and the rotating-wave approximation, is

\[
H' = \hbar \begin{pmatrix}
\delta_1 & 0 & 0 & -\frac{\sqrt{15}}{16} B_z & \frac{\sqrt{15}}{16} (b_0 - i b_1) & 0 \\
0 & \delta_2 & 0 & -\frac{\sqrt{15}}{16} B_z & \frac{\sqrt{15}}{16} (b_0 + i b_1) & -\frac{\sqrt{15}}{16} b_z \\
0 & 0 & \delta_3 & 0 & -\frac{\sqrt{15}}{16} (b_0 + i b_1) & \frac{\sqrt{15}}{16} (b_0 - i b_1) \\
-\frac{\sqrt{15}}{16} B_z & -\frac{\sqrt{15}}{16} (b_0 - i b_1) & 0 & \delta_4 & 0 & 0 \\
\frac{\sqrt{15}}{16} (b_0 + i b_1) & -\frac{\sqrt{15}}{16} B_z & \frac{\sqrt{15}}{16} (b_0 - i b_1) & 0 & \delta_5 & 0 \\
0 & 0 & -\frac{\sqrt{15}}{16} B_z & 0 & 0 & \delta_6
\end{pmatrix},
\]

(A7)

where the Rabi frequencies \( b_k (k = x, y, z) \) depend on the atom ensemble position in the microwave cavities. The parameters \( \delta_i \) are given below:

\[
\begin{align*}
\delta_1 &= -\pi (\nu_0 + \Delta \nu - \nu_0 \sqrt{1 + x_B^2/2 + x_B^2}) + B_x g_I \mu_B / \hbar, \\
\delta_2 &= -\pi (\nu_0 + \Delta \nu - \nu_0 \sqrt{1 + x_B^2/2 + 1/2}) + B_y g_I \mu_B / \hbar, \\
\delta_3 &= -\pi (\nu_0 + \Delta \nu - \nu_0 \sqrt{1 - x_B^2/2 + 1/2}) - B_z g_I \mu_B / \hbar, \\
\delta_4 &= +\pi (\nu_0 + \Delta \nu - \nu_0 \sqrt{1 + x_B^2/2 + 1/2}) - B_x g_I \mu_B / \hbar, \\
\delta_5 &= +\pi (\nu_0 + \Delta \nu - \nu_0 \sqrt{1 - x_B^2/2 + 1/2}) + B_y g_I \mu_B / \hbar, \\
\delta_6 &= +\pi (\nu_0 + \Delta \nu - \nu_0 \sqrt{1 - x_B^2/2 + x_B^2}) - B_z g_I \mu_B / \hbar.
\end{align*}
\]

APPENDIX B: MICROWAVE FIELD DISTRIBUTION IN THE FOUNTAIN CAVITIES

The PTB fountains CSF1 and CSF2 have different rectangular state selection cavities (supporting the \( TE_{201} \) and \( TE_{401} \) modes, respectively) and identical cylindrical Ramsey cavities (supporting the \( TE_{011} \) mode). The solution of the Maxwell equations for a real cavity with apertures and resistive losses can be obtained using finite-element analysis methods [34]. In this work, we use the TEM modes of an ideal rectangular (state selection) or cylindrical (Ramsey) cavity with no apertures as an approximation of the spatial microwave field distributions.

The expressions for the magnetic components of the microwave field of a \( TE_{m01} \) mode of the rectangular state
TABLE II. Cavity parameters for PTB fountains CSF1 and CSF2 (in millimeters).

<table>
<thead>
<tr>
<th>Parameter</th>
<th>CSF1</th>
<th>CSF2</th>
</tr>
</thead>
<tbody>
<tr>
<td>$d_z^c$ (state selection)</td>
<td>48.00</td>
<td>84.46</td>
</tr>
<tr>
<td>$d_x^c$ (state selection)</td>
<td>12.00</td>
<td>38.00</td>
</tr>
<tr>
<td>$d_y^c$ (state selection)</td>
<td>22.00</td>
<td>25.67</td>
</tr>
<tr>
<td>Height (above loading zone)</td>
<td>346</td>
<td>186</td>
</tr>
<tr>
<td>$d$ (Ramsey)</td>
<td>28.62</td>
<td>28.62</td>
</tr>
<tr>
<td>$r$ (Ramsey)</td>
<td>24.20</td>
<td>24.20</td>
</tr>
<tr>
<td>$r_c$ (aperture)</td>
<td>5.00</td>
<td>5.00</td>
</tr>
<tr>
<td>Height (above loading zone)</td>
<td>442</td>
<td>635</td>
</tr>
</tbody>
</table>

Selection cavities in Cartesian coordinates are [35]

$$ B_{x,0}^\text{RF} = \frac{B_0^\text{RF}}{2 \pi} \left( \frac{m}{d_x^c} \right) \sin \left( \frac{\pi m}{d_x^c} \right) \sin \left( \frac{\pi}{d_x^c} \right), $$

$$ B_{y,0}^\text{RF} = 0, $$

$$ B_{z,0}^\text{RF} = \frac{B_0^\text{RF}}{2 \pi} \cos \left( \frac{\pi m}{d_x^c} \right) \cos \left( \frac{\pi}{d_x^c} \right), $$

where the field components are normalized such that the $z$ component is equal to $B_0^\text{RF}$ in the center of the cavity. The horizontal dimension of the cavity along the $x$ axis is $d_x^c$, and the cavity height is $d_z^c$. The mode number is $m = 2$ for CSF1 and $m = 4$ for CSF2. The distances along $x$ and $z$ are measured from the cavity center.

