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Spin waves and Collisional Frequency Shifts of a Trapped-Atom Clock

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We excite spin-waves with spatially inhomogeneous pulses and study the resulting frequency shifts of a chip-scale atomic clock of trapped ⁸⁷Rb. The density-dependent frequency shifts of the hyperfine transition simulate the s-wave collisional frequency shifts of fermions, including those of optical lattice clocks. As the spin polarizations oscillate in the trap, the frequency shift reverses and it depends on the area of the second Ramsey pulse, exhibiting a predicted beyond mean-field frequency shift. Numerical and analytic models illustrate these observed behaviors.

Quantum scattering in an ultracold gas of indistinguishable spin-1/2 atoms leads to rich and unexpected behaviors, even above the onset of quantum degeneracy. Among these, spin waves are a beautiful macroscopic manifestation of identical spin rotation (ISR) [1–4]. ISR also inhibits dephasing, which can dramatically increase the coherence time in a trapped ensemble of interacting atoms to tens of seconds [5, 6], with possible applications to compact atomic clocks and quantum memories. Another example are collisional interactions in optical lattice clocks [7–10]. Their detailed understanding is a prerequisite for optical lattice clocks to realize their full potential as future primary standards.

At ultracold temperatures, the scattering is purely swave, which is forbidden for indistinguishable fermions, suggesting that clocks using ultracold fermions are immune to collision shifts [11, 12]. However, spatial inhomogeneities of the clock field, which are naturally larger for optical frequency fields than for radio-frequencies, allow fermions to become distinguishable and therefore can lead to s-wave clock shifts [8–10]. A series of experiments attributed the collision shifts of Sr lattice clocks to these novel s-wave fermion collisions with inhomogeneous clock field excitations [7, 13, 14]. However, subsequent work showed that p-waves dominate for Yb lattice clocks, and p-wave scattering is consistent with all the observed Sr collisional frequency shifts [15].

Bosons with state-independent scattering lengths have fermion-like exchange interactions [8, 16, 17]. This allows us to simulate the s-wave fermion collisional shift with a chip-scale clock that traps ⁸⁷Rb, a boson with nearly equal scattering lengths. We observe the distinguishing feature that the collisional shift in the presence of inhomogenous excitations depends on the area of the second Ramsey clock pulse. This dependence sets it apart from the well-known s-wave shift for homogeneous excitations, which is absent for fermions and, for bosons, depends only on the first pulse area, and hence the population difference of the two clocks states [8, 18]. Further, inhomogeneous excitations directly excite spin waves. We show an inextricable link between spin-waves and the s-wave fermion collisional shifts. Notably, we observe frequency

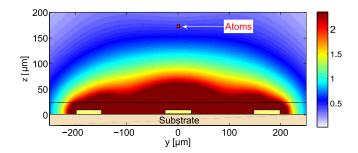


FIG. 1. Calculated microwave field amplitude of the coplanar waveguide on our atom chip, in arbitrary units. The rapidly decaying near-field causes a small vertical gradient of the spin-polarization across the trapped atomic cloud.

shifts that change sign as spin polarizations oscillate in the trap.

We perform Ramsey spectroscopy with two spatially inhomogeneous pulses to study spin waves and the collisional frequency shifts of trapped ⁸⁷Rb atoms. The first clock pulse creates an inhomogeneous spin polarization, which varies linearly in space. We directly observe a spatiotemporal oscillation of this spin polarization, which characterizes the strength of the atomic interactions. Driving a second Ramsey pulse, we measure frequency shifts of this clock. Here we vary the areas of each pulse and the interrogation time between the two pulses, to probe the unique behaviors of s-wave fermion clock shifts. We develop analytic and numerical models that describe the observed spin-waves and the novel dependence on the area of the second Ramsey pulse.

