Rydberg atoms on a chip and in a cell
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Link to publication

Citation for published version (APA):
Tauschinsky, F. A. (2013). Rydberg atoms on a chip and in a cell
In this chapter we discuss the spatially resolved, coherent excitation of Rydberg atoms near the surface of an atom chip. Electromagnetically Induced Transparency (EIT) is used to investigate the properties of the Rydberg atoms near the gold coated chip surface. We measure distance dependent shifts ($\approx 10 \text{ MHz}$) of the Rydberg energy levels caused by a spatially inhomogeneous electric field. The measured field strength and distance dependence is in agreement with a simple model for the electric field produced by a localised patch of Rb adsorbates deposited on the chip surface during experiments. The EIT resonances remain narrow ($< 4 \text{ MHz}$) and the observed widths are independent of atom-surface distance down to $\approx 20 \mu\text{m}$, indicating relatively long lifetime of the Rydberg states. Our results open the way to studies of dipolar physics, collective excitations, quantum metrology and quantum information processing involving interacting Rydberg excited atoms on atom chips.
7.1 INTRODUCTION

In this chapter we report the coherent excitation of Rydberg atoms on an atom chip [134]. To probe the atom-surface potential and lifetime of the Rydberg states we employ excited-state Electromagnetically Induced Transparency (EIT) [65, 135] as discussed in chapter 2. The position and width of the narrow transmission resonance reflect the energy and lifetime of the Rydberg state and provide a sensitive probe of the atom-surface interaction. This is related to a recent experiment investigating Rydberg excitation in a room temperature glass vapour cell at very small distances to the walls [117]. Here we investigate the effects of a metallic atom-chip surface with ultracold Rydberg atoms.

This is motivated by our aim to employ Rydberg blockade to optically control interactions between atomic ensembles prepared in separate magnetic micro-traps on a magnetic lattice atom chip [86, 87], similar to the chip design discussed in Chapters 4 and 8. Here, an unknown factor is the influence of the nearby metallic surface on the lifetime and coherence properties of the excited Rydberg atoms.

7.2 ELECTROMAGNETICALLY INDUCED TRANSPARENCY

Electromagnetically induced transparency is a coherent interference effect where the absorption on a resonant transition between two states is strongly modified by a coupling to a third state, creating a narrow transparency window. A common EIT configuration [71, 136] is the ladder-type system investigated here. We probe the absorption on the 5s-5p transition of $^{87}$Rb, with the 5p-state strongly coupled with a resonant laser to a highly excited nd or ns Rydberg state, as is depicted in Figure 7.1 a).

In this experiment the coupling laser is frequency stabilised directly to the $5p \rightarrow n\ell$-transition such that the transparency window is near the center of the resonance. In the limit of low probe intensity the susceptibility is given by

$$\chi(\Delta_p) \propto \frac{i \Gamma_p}{\Gamma_p + 2i \Delta_p + \frac{\Omega_c^2}{\Gamma_c + 2i(\Delta_p + \Delta_c)}}$$

where the probe absorption is proportional to the imaginary part $\text{Im}(\chi)$ [71], also see 2.3.1 and Equation 2.23. In this equation $\Gamma_p$ and $\Gamma_c$ denote the decay rates of the probe- and coupling resonances respectively, $\Delta_p$ and $\Delta_c$ the detunings and $\Omega_c$ the Rabi frequency of the upper transition (Figure 7.1 a).

If the Rydberg energy level shifts due to atom-atom or atom-surface interaction this leads to a detuning of the coupling laser from resonance which is visible as a shift of the transparency window in the absorption profile by a frequency $\Delta_c$. Furthermore, if the lifetime of the Rydberg state decreases, e.g. through induced decay to neighbouring states, this leads to a broadening of the transparency window, visible as an increase of $\Gamma_c$. Excited-state EIT is thus a sensitive probe to measure interactions in a Rydberg state. Compare the discussion in chapter 2 for more background on Rydberg states as well as origins and effects of EIT.
Figure 7.1: (a) Level scheme of our system; shifts in the Rydberg state are denoted by $\Delta_c$, additional decay channels lead to a broadening of the Rydberg transition, i.e. an increase in $\Gamma_c$. (b) Schematic picture of the experimental Setup. Probe beam, coupling beam and the imaging system are depicted. (c) Typical absorption image taken on the 5s – 5p transition. The chip is located at the top of the figure. The induced transparency due to the coupling laser is visible around $x = 100 \mu m$.

