3-2008

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Todd A. Johnson
College of Saint Benedict/Saint John's University, toddjohnson@csbsju.edu

E. Urban

T. Henage

L. Isenhower

D. D. Yavuz

See next page for additional authors

Recommended Citation

Johnson TA, Urban E, Henage T, Isenhower L., Yavuz DD, Walker TG, Saffman M. 2008. Rabi oscillations between ground and Rydberg states with dipole-dipole atomic interactions. *Physical Review Letters* 100: 113003.

DOI: 10.1103/PhysRevLett.100.113003

Rabi Oscillations between Ground and Rydberg States with Dipole-Dipole Atomic Interactions

T. A. Johnson, E. Urban, T. Henage, L. Isenhower, D. D. Yavuz, T. G. Walker, and M. Saffman

Department of Physics, University of Wisconsin, 1150 University Avenue, Madison, Wisconsin 53706, USA

(Received 2 November 2007; published 19 March 2008)

We demonstrate Rabi oscillations of small numbers of ^{87}Rb atoms between ground and Rydberg states with $n \leq 43$. Coherent population oscillations are observed for single atoms, while the presence of two or more atoms decoheres the oscillations. We show that these observations are consistent with van der Waals interactions of Rydberg atoms.

DOI: 10.1103/PhysRevLett.100.113003 PACS numbers: 32.80.Rm, 03.67.−a

Atoms in highly excited Rydberg states with principal quantum number $n \gg 1$ have very large transition dipole moments which scale as $d \sim qa_0n^2$, with q the electron charge and a_0 the Bohr radius. Two such Rydberg atoms can be strongly coupled via a dipole-dipole interaction. It was recognized in recent years that the large interaction strength can potentially be used for fast quantum gates between qubits stored in stable ground states of neutral atoms [1,2]. When several atoms are sufficiently close together the presence of a single excited atom can cause a shift in the energy of all other atoms which is large enough to prevent resonant excitation of more than one atom in a sample [3]. This ''dipole blockade'' mechanism has the potential for creating strongly coupled ensembles containing moderate numbers of atoms. Such ensembles can be used for gates [3] as well as several other quantum-information tasks including single-atom state preparation, fast measurement protocols, and collective encoding of multiqubit registers [4].

A number of recent experiments have revealed signatures of the Rydberg interactions needed for dipole blockade by showing that the probability of multiple excitation is suppressed at high n or high atomic density [5]. However, none of the experiments to date have observed Rydberg interactions at the level of a single atomic excitation which is crucial for applications to quantum-information processing. In order to be useful for quantum gates it is also necessary to be able to coherently excite and deexcite a Rydberg state so that the atom is available for further processing. In this Letter we demonstrate important progress towards the goal of a fast neutral atom Rydberg gate by observing interaction effects between as few as two atoms and by observation of coherent Rabi oscillations between ground and Rydberg levels. We prepare single-atom states in micron-sized optical traps and observe coherent Rabi oscillations between ground and Rydberg states with $n \leq 43$ at rates as high as $\Omega_R/2\pi = 0.5$ MHz. We then show that the presence of two or more atoms in the trap causes dephasing of the Rabi oscillations. Comparison with theoretical calculations of the strength of the Rydberg van der Waals interactions [6], confirms that our observations are consistent with the presence of Rydberg interactions.