Our chip-scale atomic clock magnetically traps between 10^3 and 10^5 atoms at a distance $z_0=156~\mu\mathrm{m}$ below a microwave coplanar waveguide on our atom chip [5, 19]. A microwave and radio-frequency, two-photon excitation drives the clock transition, $|\downarrow\rangle \equiv |F=1, m_F=-1\rangle$ to $|\uparrow\rangle \equiv |F=2, m_F=1\rangle$. The near field of the microwave guide (Fig. 1) creates a slightly inhomogeneous Rabi frequency in the vertical z direction, $\Omega(\mathbf{r}) = \Omega_0(1 + \delta_1 z + \delta_2 z^2 + \ldots)$. The trap frequencies are

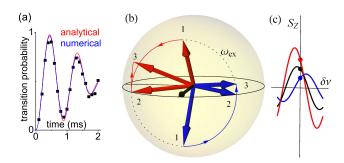


FIG. 2. (a) Inhomogeneous Rabi flopping with $\Omega(\mathbf{r}) =$ $\Omega_0(1+\delta_1z)$. The data are fitted to models of two trapped atoms: (red) an analytic model with a single motional sideband gives $\delta_1 = 0.147(4) \xi_z^{-1}$. (blue) a numerical simulation for representative vibrational states $\eta_z = 30 \pm 8$ yields $\delta_1 = 0.090(4) \xi_z^{-1}$. (b) Bloch sphere evolution of the spins of two representative atoms (blue and red), which are initially on opposite sides of the trap in state $|\downarrow\rangle$. The first inhomogeneous Ramsey excitation pulse rotates the spins differently $(\theta_{red,1} = 4\pi/5 \text{ and } \theta_{blue,1} = \pi/5)$. During the Ramsey interrogation time, ISR rotates the two spins around their sum (black arrow), here, by $\omega_{ex}T_R = \pi/2$. We take a second pulse with the same inhomogeneity as the first, but weaker, $\overline{\theta_2} = \pi/8$. It barely moves the blue spin, whereas the red rotates to be more vertical. We detect the vertical projection of each spin S_z , corresponding to the points in (c). In (c) we trace S_z as a function of detuning, showing the resonance shifted to a negative detuning. For $\omega_z T_R = (2j+1)\pi$, both atoms have switched sides of the trap so that the inhomogeneity of the 2nd pulse instead gives the blue spin a larger rotation, hence higher Ramsey fringe contrast. Thus the frequency shift changes sign as the spins oscillate in the trap.

 $(\omega_x, \omega_y, \omega_z) = 2\pi(32(1), 97(1), 119.5(5))$ Hz. The temperature of the cloud is 175(6) nK, at least 30 nK above the onset of Bose Einstein condensation, with no measurable dependence on the atom number. At this temperature, the identical spin rotation rate ω_{ex} , of order $\omega_{MF} \equiv 4\pi\hbar |a_{\uparrow\downarrow}| \bar{n}/m$, dominates in our experiments. The lateral collision rate $\gamma_c \propto a_{\uparrow\downarrow}^2 \bar{n} v_T$ is always much lower than the trap frequencies, corresponding to the Knudsen regime. Here $a_{\uparrow\downarrow}$ is the inter-state scattering length, \bar{n} the mean density, m the atomic mass and v_T the thermal velocity. The magnetic field at the trap center is tuned to minimize the inhomogeneous spread of transition frequency [20] so that dephasing can be neglected on the timescales we consider [21].

The variation of the Rabi frequency across the atomic cloud is determined by fitting the resonant Rabi flopping using $\Omega_0 \gg \omega_z$ so that atomic motion during the pulse can be neglected (Fig. 2(a)). We find $\delta_1 \approx 0.1 \, \xi_z^{-1}$ and $\delta_2 \approx 0$ which is reasonable since the r.m.s. cloud radius $\xi_z = 4.1 \mu \text{m} \ll z_0$. [22].

