Back and forth transfer and coherent coupling in a cold Rydberg dipole gas

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Coupling by the resonant dipole-dipole energy transfer between cold cesium Rydberg atoms is investigated using time-resolved narrow-band de-excitation spectroscopy. This technique combines the advantage of efficient Rydberg excitation with high-resolution spectroscopy at variable interaction times. Dipole-dipole interaction is observed spectroscopically as avoided level crossing. The coherent character of the process is linked to back and forth transfer in the $np + np \leftrightarrow ns + (n + 1)s$ reaction. Decoherence in the ensemble has two different origins: the atom motion induced by dipole-dipole interaction and the migration of the $s$-Rydberg excitation in the environment of $p$-Rydberg atoms.

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Cold ensembles of Rydberg atoms are particularly interesting quantum systems because of the strong but controllable interactions between the atoms. Thermal motion of the atoms is mostly negligible on the timescale of Rydberg excitation. In such "frozen Rydberg gases" resonant dipole-dipole energy transfer has been observed, quite analogous to the migration of excitons in an amorphous solid. The many-body nature and coherent character of these resonant transfer processes have been clearly demonstrated.

This makes cold Rydberg gases promising candidate systems for fast quantum information schemes as was suggested using tunable resonant dipole-dipole interaction associated with energy transfer resonances. In particular, the Rydberg-Rydberg interaction may be exploited to induce a phase in a conditional phase-gate operation and to inhibit excitation of pairs of Rydberg atoms within a mesoscopic volume. The signature of local blockade of Rydberg excitation due to long-range interaction has been observed in experiments with narrow-band laser excitation and line broadening due to resonant dipole-dipole interactions has been observed using microwave spectroscopy. It has recently been realized that dipole-dipole interactions between Rydberg atoms may also feature dynamics with dramatic effects such as state redistribution phenomena, Penning ionization, and subsequent plasma formation.

The aim of this Letter is to understand the complete dynamics of reacting pairs of close Rydberg atoms in the environment of a dense Rydberg sample taking into account many-body processes as well as the relative motion of the interacting pairs of atoms. We show that both effects lead to a loss of coherence in the evolution of the quantum Rydberg ensemble. We consider the process

$$np_{3/2} + np_{3/2} \leftrightarrow ns + (n + 1)s \quad (1)$$

with $n = 24$ and $n = 25$, which is resonant at electric fields $E_0 = 59.11$ and $E_0 = 44.05 \text{ V/cm}$, respectively. Unless stated otherwise the magnetic component of the $np_{3/2}$-atoms is $|m_J| = 1/2$. In contrast to experiments employing narrow-band Rydberg excitation as a means of probing interactions between the atoms we use broad-band pulsed laser excitation and time-delayed probing by narrow-band de-excitation. This technique combines the advantage of high Rydberg densities and high-resolution spectroscopy at variable interaction times.

The Rydberg atoms are excited in a cloud of up to $10^7$ Cs atoms produced in a standard vapor-loaded MOT at residual gas pressure of $3 \times 10^{-10}$ mbar. At the trap position, a static electric field and a pulsed high voltage field can be applied by means of a pair of electric field grids spaced by 15.7 mm. The magnetic quadrupole field of the MOT is switched off during the Rydberg excitation phase. For Rydberg excitation we use a pulsed (7 ns, $\sim 100 \mu J$) dye laser with $\sim 10 \text{ GHz}$ spectral width running at 10 Hz repetition rate. The Rydberg atoms are subsequently probed by a single-mode tunable Ti:Sa laser of up to 400 mW output power. The beams of the pulsed dye and Ti:Sa lasers are coaxially aligned and weakly focused into the atomic cloud.

