Highly resolved measurements of Stark-tuned Förster resonances between Rydberg atoms

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We report on experiments exploring Stark-tuned Förster resonances between Rydberg atoms with unprecedented resolution in the Förster defect. The individual resonances are expected to exhibit different angular dependencies, opening the possibility to tune not only the interaction strength but also the angular dependence of the pair state potentials by an external electric field. We achieve a high resolution by optical Ramsey interferometry for Rydberg atoms combined with electric field pulses. The resonances are detected by a loss of visibility in the Ramsey fringes due to resonances in the interaction. We present measurements of the density dependence as well as of the coherence time at and close to Förster resonances.

Rydberg atoms in ultra cold atomic systems are particularly interesting for negligible motional dephasing (frozen Rydberg gas), strong interactions and various options to control them coherently. With this they are promising ingredients for quantum information processing [1–3] and quantum simulation [4]. Also exotic phases for Rydberg dressed ensembles of atoms [5–7] are proposed. These applications rely on coherent control of the strong interactions. Here we study the coherence in the presence of these interactions.

One possibility to control interactions between Rydberg atoms are so-called Förster resonances. Two dipole coupled pair states become degenerate and create a resonant dipole-dipole interaction between the atoms. As accidental degeneracy is unlikely, certain Rydberg states can be tuned into Förster resonance by microwave fields [8, 9] or a small electric field [10]. Different magnetic substates can be coupled by different polarizations of the coupling dipole. This generates diverse angular dependencies for different Förster resonances. Thereby Stark tuned Förster resonances offer the possibility to control both, the interaction strength and the angular dependence by switching small electric fields. They have been studied in several seminal experiments in terms of dipole blockade [11], line shape analysis [12], double-resonance spectroscopy [13] and excitation statistics [14]. Until now these experiments did not resolve the magnetic splitting of the Förster resonances.

In order to study coherent control of these interactions, interferometric methods offering phase sensitivity are well suited. As already pointed out by Ramsey in 1950 [15] interferometric schemes relying on separated oscillating fields are advantageous in many aspects compared to a single pulse of the coupling field. Besides an increased spectral resolution it allows to study coherent phenomena not being limited by spatial inhomogeneities of the coupling field. Ramsey interference methods were already used to investigate the coherence in resonant microwave coupling of single-atom Rydberg states [16] and in the coupling between pair states [17]. These experiments could not coherently control the excitation and could not study the decoherence directly at the Förster resonance. To our knowledge so far no experiment has been performed that coherently controls both the laser excitation and the interaction of Rydberg atoms.

Here, we apply optical Ramsey spectroscopy to coherently excite and de-excite $^{87}$Rb atoms to the $44d$ Rydberg state. These experiments can be viewed as an atom interferometer, similar to the atom-molecule interferometer in [18]. The phase of the two arms of the interferometer can be tuned independently by small electric fields. A full coherent control over the electronic state and the phase of the atoms is realized. Using this Ramsey spec-

FIG. 1: (a) Single shot Ramsey spectrum for a pulsed electric field of $E_z = 0.3$ V/cm (blue dots) and least square fit to the data (red line). $\Delta$ is the detuning of the exciting laser to the atomic resonance. (b) Pulse sequence used throughout this paper. (c) Color coded Ramsey spectra for varying pulsed electric fields. With increasing electric field a phase shift of these fringes occurs that depends quadratically on the electric field. A loss in visibility at 0.08 V/cm, 0.13 V/cm and 0.21 V/cm is visible, marked by the arrows in c). (a,c) Parameters for Ramsey measurements: $\tau_p = 0.15 \mu s$, $\tau_d = 1.0 \mu s$, $\rho = 1.2 \cdot 10^{12}$ cm$^{-3}$
troscopy we explore the dephasing at Förster resonances of the channel

\[ 44d_{5/2} + 44d_{5/2} \rightarrow 46p_{3/2} + 42f. \]  

(1)

Several magnetic substates of the 42f-state, split by fine structure coupling, Stark and Zeeman effect, can contribute to this resonance. These substates can be tuned into Förster resonance at slightly different electric fields, reducing the fringe visibility due to interaction induced dephasing. This splitting of the Förster resonance is resolved in the measurements, as shown in Fig. 1.[1] and 2.[2] Experimentally 87Rb atoms are initially prepared in the \( f=2 \) \( m_f=2 \) ground state in a magnetic trap. After evaporative cooling the atom number is varied by a Landau-Zener sweep and the trap offset is adiabatically tuned above the critical temperature for Bose-Einstein condensation. Throughout this paper the atoms are coherently excited to the 44d\(_{5/2}\) \( m_j=5/2 \) Rydberg state via a two-photon process, detuned by \( 2\pi \cdot 400 \text{ MHz} \) to the intermediate 5p\(_{3/2}\)-state. The total laser linewidth is below \( 2\pi \cdot 100 \text{ kHz} \) and the single atom two-photon Rabi frequency is \( \Omega \approx 2\pi \cdot 100 \text{ kHz} \). Details about the experimental setup can be found in [19]. For Ramsey spectroscopy two short laser pulses of \( \tau_p = 0.15 \mu s \) separated by a variable delay time \( \tau_d = 0 \ldots 2 \mu s \), are applied to the atoms (Fig. 1[b]). The Rydberg atom number \( N_{Ryd} \) is measured after the second light pulse by field ionization and ion detection of all Rydberg states. The sequence of excitation and detection is repeated 401 times in one atomic sample so that one entire Ramsey spectrum is measured in one atomic cloud. No averaging over spectra from different atomic samples is necessary. Fig. 1[a)] shows such a single shot spectrum. The appearence of a Ramsey fringe pattern in frequency space proves the coherence of the excitation process.

