Realization of a Rydberg-dressed atomic clock

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(Dated: October 10, 2018)

We present the experimental realization and characterization of an atomic clock based on optically trapped ultracold potassium atoms, where one state is continuously coupled by an off-resonant laser field to a highly-excited Rydberg state. We show that the observed Ramsey interference signals can be used to precisely measure the Rydberg atom-light coupling strength as well as the population and coherence decay rates of the Rydberg-dressed states with sub-kilohertz accuracy and for Rydberg state fractions as small as one part in $10^6$. This provides the means to combine the outstanding coherence properties of atomic clocks with controllable coupling to strongly interacting states, thus expanding the number of systems suitable for metrological applications and many-body physics studies.

Atomic clocks based on trapped ensembles of neutral atoms or single ions have enabled the most precise measurements ever made. Besides their importance for defining future time and frequency standards \cite{1,2}, they also hold great promise for searches for physics beyond the standard model \cite{3–5}, exploring the physics of complex quantum systems \cite{6}, and for realizing sensors capable of operating at the fundamental quantum limit \cite{7,8}. By design however, atomic clocks typically involve the coherent evolution of atoms that interact very weakly, either with one another or with external fields, seemingly precluding many possible applications.

Here we demonstrate an alkali atomic clock involving two magnetically insensitive hyperfine ground states, where one state is continuously coupled to a Rydberg state by an off-resonant laser field. This Rydberg-dressing approach provides the means to combine the outstanding coherence properties of atomic clocks with greatly enhanced sensitivity to external fields or controllable interparticle interactions mediated by the Rydberg state admixture. We show that the regime of strong Rydberg atom-light coupling can be reached with coherence times that are orders magnitude longer than the bare Rydberg state lifetime. The atomic clock is then applied to precisely measure the Rydberg-atom light coupling strength and to independently determine the effective population decay and dephasing rates for the dressed-states, thereby identifying the dominant decoherence processes. From these results we assess the suitability of Rydberg-dressed atomic clocks for metrological applications and for introducing and characterizing long-range interactions in many-body quantum systems.

The starting point for our experiments is an ultracold gas of $^{39}$K atoms held in a far detuned optical dipole trap (wavelength of 1064 nm), as shown in Fig. 1a. To prepare the sample, atoms are loaded into a three-dimensional magneto-optical trap (MOT) from a cold atomic beam within 1 s. The MOT is then compressed and overlapped with the optical dipole trap before applying a 10 ms sub-Doppler cooling phase using gray-molasses on the D1

Figure 1. Experimental realization of a Rydberg-dressed atomic clock. (a) A cloud of $^{39}$K atoms is prepared in an optical dipole trap and uniformly illuminated by the Rydberg dressing laser. Each atom can be represented by a four level system involving the clock states $|0\rangle$ and $|1\rangle$, the $|r=39P_{3/2}\rangle$ Rydberg state and an auxiliary shelving $|s\rangle$. The $|1\rangle \rightarrow |r\rangle$ transition is coupled via the dressing laser with Rabi frequency $\Omega$ and detuning $\Delta$. The Ramsey interferometry sequence consists of two $\pi/2$ rf pulses coupling $|0\rangle \rightarrow |1\rangle$, while the dressing laser is applied during the variable free evolution time $T$. (b) Ramsey fringes measured in the absence of the dressing laser showing very good coherence and (c) Ramsey fringes with the Rydberg excitation laser on with $\Delta/2\pi = -12.0$ MHz, showing a modified clock frequency and asymmetric decay of the interference contrast. The blue solid lines correspond to fits to the model described in the text.

arXiv:1810.04151v1 [physics.atom-ph] 9 Oct 2018
line [9]. This is followed by a short evaporative cooling phase in which the optical dipole trap intensity is ramped down to one tenth of its initial value in 400 ms.