7.3 EXPERIMENTAL SETUP

We trap approximately $7 \times 10^5 \ ^{87} \text{Rb} \ |F = 2, m_F = 2 \rangle$ atoms in a magnetic Ioffe-Pritchard type trap. The magnetic field required for this trap is produced by a z-shape wire 0.3 mm beneath the atom chip surface. The surface is coated with a 100 nm gold layer and electrically isolated from the rest of the chip. Unlike the new design discussed in chapter 4 this chip does not possess a quartz passivation layer above the gold coating. The chip also features a permanent-magnet FePt layer that has been used to create arrays of micro-traps [86, 87], but this does not significantly influence the experiments discussed here. The atoms are cooled to temperatures of a few $\mu K$ by forced evaporative cooling.

Prior to detection, the atoms are released from the trap and expand freely for 2 ms in a uniform magnetic field of 1 G parallel to the probe beam. Typical peak atomic densities are $3 \times 10^9 \text{ cm}^{-3}$. After expansion the vertical extension of the cloud is $100 \mu m$ and the cloud center is $130(10) \mu m$ from the surface. An absorption image of this cloud is shown in Figure 7.1 c). The expanded cloud minimises possible effects of atom-atom interactions and allows simultaneous probing of many atom-surface distances.

We measure Electromagnetically Induced Transparency (EIT) spectra by simultaneously pulsing on a circularly polarised probe and a counter-propagating linearly-polarised (perpendicular to the chip surface) coupling laser beam for a time of 0.15 ms and recording an absorption image of the probe with a CCD camera for a variable detuning $\Delta_p$. The resolution
of our imaging system is $\approx 7 \, \mu m$. The probe beam is approximately uniform and much larger than the atom cloud, whereas the counter-propagating coupling laser beam is passed through an aperture and is imaged onto the atoms. The width of the coupling beam is $\approx 60 \, \mu m$ and it extends $\approx 150 \, \mu m$ from the chip surface; significantly smaller than the size of the atom cloud, i.e. only part of the cloud is exposed to the coupling beam and thus rendered transparent. This situation is depicted in Figure 7.1 b), and the spatial extent of the coupling beam is clearly visible as a light region (low optical density) centered around $x = 100 \, \mu m$, $z = 50 \, \mu m$ in Figure 7.1 c).

The probe laser is frequency-stabilised directly to the $F = 2 \rightarrow F' = 3$ transition using Doppler-free polarisation spectroscopy in a vapour cell. We estimate the linewidth of the laser around $\approx 500 \, kHz$ by beating it against an identical laser stabilized in the same way as the laser used here. To measure absorption spectra it is frequency shifted using a pair of double-pass AOMs which allow detunings between $\pm 20 \, MHz$. The coupling beam is produced by a frequency-doubled cw diode laser (Toptica TA-SHG). It is directly locked to the Rydberg state of interest by vapour cell EIT as described in [78]. Compare subsection 3.1.4 and in particular Figure 3.3 for details on this system¹. This allows direct stable locking to both s- and d- Rydberg states in the range $n = 19 \ldots 70$; the fine structure of the d-states is also well resolved in the spectroscopy. We estimate the combined (two-photon) linewidth of the Rydberg excitation lasers to be between 0.5 and 2 \, MHz, depending on the Rydberg state used.

We extract spectra from a series of absorption images such as the one shown in Figure 7.1 c). By analysing a series of such images taken for different detunings of the probe laser we construct an absorption spectrum as a function of detuning and distance to the surface. The coupling laser remains fixed in both position and frequency. For each detuning we take one image with the coupling laser present, and a reference image without the coupling laser. By analysing each pixel of these images separately we obtain spatially resolved information about the transparency of the sample. In particular, this allows us to extract information for a large number of distances from the surface of the atom chip at once. The dashed line in Figure 7.1 c) denotes the peak EIT signal and is used to extract EIT spectra at different distances to the chip. A small angle between the beam profile and the surface normal helps to align the coupling beam parallel to the surface and minimise fringing.