Additionally a pulsed electric field \( |\vec{E}| \) is switched on within 20 ns during the entire delay time (Fig. 1[b]). The electric field component \( E_z \) along the long axis of the magnetic trap was calibrated by measuring the Stark effect of the 44d-state. Note that a small radial electric field, possibly on the order of 0.35 V/cm, can not be controlled in the experiment and contributes to \( |\vec{E}| \).

Due to the high polarizability of Rydberg atoms the electric field detunes the Rydberg state relative to the exciting laser during the delay time when no excitation light is applied. This generates a phase shift

\[ \phi = -\frac{1}{\hbar} \int_0^\tau \frac{\alpha}{2} |E(t)|^2 dt \]

between the Rydberg and the ground state atoms, where \( \alpha \) is the polarizability of the 44d-state. A phase shift in the interference fringes appears and is experimentally apparent in the quadratic dependence of the fringe pattern on the electric field in Fig. 1[b]). No loss in the visibility of the fringes is visible even for phase shifts of almost \( 12\pi \) at 0.35 V/cm. This shows the remarkable stability of the coherence with respect to homogeneous fields and realizes complete coherent control over the state of the atoms.

This Ramsey interferometer is now used to study the Förster resonances. Here, the visibility \( V \)

\[ V = \frac{\max(N_{Ryd}) - \min(N_{Ryd})}{\max(N_{Ryd}) + \min(N_{Ryd})} \]

provides an observable that is sensitive to decoherence and dephasing processes. It is obtained from a fit to each individual spectrum (see Fig. 1[b]). Figure 2 shows the normalized visibility \( \tilde{V} = V/\bar{V} \), where \( \bar{V} \) is the mean
visibility of each dataset, for different densities of ground state atoms. For high densities distinct dips in the visibility can be seen at 0.08 V/cm, 0.13 V/cm and 0.21 V/cm. For decreasing densities of ground state atoms these features diminish. For the lowest attainable density of $1 \cdot 10^{10}$ cm$^{-3}$ the noise level is increased due to the weak signal. However, none of the dips are visible. The disappearance of the features for lower densities is a clear sign for an interaction process. We attribute these features to Förster resonance interaction.

The origin of the loss in visibility at the Förster resonances can already be qualitatively understood in a two-body picture. Dipole-dipole coupling of a pair of atoms in the $44d$-state to the $46p$ and $42f$-states during the electric field pulse will lead to a phase shift of the doubly excited state relative to a state where only one atom is excited. This leads to a dephasing within the system and is visible as a reduction of the visibility in the Ramsey fringes. Furthermore, an emerging population in the $p$ and $f$-states will not be coupled by the second light pulse of the Ramsey sequence, reducing the visibility in the Ramsey fringes even more.

However, since the dipole-dipole interactions depend on the interatomic distance as $1/r^3$, inhomogeneous Rydberg atom distributions in the experiment will lead to bands of interaction shifts for the collectively excited atoms [20]. This results in an additional loss of visibility in the experiment. Furthermore, in our experimental conditions many-body effects are expected to contribute to the strength of the dephasing and the lineshape [21, 22] and have to be taken into account for a qualitative study. However, the resonance positions are not expected to be notably shifted by many-body phenomena.

Note that the loss of coherence due to the Förster interaction does not lead to a reduced resolution in the spectroscopy since in the experimental sequence the atoms are excited far off the Förster resonance.

To calculate the required fields for a Förster resonance we diagonalize the Hamiltonian

$$H = H_0 + H_F + H_B,$$

where $H_0$ is the single atom field-free Hamiltonian, $H_F = \vec{d} \cdot \vec{E}$ is the electric field and $H_B = \vec{\mu} \cdot \vec{B}$ the magnetic field Hamiltonian. The electric and magnetic dipole matrix elements, $d_{ij}$ and $\mu_{ij}$ respectively, are calculated from numerical integrations of the Schrödinger equation, using the quantum defects from [23–25]. These calculations are done following the approach of [10] but taking a magnetic field into account. From the eigenstates in the magnetic and electric field the crossings of the pair states of equation 1 and the angular dependent interaction strength

$$U(\Theta) = \sqrt{2} \cdot \langle 42f | \langle 46p | V_{dd}(\Theta) | 44d \rangle | 44d \rangle$$