The excitation scheme used in the first experiment presented in this Letter is schematically depicted in Fig. 1(a). Up to $6 \times 10^5$ atoms are excited by the pulsed dye laser from the $6p_{3/2}$ upper MOT state to the $25p_{3/2}$ Stark-Rydberg state. To fulfill the energy transfer res-
resonant coupling of the electric field is tuned to a value such that the energy of a pair of $np$-atoms denoted by $|pp\rangle$ matches the energy of a pair of $ns$- and $(n+1)s$-atoms $|ss\rangle$ by the Stark effect. In the two-atom picture, resonant dipole-dipole interaction lifts degeneracy of the unperturbed states which linearly combine to the coherent repulsive and attractive interaction states, $|+\rangle = (|pp\rangle + |ss\rangle)/\sqrt{2}$ and $|-\rangle = (|pp\rangle - |ss\rangle)/\sqrt{2}$, respectively. The corresponding energy curves $V_+ (E)$ and $V_- (E)$ are the eigenvalues of the perturbed two-level Hamiltonian as a function of electric field $E$:

$$
\begin{pmatrix}
V_{pp}(E) & \Delta V \\
\Delta V & V_{ss}(E)
\end{pmatrix},
$$

where $V_{pp}(E)$ and $V_{ss}(E)$ stand for the energies of the unperturbed states $|pp\rangle$ and $|ss\rangle$ and $\Delta V$ denotes the dipole-dipole interaction energy. The coherent character of the transfer has been demonstrated using a Ramsey interference method. In that work, the migration of the products of resonant energy transfer in the Rydberg ensemble is associated with observed dephasing of interference fringes.

In the experiment, up to 50% of the population of $p$-atoms which are in the interacting magnetic sublevel are transformed into pairs of $s$ and $s'$-atoms within 300 ns interaction time. Subsequently, the $25p$-Rydberg atoms are coupled to the $7s$ state by the Ti:Sa laser. The Ti:Sa laser is applied continuously during 1 μs before selectively detecting the number of $25p$ and $26s$ atoms by applying a field ionizing high voltage pulse with $\sim 1$ μs rise time and recording the ion signal with gated integrators. Since the lifetime of the $7s$ state is short (56.5 ns) compared with the Rydberg lifetime ($\sim 10$ μs), tuning the Ti:Sa laser into resonance leads to a drop of the number of detected Rydberg atoms. The Ti:Sa laser frequency is recorded using a commercial wavemeter with 3 MHz resolution (Angström WS-8).

Typical depumping spectra of the $25p_{3/2}$ state at two different values of the electric field are displayed in Fig. 2. The number of $25p$ and $26s$ Rydberg atoms is normalized to the number of excited atoms without depumping laser, which is $6 \times 10^5$ and $1.5 \times 10^5$, respectively. The displayed spectra are the results of averaging over 4 individual scans and low-pass filtering to eliminate shot-to-shot fluctuations which are in the range of 30%. On resonance, the number of $25p$-atoms drops to about 40%, which is mainly due to the fact that both $|m_j|=1/2$ and $|m_j|=3/2$ components are excited by the broadband pulsed laser but only $|m_j|=1/2$ is coupled by the narrow-band depumping laser. Remarkably, we are also able to observe the reduction of $26s$ atoms by as much as 40%, as depicted in Fig. 2. This observation implies that resonant coupling of $s$-atoms back to $p$-atoms is also efficient. However, the position of the depumping resonance of the $s$-signal is shifted to higher frequencies when the electric field is tuned to values slightly higher than transverse resonance (Fig. 2(b)). Apparently, resonant coupling of $s$-atoms back to $p$-atoms is efficient only when exciting the symmetric repulsive state $|+\rangle$, which in turn remains nearly perfectly coherent during the depumping period. These results can be interpreted as a consequence of relative motion of the pairs of atoms leading for the attractive case ($|-\rangle$) to $I$-mixing of states in the manifold. For pairs of atoms which are more closely spaced by a factor 5 than average the displacement within 1 μs is about 10 μm, which is comparable with the average distance between Rydberg atoms. This assumption is supported by recent work demonstrating the motion of atoms induced by dipole forces. However, in our experiment we cannot directly observe such state mixing.