The experiment starts by loading a far-off-resonance optical trap (FORT) from a ^{87}Rb vapor cell magneto-optical trap (MOT) as described in our recent Letter [7]. For the experiments reported here, between 1 and 10 atoms are loaded into a 10 mK deep FORT (570 mW of 1030 nm light focused to a $1/e^2$ intensity radius waist of $w = 2.7$ μm). The radial and axial oscillation frequencies are 130 and 12 kHz. The average number of atoms is controlled by varying the amount of time for which the MOT and FORT lasers are simultaneously on from 25–400 ms. Atom temperatures in the FORT are measured by performing a drop and recapture measurement, and comparing the probability of recapture with numerical calculations. We consistently find temperatures from 5%–10% of the FORT depth, which corresponds to $T = 0.5 - 1$ mK for our typical parameters. These temperatures are much higher than Doppler cooling temperatures for ^{87}Rb, which we attribute to degradation of the laser cooling by large FORT induced differential Stark shifts of the $5s_{1/2}$ and $5p_{3/2}$ levels. Using an estimate of $T = 0.5$ mK, the spatial distribution of the atoms is quasi one-dimensional with standard deviations of $\sigma_z = 0.30$ μm and $\sigma_x = 3.5$ μm.

We prepare single-atom states in the FORT despite Poissonian loading statistics by using a two-measurement sequence. After the MOT to FORT loading period, we conduct a first measurement of the number of atoms in the FORT by scattering MOT light (3 pairs of counter-propagating beams) detuned by -5 MHz with respect to the cycling transition while chopping the FORT on and off at rates between 0.5×10^6 and 2.0×10^6 s^{-1}. The probing MOT light is chopped out of phase with the FORT, eliminating the need for tuning to the Stark shifted atomic resonance. Scattered photons are collected with a fast lens (with a numerical aperture of 0.4) and focused onto a cooled electron-multiplying CCD camera. We estimate our detection efficiency including finite solid angle, optical losses, and camera quantum efficiency to be about 2.7%. We observe single-atom photoelectron rates of about 10^4 s^{-1} (here time is the probing time with the MOT beams on, with the total measurement time about 2.5 times longer due to chopping) which gives the histogram shown on the abscissa of Fig. 1 for a 12 ms (probing time) first measurement. This is much shorter than the background gas limited

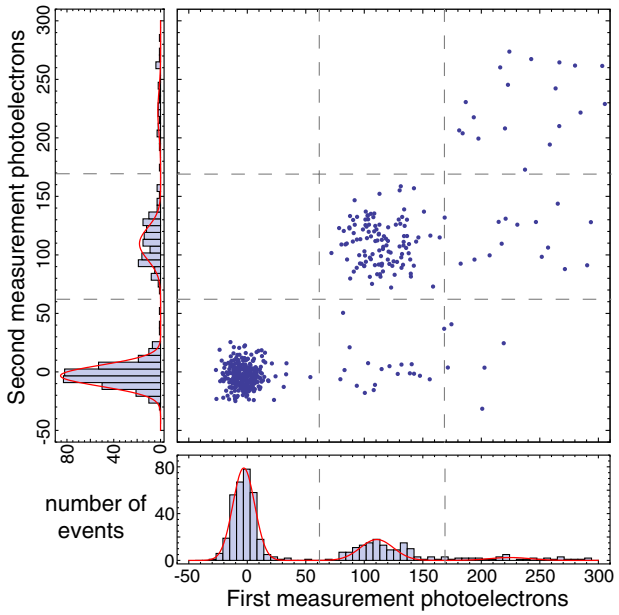

FIG. 1 (color online). Correlation between first and second measurement distributions, without Rydberg excitation between measurements. The dashed lines show cuts for selecting single-atom states and the red curves are fits based on a Poissonian model.