To realize our atomic clock we use the magnetically insensitive hyperfine ground states $|0\rangle = |4S_{1/2}, F = 1, m_F = 0\rangle$ and $|1\rangle = |4S_{1/2}, F = 2, m_F = 0\rangle$. The sample is prepared in the initial $|0\rangle$ state by spin-polarizing the atoms using a sequence of optical pumping pulses and radio-frequency transfer pulses (similar to the procedure in Ref. [10]). The final result is a spin polarized sample of approximately $10^9$ atoms in the $|0\rangle$ state with a peak density of $1 \times 10^{10}$ cm$^{-3}$ and temperature of 10 µK. From there we apply a Ramsey interferometry sequence consisting of two $\pi/2$ radio-frequency (rf) pulses separated by a variable evolution time $T$ (Fig. 1a). The rf field originates from a programmable direct-digital-synthesis device which is referenced to a commercial rubidium standard. This is amplified and fed to an antenna inside the vacuum chamber to realize $\pi/2$ state rotations between the $|0\rangle$ and $|1\rangle$ states in 90 μs. To read out the clock we measure the population in the $|1\rangle$ state using absorption imaging with a weak probe laser resonant to the $|4S_{1/2}, F = 2\rangle \rightarrow |4P_{3/2}, F = 3\rangle$ transition.

Figure 1b shows the measured Ramsey fringe pattern taken without the dressing field for free evolution times up to 15 ms. During this time the signal is very well described by a sinusoidal oscillation with essentially no visible loss of contrast. By fitting the oscillation frequency we extract the Ramsey detuning of $\delta/2\pi = 914.0(6)$ MHz. This corresponds to an absolute frequency for the $|0\rangle \rightarrow |1\rangle$ transition of 461.79748(6) MHz (statistical uncertainty in the fractional frequency of $\Delta f/f = 1.3 \times 10^{-9}$ which is within 30 Hz of previous measurements of the $^{39}$K hyperfine splitting [10, 11]. The small discrepancy can be accounted for by the expected quadratic Zeeman shift of 31(11) Hz given that we apply a relatively large bias field of 60(10) mG and additional smaller corrections due to the differential AC Stark shift from the optical dipole trap and cold collisions.

Next we realize the Rydberg-dressed atomic clock by applying a laser field during the free evolution time which is detuned by $\Delta/2\pi = -12.0$ MHz from the $|1\rangle \rightarrow |r = 39P_{3/2}\rangle$ transition. In the dressed-state picture [12], this admixes a small Rydberg state component into the clock state (i.e. $|\bar{1}\rangle \approx |1\rangle + \sqrt{f_r}|r\rangle$, where $f_r$ is the relative population of the Rydberg state). The laser light is generated by frequency doubling a continuous-wave dye laser, with a second harmonic wavelength of 286 nm and a power of 65 mW. It is frequency stabilized using an ultra-low-expansion (ULE) material cavity and it is weakly focused such that the intensity is effectively uniform over the atom cloud. Fig. 1c shows the corresponding Ramsey signal. In comparison to the case of the bare atomic clock (fig. 1b), we observe an asymmetric decay of interference contrast and a visible shift of the clock frequency.

To interpret the effects of Rydberg dressing we derive a simple analytical formula for the Ramsey signal, including the influence of noise on the Rydberg dressing laser and neglecting interaction effects, which is justified given the low densities used for the experiment. We start by considering an ensemble of identical four-level atoms, comprised of the clock states $|0\rangle, |1\rangle$, the Rydberg state $|r\rangle$, and an auxiliary shelving state $|s\rangle$ which collectively describes all states into which the Rydberg states can decay and that no longer participate in the dynamics. The system evolution is well described by a quantum master equation for the density matrix $\rho$ in Lindblad form:

$$\dot{\rho}_{\text{NH}} = \frac{\Omega}{2} (|1\rangle\langle r| + |r\rangle\langle 1|) - \left(\Delta + \frac{i\Gamma}{2}\right) |r\rangle\langle r| - \delta |0\rangle\langle 0|,$$  \hspace{1cm} (1)

where $\Omega$, $\Delta$ are the amplitude and detuning of the Rydberg laser coupling and $\delta$ is the detuning of the rf field from the clock transition. Rydberg state decay appears as an additional imaginary detuning $i\Gamma/2$ which describes overall population loss from the Rydberg state out of the system with rate $\Gamma$.