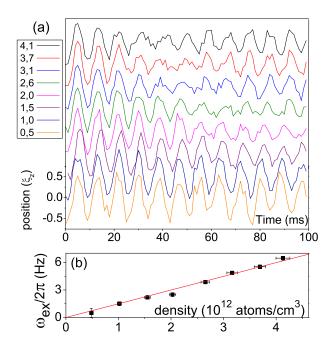


FIG. 3. (a) Center of mass position of the $|\uparrow\rangle$ component of the cold atom cloud versus time after a single $5\pi/2$ pulse for various atomic densities (in 10^{12} atoms/cm³). Successive curves are successively offset vertically by $0.5\xi_z$. As the density increases, a beat appears between the trap frequency and the increasingly faster identical spin rotation rate ω_{ex} , characteristic for spin waves. (b) Fitting ω_{ex} versus density gives density dependence $2\pi 1.4 \,\mathrm{Hz}/(10^{12} \,\mathrm{atoms/cm}^3) \times n$.

We initiate a spin-wave with a single $\tau = 1.05$ ms excitation pulse of area $\Omega_0 \tau = 2.5\pi$. We use a multiple of $\pi/2$ to produce a larger spin inhomogeneity. This inhomogeneous spin population then oscillates in the trap and we observe the oscillation by holding the atoms in the trap for various times t_h after the Rabi pulse, followed by a 7 ms of time-of-flight and state-selective absorption imaging. Fig 3(a) shows the center of mass of the $|\uparrow\rangle$ component. The data for our lowest atomic density exhibits a simple oscillation at ω_z , and the $|\downarrow\rangle$ cloud (not shown) oscillates out of phase. The center of mass of the total population shows no measurable oscillation. Increasing the density, we observe a collapse and revival of the oscillation at shorter and shorter times. At $t_h = 80 \,\mathrm{ms}$, the oscillation for the highest density is out of phase with the lowest. As we show below, this is a signature of a spin wave driven by ISR.

We can intuitively illustrate the spin dynamics by considering two localized atoms oscillating in a onedimensional trap with frequency ω_z (Fig. 2(b)). Putting the atoms initially on opposite sides of the trap center in state $|\downarrow\rangle$, they are excited with a short Rabi pulse of mean area $\theta_1 = \Omega_0 \tau = \pi/2$, typically. Since $\Omega = \Omega(z)$, the atoms experience different Bloch vector rotations $\theta_1 \pm \Delta \theta_1$. After the pulse, the atoms oscillate in the trap during the Ramsey interrogation time T_R as in Fig. 3(a). In the absence of interactions, each atom maintains its spin orientation, in a frame that rotates at the atomic transition frequency, and thus the spatial spin populations simply oscillate at ω_z . Note that the phase of each atom's coherence is constant. However, if there are exchange interactions, the two spins will rotate with an ISR rate ω_{ex} around their total spin as they repeatedly collide (Fig. 2(b)) [5, 16]. In a time π/ω_{ex} , the two atoms exchange their spin polarizations, producing a beat between ω_{ex} and ω_z . This introduces the frequency ω_{ex} into the spatial oscillation of the spins, producing a beat between ω_{ex} and ω_z . For each curve, we extract ω_{ex} , which varies linearly with density (Fig. 3(b)), as $2\pi 1.4 \,\mathrm{Hz}/(10^{12} \mathrm{at/cm^3}) \times n$, within our uncertainty. [23]

The spatiotemporal spin oscillation has important consequences for Ramsey spectroscopy. The second Ramsey pulse reads-out the phase of the atomic coherences. Between the pulses, the exchange interaction modulates the phase of each atom's coherence as the spins rotate about one another (Fig. 2(b)). If we were to measure the transition probability of one of the atoms above [8], the apparent resonance frequency would depend on T_R - it would be modulated at ω_{ex} . In the usual case when both atoms are detected, the frequency excursion of this modulated collision shift is reduced (Fig. 2(c)). The shift averages to zero if the second Ramsey pulse is homogeneous. For an inhomogeneous second pulse, the two Bloch vectors experience different rotations $\theta_2 \pm \Delta \theta_2 = \Omega(z_i(T_R))\tau$ depending on their positions at the time of the pulse. This gives them different weights in the Ramsey measurement, making the cancellation incomplete, unless the second pulse is an odd multiple of $\pi/2$, which reads out the phases of both atoms with the same sensitivity [8]. This simple model predicts a clock shift

$$\delta\nu = \frac{\Delta\theta_1\Delta\theta_2\sin\left(\omega_{ex}T_R\right)\cos\left(\omega_zT_R\right)\cos\theta_2}{4\pi T_R\sin\theta_1\sin\theta_2}.$$
 (1)

It extends the results in [8] to $\omega_{ex}T_R \geq 1$ and unresolved sidebands. Here we linearize the dependence on $\Delta\theta_i$.