In Fig. 3(a) the positions of $p$ and $s$-lines obtained from fitting Lorentzian functions to the experimental spectra are plotted versus electric field. The corresponding $26s$-atom number normalized to the $25p$-atom number without depumping laser is shown in Fig. 3(b) including a Lorentzian fit curve. The linear fit curve in Fig. 3(a) illustrates the Stark shift of the $p$-state $V_{pp}$. The dashed lines result from fitting to the $s$-line positions the eigenvalue curve $V_+ (E)$ of expression 2. Free fit parameters are $V_{ss'}$, which is assumed to be independent of the electric field, and $\Delta V$. The resulting value $\Delta V/h = 15(3)$ MHz has to be compared with the expected value, which can be estimated by $\Delta V_{theo} \approx (\mu_{25s25p}\mu_{26s25p})/R^3$ in atomic units (a.u.), where $\mu_{25s25p} = 241$ a.u. and $\mu_{26s25p} = 237$ a.u. denote the transition dipole moments, and $R$ stands for the mean distance between interacting Rydberg atoms. The resulting value $\Delta V_{theo}/h \approx 0.5$ MHz is much smaller than the experimental one which is attributed to many-body effects inducing a wide energy band as in a disordered solid rather than a splitting of degenerate levels in the simple two-atom picture. We have carefully
Eq. 2 obtained from a least-squares fit of $V$ in an incoherent sum of spectra (open and filled diamonds, respectively) for various $s$-values of the electric field as shown in Fig. 2. The straight line is a linear fit of the positions of $p$-lines, the dashed lines indicate the interaction potential curves $V_s(E)$ and $V_p(E)$ from Eq. 2 obtained from a least-squares fit of $V_s(E)$ to the $s$-line positions. (b) The filled circles and the Lorentzian fit curve represent the number of 26$s$ atoms relative to the number of 25$p$ atoms without depumping laser.

checked that the effect was not an artefact due to the shape of the field ionization pulse which could lead to (a-)diabatic following of the Stark manifold.

Coherence of the dipole coupled interaction state $|+\rangle$ is interrogated by measuring the back and forth coupling efficiency for variable evolution times of the Rydberg sample. For this, the narrow-band depumping pulse is time-delayed with respect to the excitation pulse. The field ionizing high-voltage ramp is applied at constant time 10$\mu$s after the excitation pulse. The open diamonds in Fig. 3 represent the depletion amplitudes of 26$s$-atoms which are obtained from Lorenzian fits of the 26$s$-lines. The plotted amplitudes are normalized to the 26$s$-signal recorded without depumping pulse. The initial depumping amplitude of up to 60$\%$ continuously decreases down to roughly 30$\%$ at 1$\mu$s and longer interaction time. We interpret this behavior as a loss of the coherence after 1$\mu$s, due to the many-body migration of the products $s, s'$ of the reaction $|+\rangle$ through the inhomogeneous $p$-Rydberg ensemble $|+\rangle$. For longer times, the ensemble is in an incoherent sum of $p$ and $s, s'$-populations, where secondary reactions between $s$ and $s'$ atoms explain the residual depumping. The contribution to decoherence from the thermal motion of the Rydberg atoms is expected to be negligible since the average displacement of an atom in 1$\mu$s is only $\sim 0.1$ nm.

In a complementary experiment, the dynamics of resonant excitation transfer is investigated on the timescale of several microseconds. Rydberg atoms are now excited by a combined scheme of pulsed excitation, spontaneous emission, and cw-excitation of the Rydberg levels as illustrated in Fig. 1 (b). The branching ratio for transitions from 7$p_{3/2}$ to 7$s$ intermediate state is 44$\%$. Atoms are excited selectively to the $24p_{3/2}|m_f|=1/2$ Rydberg state by switching on the Ti:Sa laser during $t_1=0.3$ $\mu$s after the dye laser pulse by means of an acousto-optic modulator. This pump pulse transfers the maximum number of atoms into the Rydberg state. After an off-period of $\Delta t = 0.1-10$ $\mu$s a second pulse of $t_2=1$ $\mu$s duration is applied for down-stimulating the Rydberg atoms to the short-lived 7$s$-state as in the previous experiment. The field ionizing high-voltage pulse is applied at a fixed time of 5$\mu$s after the dye laser pulse for the measurements with $\Delta t = 0-3$ $\mu$s and 15$\mu$s after the dye laser pulse for the measurements with $\Delta t = 4-10$ $\mu$s.