In the weak dressing limit $|\Delta| \gg \Omega, \Gamma$, the $|r\rangle$ state can be adiabatically eliminated yielding an effective Hamiltonian for the slowly evolving clock states,

$$\hat{H}_{\text{eff}} = f_r \left(\Delta - \frac{i\Gamma}{2}\right) |1\rangle\langle 1| - \delta |0\rangle\langle 0|,$$ \hspace{1cm} (2)

where $f_r = \Omega^2/(\Gamma^2 + 4\Delta^2)$ can be recognized as the steady state Rydberg fraction of the dressed $|1\rangle$ state. Finally, intensity and frequency noise of the dressing laser can be included by treating the light shift as a fluctuating quantity $E_{\text{LS}} \rightarrow E_{\text{LS}} + \sqrt{X}\xi(t)$, where $\xi(t)$ is assumed to be a zero mean white noise process with $\langle \xi(t)\xi(t')\rangle = \delta(t-t')$. We define the noise variance,

$$X \approx \left(\epsilon_I^2 + \epsilon_\Delta^2\right) \frac{\Omega^4}{16\Delta^2},$$ \hspace{1cm} (3)

in terms of the spectral densities of the relative intensity and detuning noise $\epsilon_I^2$ and $\epsilon_\Delta^2$ respectively, with units (frequency)$^{-1}$.

Following [13] we write the stochastic time-dependent von Neumann equation for the density matrix $\rho_{\text{eff}}$ as,

$$\dot{\rho}_{\text{eff}} = -i[H_{\text{eff}}, \rho_{\text{eff}}] - i\sqrt{X}\xi(t)|1\rangle\langle 1|, \rho_{\text{eff}}].$$ \hspace{1cm} (4)

Performing the Markov approximation and using the identity for multiplicative linear white noise in [13], we arrive
At
\[ \dot{\rho}_{\text{eff}} = \left( \mathcal{L}_0 + \frac{1}{2} X \mathcal{L}_1 \right) \rho_{\text{eff}}(t), \]
(5)
where \( \mathcal{L}_0 \rho_{\text{eff}} = -i [H_{\text{eff}}, \rho_{\text{eff}}] \) and \( \mathcal{L}_1 \rho_{\text{eff}} = -i [1 \langle 1 |, \rho_{\text{eff}}] \).

Considering the Ramsey sequence in figure 1a and applying Eq. (5) during the free evolution time, the population in the \( |1 \rangle \) state after the second \( \pi/2 \) pulse is:
\[ P_1 = \frac{1}{4} \left( 1 + e^{-\delta X} + 2 e^{-\frac{1}{2} \left( \delta + f \Delta \right) t} \cos \left[ \left( \delta + f \Delta \right) t + \phi \right] \right), \]
(6)
where we have included the possibility of a small phase shift \( \phi \) caused by the detuning during the two \( \pi/2 \) pulses. Here the effects of Rydberg dressing can be identified as a light shift of the \( |1 \rangle \) state which changes the clock frequency by an amount \( E_{LS} = f \Delta \), an overall population decay rate \( \Gamma_{\text{eff}} = f \Gamma \) and pure dephasing rate \( X \).

In the following we analyze the performance of the Rydberg-dressed atomic clock for different detunings of the dressing laser \( \Delta \). For this, we fit the Ramsey measurements to Eq. (6) including an experimentally determined phaseshift \( \phi = 0.55 \). As exemplified by the solid lines in Fig. 1b,c the model reproduces the data extremely well. For further analysis we extract from the fits the detuning dependent parameters \( E_{LS}, \Gamma_{\text{eff}}, \) and \( X \).