A singlet-triplet basis provides helpful insight and also leads to Eq. (1). Before the first pulse the two atoms are in the triplet state $|S,m_s\rangle=|1,-1\rangle$. The inhomogeneous excitation pulse makes them partially distinguishable and populates the singlet state $|0,0\rangle$ [24], which then accrues a collisional phase shift during the Ramsey interrogation time [25]. When $\Omega_0 \approx \omega_z, \omega_{ex}$, we numerically calculate the evolution of the S=1 pseudo-spin system, coherently including all transitions up to the 5th sideband. We also treat the 5% scattering length difference $a_{\uparrow\uparrow} \gtrsim a_{\uparrow\downarrow} \gtrsim a_{\downarrow\downarrow}$.

To experimentally test eq. (1), we measure the shift of the clock's frequency with a Ramsey sequence for the same range of densities as in Fig. 3. Fig. 4(a) shows the measured shift as a function of density for two $\tau_{1,2}=1.05\,\mathrm{ms}$ pulses separated by a $T_R=100\,\mathrm{ms}$ interrogation time, which is close to a multiple of the trap period. The first pulse area is $\theta_1=5\pi/2$ and the second is $\theta_2=2.2\pi$. Like above, a large pulse area is used to increase the inhomogeneity. The observed frequency shift indeed oscillates as a function of density, giving a frequency shift that is inconsistent with the often-used mean-field shift [7, 9, 12, 18]. Moreover, the first zero of $\delta\nu$ indeed occurs for $\omega_{ex}T_R=\pi$, with the value of ω_{ex} being determined from the data in Fig. 3 for this density. This confirms a distinguishing prediction of eq. (1).

We also measure the shift as a function of T_R (Fig. 4(b)). Again we use $\theta_1 = 2.5\pi$ and $\theta_2 = 2.2\pi$ and determine the frequency shift $\delta\nu$ for each of the atomic densities, $\overline{n} \approx \{0.4, 0.8, 1.3, 1.7\} \, 10^{12} \, \text{at/cm}^3$. From a linear fit we extract the slope $\alpha = d\delta\nu/dN_{at}$. This suppresses potential density-independent frequency shifts that vary with T_R . Whenever the spatial spin distribution is the same for the first and second Ramsey pulses, the shift has the same sign and has the opposite sign when the spatial spin distribution reverses. This emphasizes the importance of the correlation between the inhomogeneities of the first and second Ramsey pulses. In the low-density regime, ($\overline{n} \lesssim 10^{12} \text{at/cm}^3$), Eq. 1 predicts that α in Fig. 4(b) should oscillate at ω_z . Here, the mean frequency shift is offset, as expected from the small scattering length differences.

A distinguishing feature of the s-wave fermion collisions (eq. 1) is that the frequency shift spectroscopy depends on the area of the second Ramsey pulse [8]. For homogeneous excitations of ultracold bosons, the frequency shift depends on the area of the first Ramsey pulse, which determines the population difference of the clock states for the collisions during the Ramsey interrogation time. This unique feature was not demonstrated in the observations of collisional shifts of lattice clocks [7, 9, 10, 13–15]. We apply $\theta_1 = 2.5\pi$, T = 92 ms and vary the second pulse area θ_2 via the microwave power, keeping the duration fixed. The resulting α is shown in Fig. 4(c) (black squares). The $1/\tan\theta_2$ dependence predicted in [8] and (1) is clearly visible.