The observed 24$p$-resonance is depicted as a dashed line in Fig. 4 for an electric field $E = 59.11$ V/cm which corresponds to maximum excitation transfer. Depumping of the $p$-atoms takes place at the center of the excitation line for two reasons. On the one hand, the Rabi frequency is largest on resonance which leads to maximum depletion of the Rydberg. On the other hand, a small number of photo-ions ($\lesssim 100$) is created by the Ti:Sa laser, presumably by photo-ionization of the 7$p_{3/2}$-state. The space charge from these ions induces inhomogeneous Stark broadening during Rydberg excitation. However, the ions are accelerated out of the Rydberg sample within 300 ns at 59.11 V/cm and the depumping line is unaffected by photo-ions. The depumping minimum nearly reaches zero in this scheme since all excited Rydberg atoms are subject to subsequent de-excitation.

The $s$-lines for three different off-periods of the Ti:Sa laser are plotted vertically shifted from each other for the sake of distinctiveness. The vertical scale indicates the $s$-signal amplitude relative to the maximum $p$-signal. Up to 50$\%$ of $p$-atoms are converted into $s$-atoms, which requires many-body interactions to govern the excitation transfer process $|+\rangle \rightarrow |s\rangle$. When comparing the depumping minima of $p$ and $s$-lines one notices that again depletion of the $s$-atoms is efficient only on the high-frequency side of the spectral line. In fact, the $p$ and $s$-depumping line positions qualitatively reproduce a similar $E$-field dependence as the one shown in Fig. 3. However, the fitted interaction energy amounts to $\Delta E \approx 1$ MHz which can be explained by a factor 6 lower Rydberg density than in the first experiment using pulsed Rydberg excitation.

For an increasing off-period $\Delta t$ of the Ti:Sa laser the depumping efficiency clearly diminishes. In fact we ob-
serve a hole in the spectrum only up to $\Delta t \sim 1.5 \mu s$, and a reduced signal amplitude for longer times. In order to quantify the depumping dynamics the $s$-lines are fitted by the heuristic model function $N_s(\nu) = L(\nu) \times (1 - G(\nu))$, where $L$ and $G$ stand for Lorentzian and Gaussian functions. During the fitting procedure the peak positions are held fixed as well as the Lorentzian and Gaussian functions. The resulting Gaussian amplitude factor which may vary between zero (no depumping) and unity (depumping down to zero) is plotted versus $\Delta t$ as filled diamonds in Fig. 4. Depumping efficiency drops fastest during the first microsecond which reflects loss of coherence of the system as previously mentioned. In the entire time range up to $10 \mu s$, the data are modelled by a power-law decay which is plotted as smooth line to guide the eye. When comparing the data for broad-band and narrow-band Rydberg excitation one notices different initial depumping amplitudes which has already been discussed, but time evolution qualitatively agrees for both experiments.

In conclusion, we have reported on narrow-band time-resolved spectroscopy of a gas of interacting Rydberg atoms, leading to a better understanding of decoherence of an interacting Rydberg ensemble. Excitation transfer at the attractive potential branch is not observed, presumably due to state-mixing dipole-induced collisions. This first result indicates the frontier between a frozen Rydberg gas where the thermal motion of atoms is negligible and a dipole gas where the motion between atoms induced by the dipole forces dominates the dynamics. In such a dipole gas, any initial coherence is rapidly destroyed. Decoherence of the repulsive interaction state takes place on a time scale of $\sim 1 \mu s$ which can be attributed to the far migration of the products of reacting pairs of atoms. This second result seems to confirm the dynamics of coherence of such quantum Rydberg ensembles as observed by $[4]$. Further experiments and new theoretical developments are necessary to reach complete understanding of such quantum systems.

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