of the resonances can be calculated. $V_{dd}(\Theta)$ is the dipole-dipole interaction operator and $\Theta$ the angle between the direction of the electric field and the interatomic axis. Compared to calculations without magnetic field we get additional splittings and more resonances. Figure 3 shows the dependence of the electric fields that tune the pair state potentials into resonance as a function of the magnetic field. Here, the magnetic and electric fields are parallel. Most of the resonant pair states are not coupled, as the involved single atom states, depending on their eigenstates in the magnetic and electric field, are not dipole coupled. Hence they do not induce interactions. The angular maximum $U_{\text{max}}$ of the strength of the interaction at resonance is indicated by the diameter and the color of the dots. In zero magnetic field three resonances exist that are dipole-dipole coupled. They differ in the involved magnetic substate of the $46f$-state. In a basis formed of magnetic quantum numbers for electron spin and orbital angular momentum $(|m_s, m_l\rangle)$ the substates at $B = 0$ G can be identified as $|\frac{1}{2}, 3\rangle$, $|\frac{1}{2}, 2\rangle$ and $|\frac{1}{2}, 1\rangle$ for the resonances at 0.234 V/cm, 0.182 V/cm and 0.164 V/cm respectively. Small differences to [10] are due to the different quantum defects. In a finite magnetic field the states mix and split in several substates. The resonance at $|\vec{E}| = 0.234$ V/cm is almost independent of the magnetic field.

In Fig. 2 the calculated resonant electric fields at 13.55 G are indicated by diamonds. In the experiment only three resonances could be clearly identified. The resonance at $E_z = 0.21$ V/cm was observed to be not shifted by the magnetic field, as shown in Fig. 3. Based on this magnetic field dependence the measured resonances were attributed to the calculations as indicated by the dashed lines in Fig. 2. Calculations show that the discrepancy can be explained by a reasonable, experiment-
The Ramsey interferometer in combination with tuneable Stark shifts of the pair states. This is clearly below the splitting of the resonances and opens the possibility to tune the angular dependence of the interaction. So far the resolution was on the same order as the splitting [9]. Furthermore coherence times for atoms in the 44d-state at and near the Stark-tuned Förster resonance were obtained. The next steps will involve a detailed study of the origin of the dephasing process. Dephasing measurements on the resonant energy transfer process [17] will be performed in future measurements. In order to reduce the dephasing due to an inhomogeneous arrangement of atoms a more ordered system like an optical lattice or several small dipole traps could be used, where possibly interactions induce a collective phase shift of the atoms. Control over the angular dependence of the interaction will create an anisotropy in the Rydberg blockade and accordingly an anisotropic Rydberg density distribution. The Ramsey interferometer in combination with tuneable strong two-body interactions is a well suited tool to study and control this angular dependence of the interactions.

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[1] D. Jaksch et al., Phys. Rev. Lett. 85, 2208 (2000)
[2] M.D. Lukin et al., Phys. Rev. Lett. 87, 037901 (2001)
[3] M. Saffman et al., Rev. Mod. Phys. 82, 2313 (2010)
[4] H. Weimer et al., Nature Phys. 6, 382 (2010)
[5] N. Henkel et al., Phys. Rev. Lett. 104, 195302 (2010)
[6] G. Pupillo et al., Phys. Rev. Lett. 104, 223002 (2010)
[7] J. Honer et al., Phys. Rev. Lett. 105, 160404 (2010)
[8] K. Afrousheh et al., Phys. Rev. Lett. 93, 233001 (2004)
[9] P. Bohlouli-Zanjani et al., Phys. Rev. Lett. 98, 203005 (2007)
[10] A. Reinhard et al., Phys. Rev. A 75, 032712 (2007)
[11] T. Vogt et al., Phys. Rev. Lett. 97, 083003 (2006)
[12] I.I. Ryabtsev et al., Phys. Rev. Lett. 104, 073003 (2010)
[13] A. Reinhard et al., Phys. Rev. Lett. 100, 233201 (2008)
[14] A. Reinhard et al., Phys. Rev. A 78, 060702 (2008)
[15] N.F. Ramsey, Phys. Rev. 78, 695 (1950)
[16] I.I. Ryabtsev et al., J. Phys. B 36, 297 (2003)
[17] W.R. Anderson et al., Phys. Rev. A 65, 063404 (2002)
[18] B. Butscher et al., Nature Phys. 6, 970 (2010)
[19] R. Löw et al., arXive: quant-phys 0706.2639 (2007)
[20] D. Comparat et al., J. Opt. Soc. Am. B 27, 208 (2010)
[21] I. Mourachko et al., Phys. Rev. Lett. 80, 253 (1998)
[22] K.R. Younge et al., Phys. Rev. A 79, 043420 (2009)
[23] W. Li et al., Phys. Rev. A 67, 052502 (2003)
[24] C.-J. Lorenzen et al., Phys. Scr. 27, 300 (1983)
[25] J. Han et al., Phys. Rev. A 74, 054502 (2006)
[26] L. Allen et al., Optical Resonance and Two-level Atoms Dover Publications (1987)
[27] I.I. Beterov et al., Phys. Rev. A 79, 052504 (2009)