To show the quantitative agreement between the data of Fig. 4 and our models we use the experimental parameters, including δ_1 which is independently determined from the resonant Rabi flopping with the respective model (Fig. 2(a)). The numerical model reproduces the data [26]. The analytical model eq. 1 reproduces the oscillations but overestimates the amplitude of the shift by 60%. Including many sidebands, the numerical model gives better agreement as expected.

We also varied the area of the first Ramsey pulse θ_1 , keeping $\theta_2 = 5\pi/2$ fixed. Surprisingly, the shift is comparable to when θ_2 is varied. Eq. (1) has a small dependence on θ_1 when θ_2 is not exactly $5\pi/2$ but the predicted shift is much smaller than observed and would not

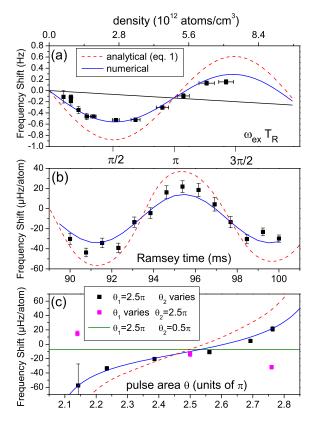


FIG. 4. (a) Measured frequency shifts for a Ramsey sequence $T_R = 100 \text{ ms}, \ \theta_1 = 2.5\pi \text{ and } \theta_2 = 2.2\pi \text{ versus interaction}$ strength, which is proportional to density. The shift is nonlinear; in fact it oscillates as the density increases. For reference we plot the known shift for homogeneous excitations, $\delta\nu = (a_{\uparrow\uparrow} - a_{\downarrow\downarrow})/(2\pi a_{\uparrow\downarrow})\omega_{ex}$ (black line). (b) Density shift per atom $d\delta\nu/dN_{at}$ in the low interaction regime versus T_R with $\theta_1 = 2.5\pi$ and $\theta_2 = 2.2\pi$. The shift oscillates as the spin polarizations oscillate in the trap. (c)Dependence on the first and second pulse areas. Black dots: $d\delta\nu/dN_{at}$ for $T_R = 92 \text{ ms}$ with $\theta_1 = 2.5\pi$ fixed and θ_2 variable. The green horizontal line is a reference measurement with $\theta_2 = \pi/2$ giving $-6 \,\mu\text{Hz/atom}$, the expected frequency shift in the absence of ISR and predicted from the difference of scattering lengths. A numerical calculation based on the singlet-triplet model for two atoms (blue line) reproduces the data with no free parameters. Eq. 1 qualitatively reproduces the observed behaviours, but overestimates the shift by 60% (red line). Magenta dots: θ_1 variable and $\theta_2 = 2.5\pi$ fixed. This dependence is not reproduced by either of the models.

change sign near $\theta_1 = 2.5\pi$. Similarly, shifts due to the small difference of scattering lengths $a_{\uparrow\uparrow} + a_{\downarrow\downarrow} - 2a_{\uparrow\downarrow}$ are too small. However, we note that our first pulse motionally excites the spin components, beyond a simple dipolar excitation. This leads to an oscillation of the cloud size, which unexpectedly varies with θ_1 . We speculate that this may produce some dephasing [27], and thereby leads to a dependence on θ_1 .