Light shifts:– Figure 2a shows the Rydberg induced light shift \( E_{LS} \) as read out from the clock frequency for different detunings of the Rydberg dressing laser in the range \( \pm 40 \text{ MHz} \). The density plot in the background of the figure corresponds to the Fourier transform of each respective Ramsey signal which gives additional information about the spectral width of the Ramsey signal. To account for the cases close to resonance where the clock frequency \( E_{LS} + \delta \) becomes negative we map the sign of the data points such that the frequency is monotonic as a function of \( \Delta \) (these data points before mapping are displayed in gray in Fig. 2a).

Although the measured light shifts are relatively small, \( \lesssim 1 \text{ kHz} \) over most of the range, they are clearly resolved with an average statistical uncertainty of \( \sim 2 \times 10^{-3} \) for \( |\Delta|/2\pi > 10 \text{ MHz} \). Within this accuracy we observe small deviations from the simple two-level weak-dressing prediction of \( E_{LS} = \Delta \Omega^2 / (\Gamma^2 + 4 \Delta^2) \). Therefore, we perform a fit that include the additional light shifts caused by the nearby transition \( |0 \rangle \rightarrow |39P_{1/2} \rangle \) which is resonant for \( \Delta/2\pi = 76 \text{ MHz} \). The quality of the fit including the extra state, shown as a solid line in Fig. 2a, reflects a very good quantitative understanding of the dominant effects influencing the frequency of the Rydberg-dressed atomic clock. It also confirms that interaction effects such as the Rydberg blockade do not play an important role for the current experimental parameters.

From the fit shown in figure 2a we obtain a precise determination of the Rabi frequency \( \Omega/2\pi = 163(1) \text{ kHz} \). The value of \( \Omega \) is more than a factor of two smaller than an independent estimate based on the power and waist of the laser beam and the expected transition dipole matrix element. This highlights the importance of direct experimental measurement of the atom-light interaction parameters over indirect estimates. The range of resolved light shifts corresponds to Rydberg fractions from approximately \( 4 \times 10^{-4} \) to a minimum value of \( 4 \times 10^{-6} \),
The decaying contrast of the Ramsey fringes gives further information on the Rydberg-dressed states, specifically their degree of coherence which is influenced by fluctuations of the dressed state energy and population decay. While energy fluctuations result in a symmetric loss of contrast with respect to the mean (dephasing), population loss causes an overall reduction of the mean value. As such, these contributions can be separately extracted from the fits to Eq. (6) and are shown in Fig. 2b on a logarithmic scale. Close to resonance we find that the contributions are approximately equal with a rate around 1 kHz. This is comparable to the bare Rydberg state decay rate estimated as \( \Gamma / 2\pi = 5.6 \text{kHz} \) which includes photoionization from the optical dipole trap, blackbody redistribution and spontaneous decay. Interestingly, the maximum dephasing rate is two orders of magnitude smaller than the independently measured laser linewidth showing that the correspondence between laser phase noise and the loss of atomic clock coherence can be subtle.

For large detunings \(|\Delta|/2\pi > 5 \text{MHz}\) the effective loss and dephasing rates drop off rapidly, reaching values that are orders of magnitude lower than the bare Rydberg state decay rate. Here we find that dephasing is the dominant effect governing the loss of coherence rather than intrinsic atom loss. Nevertheless, the dephasing rate remains small compared to the light shift over most of the measurement range. This can be quantified by the strong-coupling parameter \( C = E_{LS}/(\Gamma_{eff} + X) \) that varies in the range \( 7 \leq C \leq 18 \) as \( \Delta \) is varied over the range \( \pm 40 \text{MHz} \). Looking closer at the detuning dependent dephasing rate we find that it is well described by a power-law which, below resonance, scales with \( \Delta^{-1.98(4)} \). This is close to the expected scaling for intensity noise, which according to Eq. (3) should scale as \( X \propto |\Delta|^{-2} \), as opposed to laser frequency noise which should scale with \( |\Delta|^{-4} \) since the relative frequency noise \( \epsilon_\Delta^2 \) scales as \( |\Delta|^{-2} \). From the power-law fit we extract the normalized intensity spectral density: \( 2\pi \epsilon_\Delta^2 = 0.2 \text{kHz}^{-1} \) which is compatible with residual intensity noise from the resonant cavity frequency doubler.