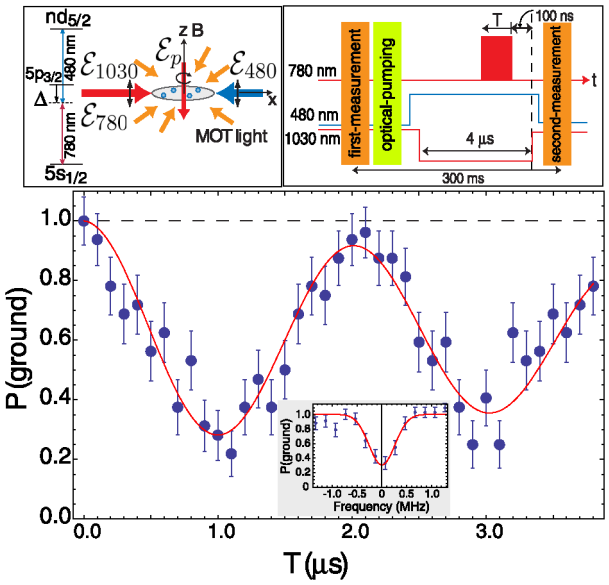

FIG. 2 (color online). Rabi oscillation experiment to $43d_{5/2}$ with $P_{780} = 1.85\ \mu$W, $P_{480} = 10.7$ mW, and $\Delta/2\pi = -3.4$ GHz. The experimental geometry is shown in the upper left and the timing sequence in the upper right. Each data point is the average of 40 preselected single-atom experiments, with the bars showing ± 1 standard deviation. The inset shows spectroscopy of the resonance obtained by scanning the frequency of the 780 nm light.

$1/e$ FORT lifetime of about 3 s. We verify the reliability of preselecting single-atom states by performing a second measurement, shown on the ordinate. We see that despite some loss of atoms during the first measurement, single-atom states can be prepared with about 85% probability, with a 15% admixture of zero atom states. Note that the reliability of selecting states with two or more atoms is much worse. We believe that this is due to light assisted collisions causing rapid loss out of the FORT during the first measurement [8].

We excite Rydberg states using two-photon transitions with 780 and 480 nm lasers as shown in Fig. 2. The Rydberg beams $\mathcal{E}_{780}$, $\mathcal{E}_{480}$ are focused to waists of $w \simeq 10\ \mu$m and spatially overlapped with the FORT. These beams are generated by locking a 780 nm laser and a 960 nm laser to different longitudinal modes of the same stable reference cavity with finesse $\mathcal{F} \sim 120\,000$ and linewidth $\sim$4 kHz. The cavity is constructed of ultralow expansion glass and is placed inside a temperature stabilized vacuum can. We obtain long term instability of a few hundred kHz, and short term instabilities of both lasers relative to the cavity line of a few hundred Hz at $<10\ \mu$s averaging time. The 960 nm light is then amplified and frequency doubled to create the 480 nm Rydberg excitation light. Acousto-optic modulator (AOM) based noise eaters are used as necessary to reduce amplitude fluctuations to a few percent. The frequencies of both lasers are then shifted with AOM's to match the desired Rydberg level.

After preparing a single-atom state the probability of transition to a Rydberg level is measured as a function of the pulse length of $\mathcal{E}_{780}$, $\mathcal{E}_{480}$. If the atoms are not prepared in a single ground state Zeeman level they will be coupled to a superposition of different Rydberg Zeeman levels. In the presence of background magnetic fields these levels have different Zeeman shifts which decohere the coherent population oscillations we are interested in. We therefore start by optically pumping into the $|f = 2, m_f = 2\rangle$ Zeeman state using σ_+ polarized light ($\mathcal{E}_p$ in Fig. 2) near resonant to the $|5s_{1/2} f = 2\rangle \leftrightarrow |5p_{3/2} f' = 3\rangle$ transition with a $B = 1.1 \times 10^{-3}$ T bias field. The $\hat{z}$-polarized excitation light only couples this state to the $|nd_{5/2}, m_j = 1/2\rangle$ Zeeman state. We have verified the expected shift of -6.2 MHz of the Rydberg excitation frequency in the presence of the bias field by performing spectroscopy with and without the field on.