It is important to realise that the atom chip used in the experiments discussed here is not the same as that described in chapter 4, but rather an earlier generation, described in more detail in [87]. Of the differences between those two generations only one is of particular relevance to the results discussed here: The older generation did not have a SiO$_2$ passivation layer above the reflective gold surface. Instead the gold was exposed directly, leading to a large dipole moment for adsorbed rubidium atoms, and subsequently to the effects discussed here. Further differences include changes in the silver foil defining macroscopic wires, different locking methods for the probe laser and a different imaging system for perpendicular imaging. These changes however do not have a significant bearing on the results presented here. We can not yet present measurements of the electric fields near the new generation atom chip.

¹While the setup discussed in chapter 3 is based on saturated absorption $\text{fm}$ spectroscopy rather than polarisation spectroscopy for stabilizing the frequency of the probe laser, this is the only difference to the setup discussed here, and of no further consequence for the results presented here.
7.4 EIT SPECTROSCOPY

Figure 7.2: Optical spectra extracted from a series of absorption images for 80 probe detunings, with coupling to the $38d_{5/2}$ Rydberg state. Square points indicate the measured optical density spectrum taken in the absence of the coupling laser, with a fit to Equation 7.1 with $\Omega_c = 0$ (dashed line). Circles correspond to the EIT spectrum with the solid line a fit to the data using Equation 7.1. Both spectra are obtained at a distance of $\approx 200 \mu m$ from the surface of the chip.

Figure 7.2 shows an absorption spectrum measured in the absence of the coupling beam (square points), along with a spectrum measured with the coupling beam present (circles), for a relatively large atom-surface distance of $z \approx 200 \mu m$. The former simply reflects the natural absorption line-shape of the $^{87}\text{Rb} \, 5s \rightarrow 5p$ transition and is used as a reference. The latter spectrum shows the EIT dip resulting from strong coupling to the $38d_{5/2}$ Rydberg state. The transparency window is close to the center of the $5s \rightarrow 5p$ resonance. We can extract relevant parameters, in particular the linewidth and the detuning of the Rydberg state from these measurements by fitting the model of Equation 7.1. For the spectrum of Figure 7.2 we find $\Delta_c = 0.54(7) \text{ MHz}$ and $\Gamma_c = 0.61(8) \text{ MHz}$. The narrowest spectrum we have observed had a linewidth of $0.4(3) \text{ MHz}$ for the state $23s_{1/2}$ at a distance of $115 \mu m$ from the chip, consistent with our estimates for the combined laser linewidth. The non-zero fitted value of $\Delta_c$ is due to a small offset in the locking of the probe- and coupling lasers, and constitutes a constant
offset which is subtracted in later analysis. The measured EIT spectra demonstrate the coherent excitation of Rydberg atoms on an atom chip, in the distance range relevant to typical atom chip experiments. The width of the resonance is currently determined by the linewidth of the excitation laser system.

7.5 SURFACE EFFECTS

Figure 7.3 shows the fitted detuning of the EIT resonances as a function of distance to the surface for various excited states in the range \( n = 22 \rightarrow 36 \). Example spectra with fits for some observed level shifts are shown in Figure 7.3 (a,b) for the state \( 36d_{5/2} \) and in Figure 7.3 (c) for the state \( 23s_{1/2} \). The dashed vertical line indicates the extrapolated position of the EIT resonance for large distances (subtracted in Figure 7.3 d); a significant distance-dependent shift of the fitted resonance position is clearly recognisable. A positive shift of the detuning is equivalent to a shift of the Rydberg level towards higher energies.

We investigate \( s\)-states \( \ell = 0, j = \frac{1}{2} \) as well as two different \( d \) finestructure states where either \( \ell = 2, j = \frac{5}{2} \) or \( \ell = 2, j = \frac{3}{2} \). The distance range probed in the experiments is between 20 \( \mu m \) and 200 \( \mu m \) and in this range we observe shifts between \(-15 \) MHz and \(+20 \) MHz. The measured \( n \) dependence is \( \propto n^{6.2 \pm 0.3} \), however there are significant differences between measured shifts for \( s, d_{3/2} \) and \( d_{5/2} \) states.

Notably, the measured shifts for the \( d_{5/2} \) states have the opposite sign as compared with the \( s_{1/2} \) and \( d_{3/2} \) states. This we attribute to the differing DC electric polarisability \( \alpha \) for the studied states indicating the shifts originate from spatially varying electric fields. We compute \( \alpha \) from perturbation theory based on the analytical radial matrix elements in terms of Appell \( F_2 \) functions, including their dependence on \( j \) and \( |m_j| \) quantum numbers [137], using precise values for the \(^{87}\)Rb quantum defects [56]. The calculated values are in excellent agreement with tabulated measurements for the scalar and tensor polarisabilities from previous Stark shift measurements of \( s \) and \( d \)-states of \(^{85}\)Rb [138, 139]. In particular we find that for \( nd_{5/2,|m_j|=1/2,} \), \( \alpha \) is negative for the measured states, whereas the \( s \) and \( d_{3/2,|m_j|=3/2} \) states have a positive sign.