The expressions for the magnetic components of the microwave field of a TE$_{011}$ mode of the Ramsey cavity in cylindrical coordinates are [35]

$$ B_{\rho,0}^\text{RF} = \frac{B_0^\text{RF}}{d_0} J_0 \left( \frac{x_0}{r} \right) \sin \left( \frac{\pi m}{d_x^c} \right), $$

$$ B_{\varphi,0}^\text{RF} = 0, $$

$$ B_{\varphi,0}^\text{RF} = \frac{B_0^\text{RF}}{d_0} J_0 \left( \frac{x_0}{r} \right) \cos \left( \frac{\pi}{d_x^c} \right), $$

where, again, the vertical microwave field component is equal to $B_{z,0}^\text{RF}$ in the center of the cavity, $x_0$ is the first zero of the Bessel function $J_1$, $\rho$ is the radial distance from the cavity axis, $r$ is the radius of the cavity, $z$ is the vertical distance measured from the center of the cavity, and $d$ is the cavity height. The cavity parameters for the fountains CSF1 and CSF2 are listed in Table II.

The value of the microwave field component $B_{\rho,0}^\text{RF}$ can be more conveniently expressed in terms of an effective Rabi frequency, $b_0 = \mu B_0^\text{RF} (g_j - g_i)/2h$, taking a value $b_0^c$ for the state selection and $b_0$ for the Ramsey microwave interaction. To solve the von Neumann equation (1), we convert the spatial dependence of the microwave amplitude into a time dependence assuming a constant velocity along the $z$ direction for a given atom crossing the cavity on the way up or down.

APPENDIX C: FRINGE CONTRAST IN THE 2D MODEL

For a given atom ensemble $i$, the $m_{F} = \pm 1$, $\Delta m_{F} = 0$, and $\Delta m_{F} = \pm 1$ transition probabilities as a function of the microwave frequency have a Ramsey fringe structure with a period $T_{Ri}$. The Ramsey fringe structures of these transitions overlap and extend all the way to the clock transition frequency. When the static magnetic field is changed, the fringe structure of these transitions and the corresponding frequency shift of the clock transition change accordingly, leading to a field-dependent Rabi-pulling shift with a period proportional to $1/T_{Ri}$ and a Ramsey-pulling shift with a period proportional to $2/T_{Ri}$ (see Ref. [12]). The factor-of-2 difference in the field dependence is due to the twice-smaller field dependence of the $\Delta m_{F} = \pm 1$ transitions compared to the $m_{F} = \pm 1$, $\Delta m_{F} = 0$ transitions.

In practice, the fringe contrast of the transitions involving magnetically sensitive states decreases in the same way as that of the clock transition with their detuning from resonance due to the averaging over atom ensembles with different vertical trajectories and corresponding Ramsey times $T_{Ri}$. This decrease can be seen in Fig. 3 (bottom; red curve). The diminished fringe contrast of the $\Delta m_{F} = \pm 1$ and $m_{F} = \pm 1$, $\Delta m_{F} = 0$ transitions near the clock transition frequency $v_0$ generally leads to Rabi- and Ramsey-pulling shifts of reduced magnitude and actual dependences on the static magnetic field strength with a period proportional to $1/T_{Ri}$ and $2/T_{Ri}$, i.e., to the microwave interaction time in the Ramsey cavity corresponding to the cloud center [14]. Because, for the 2D atom cloud used in the calculation, the atom ensembles have identical vertical trajectories, the fringe contrast is not diminished with detuning from resonance and adjustments of the model are needed.

To cancel the $T_{Ri}$ dependence of Ramsey pulling, we used in the simulation two same-state atom ensembles at each horizontal grid point at the beginning of the first Ramsey interaction. The first ensemble had the same horizontal position during both Ramsey interactions, while the second ensemble had a diametrically opposite horizontal position during the second Ramsey interaction. For each such pair, the Ramsey fringe structure of these transitions has an opposite phase, as, due to the cylindrical cavity symmetry, the microwave components driving the $\Delta m_{F} = \pm 1$ transitions in each atom ensemble point in opposite directions during the second Ramsey interaction. At the same time, the fringe structure of the $\Delta m_{F} = 0$ transitions for the two ensembles remains the same. As a result, the Ramsey-pulling shift dependence on the Ramsey time $T_{Ri}$ is canceled out when the shift is averaged over the cloud containing such pairs of atom ensembles.

To cancel also the dependence of the Rabi-pulling shift on $T_{Ri}$, we calculated the average of two shift values corresponding to two magnetic fields in and above the Ramsey cavity: one for a field $B_{s} = B_{0}^\text{RF}$ and one for a slightly higher field, $B_{s} = B_{0}^\text{RF}$, which increases the $|3, +1 \rangle \to |4, +1 \rangle$ transition frequency by $1/2T_{Ri}$. For these two magnetic field values, the fringe structures of the $m_{F} = \pm 1$, $\Delta m_{F} = 0$ transitions have an opposite phase around $v_0$, and the Rabi-pulling shift dependence on $T_{Ri}$ is canceled. Additional calculations performed using Monte Carlo simulated 3D clouds with temperatures of the order of 1 $\mu$K showed that, after the cancellation, the results of the calculation using distribution of atom ensembles on a 2D grid resemble closely these using a 3D atom clouds filling uniformly the cavity apertures on the way up and down.