In order to measure the probability of Rydberg excitation we use the fact that the calculated photoionization rate of the d states due to the FORT light is large compared to the radiative decay rate back to the ground state [9]. The timing sequence for Rabi oscillation is shown in Fig. 2. We turn off the FORT light for a fixed length of time which is long enough to perform the Rydberg excitation yet short enough that we do not lose the atom in the absence of a Rydberg pulse. We then perform a Rydberg pulse of variable length T, after which we restore the FORT light. Photoionization by the FORT light thus performs a projective measurement of the atomic state, and after 100 ms with the FORT on, the ground state population is measured

using MOT light. The resulting data points for the ground state probability are then normalized to 1.0 at $T = 0$ to correct for a 10%–20% loss rate due to the single-atom selection measurement, motional losses during the FORT drop period, and FORT loss due to background collisions. A curve fit to the data of Fig. 2 with the function $(1 - a) + ae^{-(t/\tau)}\cos(\Omega_R t)$ gives a Rabi frequency of $\Omega_R = 2\pi \times 0.49$ MHz, whereas our theoretical value with no adjustable parameters is [10] $\Omega_R = \Omega_{780}\Omega_{480}/2\Delta = 2\pi \times 0.55$ MHz. We attribute the approximately 11% lower experimental value to spatial misalignment, and a small fraction of the Rydberg light being present in servo sidebands from the laser locks. The fit also gives a decay time of $\tau = 8$ μs which is consistent with Doppler averaged numerical calculations. The data show that the atom is returned to the ground state with better than 90% probability and that Rydberg state excitation is achieved with 70%–80% probability. The lack of perfect Rydberg excitation is due to several factors which we estimate as Doppler broadening of the excitation (~10%–20%), imperfect optical pumping (~5%), and imperfect detection efficiency (~4%) [11].

A quantum gate protocol [1] requires that we leave an atom in one site in a Rydberg level while a conditional Rydberg excitation is performed at a neighboring site. With our observed π pulse time of ~1 μs this requires staying in the Rydberg level for at least 2 μs. We have verified that coherence can be maintained over this time by performing a double pulse experiment as shown in Fig. 3. For this experiment the maximum probability of Rydberg excitation is only about 50%. This is because during the 2 μs free evolution between the pulses the 780 nm Rydberg laser induced light shifts are no longer present which results in a free evolution for 2 μs at finite detuning. We have fit the measured data by assuming a detuning of 0.53 MHz. This is consistent with the 780 nm beam induced light shift of the ground state which is $\Omega_{780}^2/4\Delta = 2\pi \times 0.58$ MHz for our estimated experimental parameters.

The above Rabi oscillation curves change dramatically when more than one atom is present in the FORT. Figure 4 shows the results with average atom numbers of $\bar{N} = 0.3$, 1.7, and 8. These data were obtained by simply adjusting the loading time for a desired $\bar{N}$ (the actual distribution is close to Poissonian), and then applying varying length Rydberg pulses, without first using the single-atom selection procedure of Fig. 1. We see that at $\bar{N} = 1.7$ the visibility of the oscillations is strongly reduced, and at $\bar{N} = 8$ there is essentially no oscillation left. We emphasize that Rabi oscillation with high visibility and good coherence between ground state levels is observed with as many as 10 atoms in the FORT [7].

In order to explain these observations we must account for Rydberg interactions. ^{87}Rb atoms excited to the $43d_{5/2}$ state experience a Förster interaction due to the near resonance of the process $43d_{5/2} + 43d_{5/2} \rightarrow 45p_{3/2} + 41f_{5/2,7/2}$. The corresponding energy defect is $\hbar\delta = U(45p_{3/2}) + U(41f_{5/2,7/2}) - 2U(43d_{5/2})$. Using recent measurements of the Rb quantum defects [12], we find $\delta/2\pi = -6.0$, -8.3 MHz for the $f_{5/2}$ and $f_{7/2}$ states, respectively. This small energy defect naively implies that for an atom pair separated by R there is a strong resonant dipole-dipole interaction $U_{\rm dd} = C_3/R^3$, out to a crossover distance $R_c = (4C_3^2/\hbar^2\delta^2)^{1/6}$ followed by a van der Waals scaling $U_{\rm vdW} = C_6/R^6$ at larger distances with $C_3 \sim d^2$ and $C_6 \sim C_3^2/\hbar\delta$. For the $43d_{5/2}$ level the crossover distance is [6] $R_c = 8.1$ μm. However, due to Zeeman degeneracy of the Rydberg levels, there are linear superpositions of two-atom $d_{5/2}$ states which are excited by the Rydberg lasers, yet have small dipole-dipole interactions [13]. Our theoretical treatment of the van der Waals interaction in the presence of Zeeman degeneracy [6] shows that the effective interaction must be modified to