From the measured shifts we conclude that the dominant states probed in these experiments are \( s_{1/2,1/2}, d_{3/2,3/2} \) and \( d_{5/2,1/2} \). The other \( |m_j| \) states are expected to shift at a different range of distances than those investigated. For example, for the state \( 22d_{5/2} \) the electric polarisability \( \alpha \) for \( |m_j| = \frac{3}{2} \) and \( |m_j| = \frac{5}{2} \) is about 3 and 10 times larger respectively than for the probed state \( |m_j| = \frac{1}{2} \).

The spectra also provide information on the Rydberg lifetime through the parameter \( \Gamma_c \). As can be seen in Figure 7.3 (e) the fitted values of \( \Gamma_c \) do not measurably depend on the distance to the chip surface. The observed linewidths are of the order of the linewidth measured at large distances to the chip, which is limited by the linewidth of our laser system (see Figure 7.2 a). For some of the data we see evidence for other \( |m_j| \) states as double EIT resonances, which may result in artificially large fitted values for \( \Gamma_c \). Some broadening of the EIT resonances (\( \Gamma_c \sim 0.2 \Delta_c \)) is also expected due to integration over the inhomogeneous electric field over the atomic distribution along the probe axis during imaging. The largest fitted value for \( \Gamma_c \) we observed in any measurement was below 4 MHz, giving a lower bound for the Rydberg lifetime of 0.04 \( \mu s \).
Figure 7.3: Panels (a) – (c) show absorption spectra for three different points marked in panel (d). Markers and colors correspond to those used in Figure 7.2. The vertical lines in these figures denote the resonance position at large distances. The main figure in panel (d) shows measured shifts of the EIT resonance for various excited states as a function of distance from the surface. The measurement of 27d_{5/2} indicates typical error bars (standard deviation) calculated from repeated measurements of the same state over a time-span of 3 weeks. The lower panel (e) shows fitted values for $\Gamma_c$ for all states presented.
Figure 7.4: Local electric field strength inferred from the measured Rydberg energy-level shifts and averaged over the probe direction. The solid line shows the results of a fit to the data assuming the field originates from an Gaussian patch of surface dipoles (see inset) with $d_0 = 7 \times 10^5$ Debye $\mu$m$^{-2}$, and radius $w = 100 \mu$m, averaged over the $e^{-1/2}$ cloud radius of $\sigma_y = 130 \mu$m. The dotted line indicates the peak field strength at the center of the atom cloud ($y = 0$).

Shown in Figure 7.4 are values for the local electric field strength $|\vec{E}| = \sqrt{2\Delta c/\alpha}$ inferred from the measured Rydberg energy-level shifts. The measurements are identical to those presented in Figure 7.3; they span from $n = 22 - 36$ and include $s$, $d_{3/2}$ and $d_{5/2}$ angular momentum states covering a wide range of electric polarisabilities. In this form, all data fall on a single curve, confirming that the measured line shifts are caused by an inhomogeneous electric field originating from the chip surface. The height dependence of the field strength roughly follows a weak power-law decay $\propto z^{-0.7}$, thus excluding effects due to Van der Waals interactions ($\propto z^{-3}$) [15] or randomly distributed patch potentials expected to scale with $z^{-2}$. We also expect a contribution of the Van der Waals interaction for e.g. 26s of only $\approx 0.1$ kHz at 80 $\mu$m. The presented data was obtained over a time-span of approximately 4 weeks of experiments, indicating the observed electric field is relatively stable and reproducible from day-to-day.