The striking connection between spin-waves and swave fermion collision shifts demonstrated here is very general. We can elucidate this connection by considering the resolved sideband regime, used in many ultraprecise atomic clocks, including optical lattice clocks. With resolved sidebands and weak interactions, even though the clock field cannot change the motional state of the atoms, we show that spin wave are excited. Here, the spatial inhomogeneity may give a low energy atom in vibrational state $|\alpha\rangle$ a large pulse area and a high energy atom in $|\beta\rangle$ a small pulse area, directly populating pair-wise singlet states $|0,0\rangle$ [8]. After the pulse, the two fermionic atoms evolve as $|\Psi(T)\rangle = se^{i\omega_{ex}T}|0,0\rangle\{|\alpha\beta\rangle\}^+ + (t|1,0\rangle +$ $u|1,1\rangle + d|1,-1\rangle \{|\alpha\beta\rangle\}^-$, where $|1,m_S\rangle$ are triplet states, $\{\}^{(-)+}$ denotes (anti)symmetrization, and u, d, t, and s are the state amplitudes. Rewriting this two particle wavefunction as $|\Psi(T)\rangle = |\downarrow\beta\rangle$ $^{-} + 1/\sqrt{2}(t$ $se^{i\omega_{ex}T}$) $\{|\downarrow\alpha\rangle|\uparrow\beta\rangle\}^- + (u|1,1\rangle + d|1,-1\rangle)\{|\alpha\beta\rangle\}^-$, we see that the $|\uparrow\rangle$ populations in state $|\alpha\rangle$ and $|\beta\rangle$ have an explicit oscillation at ω_{ex} – at different times T, the $|\uparrow\rangle$ population in the vibrational states are different. Thus, whenever there is a fermion collision shift, spin waves must also exist, and the fermion collision shift will oscillate as the spin populations oscillate in the trap.

In summary, we observe characteristic behaviors of the collisional frequency shifts due to inhomogeneous excitations in an atomic clock. The inhomogeneous excitations create spin-waves, which we show are inextricably connected to the s-wave frequency shifts of fermion clocks, including optical-frequency lattice clocks. We directly excite dipolar spin waves via an amplitude gradient of the excitation field. The spin populations oscillate, exhibiting a beat between the trap frequency and the frequency of spin rotation due to particle interactions. This leads to a collisional frequency shift that oscillates as the spin populations oscillate in the trap. We observe that the clock collision shift does not vary linearly with the atomic density and, in the spin-wave regime, varying the Ramsey interrogation time T_R (Fig. 4(b)) could help to evaluate the accuracy of atomic clocks. The frequency shift exhibits the novel dependence on the area of the second Ramsey pulse, in stark contrast to the mean field expressions for frequency shifts with homogeneous excitations [8]. While we intentionally exaggerate the spin wave excitations here, these frequency shifts can be minimized by using spatially homogenous fields, using sideband resolved pulses, and avoiding the Knudsen regime so that trap-state changing collisions further suppress the fermion shift.

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- [21] The inhomogeneous spread of transition frequencies due to the magnetic potential and the spatially varying atomic density, is less than 80 mHz.
- [22] We checked experimentally that inhomogeneity is predominantly vertical. Along y there is no detectable variation of $\Omega(\mathbf{r})$ and its variation along x is 5 times smaller than along z.
- [23] This coefficient is five times smaller than ω_{MF} [1]. Small amplitude dipolar spin waves oscillate at $\omega_{MF}/2$ [28] and models show that this frequency can decrease by a factor 2 for large amplitude spin waves. Additionally, we experimentally observe ω_{ex} decreases by a factor of two as $\theta_1 = 2.2\pi$ increases to $\theta_2 = 2.8\pi$.
- [24] Only the singlet state for fermions can have s-wave collisions. For bosons with equal inter and intra-state scattering lengths, the triplet states all have the same energy shift and the singlet state has none. Therefore, taking out the common shift of the triplet states, such bosons have the same interactions as fermions, albeit with the opposite ω_{ex} [8, 16, 17].
- [25] Eq. (1) can also be extracted in this basis. To derive it in the singlet-triplet basis, we consider short pulses, $\Omega_0 \gg \omega_z, \omega_{ex}$, neglecting interactions during the pulses, and small inhomogeneities, $\delta\theta \ll 1$, exciting only a single trap sideband. We add the contributions for the upper and lower sidebands.
- [26] To extract α from the numerical model, we calculate the shift for the 4 densities and extract a from a linear fit, in the same way that we analyze the data.
- [27] Such oscillations could create additional spatial inhomogeneities of the transition frequency through the trap, and add a new source of dephasing. Any dephasing directly couples pair-wise singlet and triplet states during the Ramsey time and generally lead to a dependence of the measured frequency on θ_1 . Since the contrast does not decrease significantly, the dephasing rate would have to be less than ω_{ex} .
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