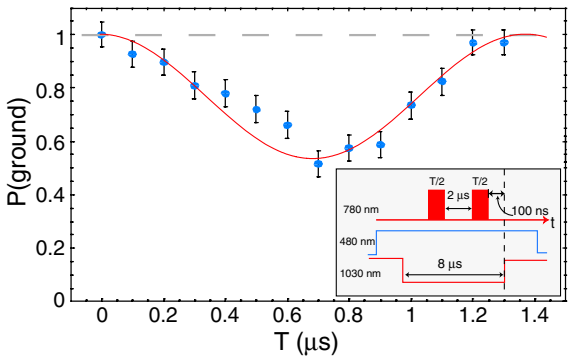

FIG. 3 (color online). Rabi oscillation experiment with single-atoms to $28d_{5/2}$ using a double pulse sequence. The excitation parameters were the same as in Fig. 2 except that $P_{780} = 3.3$ μW, $P_{480} = 9.0$ mW, and $\Delta/2\pi = -3.8$ GHz. The solid curve is a theoretical calculation assuming $\Omega_R/2\pi = 0.7$ MHz and a detuning in the 2 μs gap of 0.53 MHz. The inset shows the timing sequence.

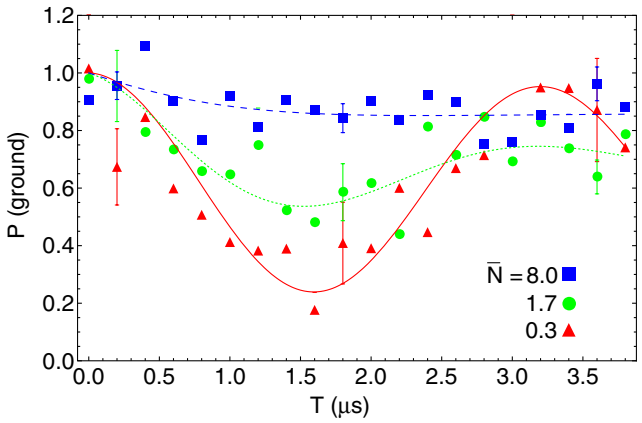

FIG. 4 (color online). Rabi oscillation experiment to $43d_{5/2}$ with different atom numbers. Each data point is the average of 150 measurements, with the bars showing ± 1 standard deviation.

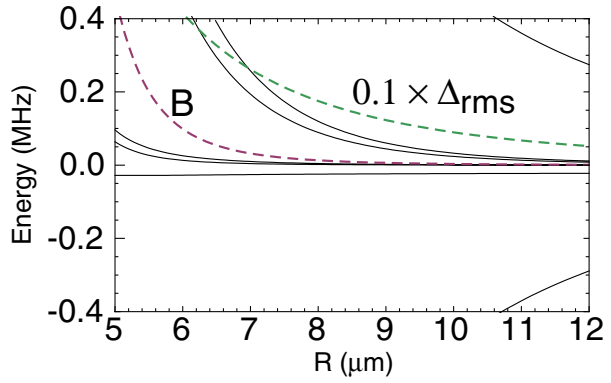

FIG. 5 (color online). Near resonant molecular energy levels for excitation of $43d_{5/2}|1/2, 1/2\rangle$ together with blockade shift B and direct interaction $\Delta_{\rm rms}$ as a function of separation R. The calculations used $C_3 = 1.8$ GHz μm^3, and $B = 0.0011$ T.