Having characterized the performance of the Rydberg-dressed atomic clock, we now assess its potential applications. As an example we consider the sensitivity of the Rydberg-dressed atomic clock to static electric fields due to the second order Stark effect of the Rydberg state, where the polarizability of the bare clock states can be totally neglected. Maximizing the derivative of Eq. 4 with respect to small perturbations and assuming shot noise limited state readout we find a minimum single shot sensitivity (standard quantum limit) \( \sigma = e(X + f_r\Gamma)/(f_r\alpha\sqrt{N}) \), where \( e \) is Euler’s number, \( F \) refers to the bias electric field strength, \( N \) is the number of atoms used in the measurement and \( \alpha \) is the electric polarizability of the Rydberg state (which scales with principal quantum number as \( n^4 \)). Importantly, the fundamental limit obtained for \( X \rightarrow 0 \) is independent of the Rydberg state fraction. Thus, the Rydberg dressing approach allows for extremely small densities of the strongly interacting particles, while at the same time enabling large overall atom numbers to minimize the atomic shot noise. For the Rydberg state used in our experiment (having \( \Gamma/2\pi = 5.6 \text{kHz} \) and \( \alpha_{SRP}/2\pi = 10.25 \text{MHz cm}^2/\text{V}^2 \) with \( N = 10^5 \) particles and applying a bias field \( F = 1 \text{V/cm} \), we find \( \sigma = 4.7 \mu\text{V/cm} \) which indicates that such clock based electrometers would rival the state-of-the-art electrometers [14–17].

While the present experiments were performed in an effectively non-interacting situation, Ramsey interferometry can serve as a powerful method to characterize atomic interactions [6, 18–21]. Rydberg dressing provides a way to induce and control these interactions, thereby introducing a tunable nonlinear evolution between the clock states. This would provide another route to generating squeezed or entangled many-body states without relying on low-energy collisions between the atoms [22–25], or the implementation of nonlinear quantum metrology protocols [26, 27] for surpassing the standard quantum limit. Alternatively, atomic clocks could provide a way to optimize Rydberg-dressing protocols [21, 28–30], even in the limit of extremely weak interactions, enabling the realization of robust quantum logic gates and quantum spin systems [31–34], or novel long-range interacting quantum fluids [35, 36] and lattice gases with beyond nearest neighbor interactions [37–39].

In conclusion we have realized and characterized the performance of a trapped atomic clock enhanced by Rydberg dressing. This includes a precise determination of the atom-light coupling, Rydberg state admixture and coherence properties of the dressed states. The basic essence of our scheme – the controllable coupling of a highly-coherent two-level system to a third state with greatly enhanced sensitivity – can be applied to numerous quantum systems, even beyond the Rydberg atom platform. Thus this approach has the potential to greatly expand the number of atomic and molecular systems suitable for metrological applications and many-body physics.

We acknowledge early contributions to the experiment by T. Wintermantel and H. Hirzler as well as valuable discussions with J. Schachenmayer. This work is part of and supported by the DFG Collaborative Research Centre “SFB 1225 (ISOQUANT)”, the Heidelberg Center for Quantum Dynamics and the “Investissements d’Avenir” programme through the Excellence Initiative of the University of Strasbourg (IdEx). S.W was partially supported by the University of Strasbourg Institute for Advanced Study (USIAS), A.A. and S.H. acknowledge support by the Heidelberg Graduate School for Fundamental Physics, S.H. acknowledges also support by the Carl-Zeiss foundation.
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