We account for our measurements by assuming the electric field is produced by a patch of Rb adsorbates deposited on the chip surface. Following [14], we treat each adsorbate as an electric dipole oriented perpendicular to the chip surface and assume a patch of adsorbates which reflects the Gaussian distribution of atoms released from the magnetic trap. Integrating
the field over the distribution of dipoles gives the total electric field as a function of distance from the surface. At the center of the patch the field is given by

$$E_z(Z) = \frac{d_0}{2w e_0} \times \left[ -Z + e^{\frac{1}{2}Z^2} \sqrt{\frac{\pi}{2}} \left( 1 + Z^2 \right) \text{Erfc} \left( \frac{Z}{\sqrt{2}} \right) \right]$$  \hspace{1cm} (7.2)$$

where $Z = z/w$, $d_0$ is the peak dipole density and $w$ is the $e^{-1/2}$ patch radius. Finally, we include averaging of the measured line shifts over the width of the atom cloud $\sigma_y = 130 \mu m$ (along the probe axis at the time of detection). This model is then fit to the data by least squares minimisation as a function of $z$ to extract the dipole density and patch size. The solid-line in Figure 7.4 shows the result of a fit to the entire data set. The fit parameters are a peak dipole density $d_0 = 7 \times 10^5$ Debye $\mu m^{-2}$ and a patch size $w = 100 \mu m$, which is comparable to the radius of the atom cloud after expansion and the distance at which the trapped cloud is released. The dashed-line indicates the inferred field strength at the center of the cloud which is roughly 20% larger than the cloud averaged field strength.

From the fit results and for a dipole moment of order 10 Debye per ad-atom [14] we estimate a total of $\sim 10^9$ adsorbates on the surface. Assuming at steady state a decay rate of the order of $2 \times 10^{-6}$ s$^{-1}$ (room temperature surface [14]) and an experimental cycle time of 30 s, this corresponds to $3 \times 10^5$ deposited atoms per shot, comparable to the number of atoms released in each run of the experiment ($N = 7 \times 10^5$). Since these measurements, we have attempted to clean the surface of adsorbates by resistively heating the z-shaped wire beneath the chip surface ($I_z = 14 A$, conductor temperature $T \approx 80 ^\circ C$) for 48 hours, however this was insufficient to significantly raise the surface temperature and did not have an appreciable effect on the measured Rydberg line shifts. Briefly exposing the surface to oxygen may be a better method to eliminate the electric field produced by rubidium adsorbates.

7.6 CONCLUSION

The present experiments focus on spatially resolved excitation of Rydberg atoms on an atom chip. We create Rydberg atoms in a cloud of rubidium atoms on an atom chip, and investigate electromagnetically induced transparency near the chip surface. Absorption imaging in conjunction with the recording of EIT signals is used to obtain spatially resolved EIT spectra. We measure significant shifts of the Rydberg levels as we approach the surface which we attribute to electric fields produced by ad-atoms adsorbed on the chip surface. A theoretical model of the field produced by a Gaussian distribution of surface dipoles is in good agreement with our data. We do not observe significant broadening of the Rydberg levels. The measured shifts are not expected to inhibit the coherent creation and investigation of Rydberg atoms on atom chips.

We clearly see that the shifts are directly proportional to the polarisabilities of the states in question. This opens new possibilities to use Rydberg excited atoms as a sensitive, spatially resolved probe of electric fields [140, 141]. A simple estimate of our present sensitivity to electric fields is $\approx 0.1 \text{ V cm}^{-1}$ with a resolution 7 $\mu m$. The field sensitivity could be straight forwardly improved using narrow linewidth lasers and higher-lying Rydberg states.
The surface effects due to ad-atoms observed here could be prevented in future experiments by incorporating a magnetic field gradient to push the atoms away from the surface at the end of each experimental cycle. Furthermore, use of other coating materials on the chip surface could decrease the dipole moment of ad-atoms, or increase the desorption rate of these atoms. This last suggestion was indeed implemented in the current generation atom chip, using a SiO$_2$ coating as topmost layer to reduce the ad-atom dipole moment, as discussed in chapter 4.

Highly excited Rydberg atoms on atom chips, as demonstrated here for the first time, open new avenues for the study of dipolar physics, strongly interacting systems, and quantum information science in tailored trapping potentials. Atom chips allow the preparation of small atomic clouds [87] ideal for investigating dipole-dipole interactions and collective excitations in dense ($10^{15}$ cm$^{-3}$) mesoscopic ensembles [24, 142]. Furthermore, Rydberg atoms on atom chips are very attractive for studying long-range Van der Waals interactions with surfaces [15] and interfacing ultracold atoms with on-chip structures [143, 144].

The next chapter will discuss our first results on loading atoms on an atom chip expressly designed for the purpose of studying such effects.