$U_{\rm vdW} = \sum_\varphi \kappa_\varphi^2 D_\varphi C_6/R^6$, where κ_φ accounts for the overlap of the laser excited state with the two-atom eigenstates $|\varphi\rangle$, and D_φ is an eigenvalue. In the quasi one-dimensional geometry provided by our optical trap the molecular axis $\hat{R}$ is close to $\hat{x}$ and is thus rotated by $\theta = \pi/2$ with respect to the quantization axis $\hat{z}$. In this situation we must account for coupling to eigenstates for which D_φ varies by a large factor of about 150 [6].

Accounting for all molecular eigenstates it can be shown that the probability of double excitation scales as [6] $P_2 \sim N\Omega_R^2/\mathsf{B}^2$, with $1/\mathsf{B}^2 \sim \frac{R^{12}}{C_6^2}\sum_\varphi \frac{\kappa_\varphi^2}{D_\varphi^2}$ and N the number of atoms. An effective dipole blockade is not achieved due to coupling to Förster zero states [13], which results in a small value of the effective blockade shift B. However, if there was no interaction we would expect to observe Rabi oscillations without decoherence with more than one atom. This is not the case since the direct interaction of two Rydberg excited atoms $\Delta_{\rm vdW} = U_{\rm vdW}/\hbar$ takes on a value larger than our experimental Rabi frequency causing dephasing of the oscillations. The situation is shown in Fig. 5 where we have plotted $\Delta_{\rm rms} \equiv \sqrt{\sum_\varphi \langle\varphi|\Delta_{\rm vdW}|\varphi\rangle^2}$, as a function of R. Curves are shown for $B = 0.0011$ T corresponding to our experiments. The theory results were obtained by numerical solutions of the dipole-dipole Hamiltonian accounting for the angular dependence of the dipole operator and all Zeeman shifts. We see that for $R > 5$ μm (which is the minimum value at which blockade is ineffective) and $R < 12$ μm (which is more than $3\sigma_x$ and therefore the largest separation that will occur) the direct interaction is $\Delta_{\rm rms}/2\pi \gtrsim 0.5$ MHz which is comparable to our Rabi frequency. Under these conditions we expect strong dephasing proportional to the number of atoms which is consistent with the data of Fig. 4. We thus have the surprising situation that the Rydberg interactions are both too weak to provide an effective blockade yet strong enough to dephase Rabi oscillations when more than one atom is present.

In conclusion we have observed coherent Rabi oscillations between ground and Rydberg states which is an important step towards demonstration of a neutral atom Rydberg gate. We have shown how van der Waals interactions lead to dephasing of the oscillations when several atoms are present, and elucidated the role of Förster zero states in the dephasing. Future work will explore alternative interaction geometries that do not couple to small eigenvalues as a means of demonstrating dipole blockade.

This work was supported by the NSF and ARO-IARPA. We are grateful to Pasad Kulatunga and Marie Delaney for experimental contributions at an early stage of this work.

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[10] For $\hat{z}$-polarized light $\Omega_{780} = \frac{\sqrt{2}}{3}\frac{q\mathcal{E}_{780}}{\hbar}R^{5_{P_{3/2}}}_{5_{S_{1/2}}}$ and $\Omega_{480} = \frac{\sqrt{6}}{5}\frac{q\mathcal{E}_{480}}{\hbar}R^{43_{D_{5/2}}}_{5_{P_{3/2}}}$. Using quantum defect wave functions we find $R^{5_{P_{3/2}}}_{5_{S_{1/2}}} = 5.14a_0$, $R^{43_{D_{5/2}}}_{5_{P_{3/2}}} = 0.029a_0$.
[11] The ratio of room-temperature radiative transitions to photoionization rates is about $(1.3 \times 10^4\ {\rm s}^{-1})/(3.1 \times 10^5\ {\rm s}^{-1})$ which implies a 4% reduction in our measured excitation probability relative to the true value.
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