Ultracold chemistry of a Rydberg atom in a rubidium-87 BEC

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Stuttgart, 9. Mai 2016

Felix Engel

List of publications

[S1] M. Schlagmüller, T. C. Liebisch, H. Nguyen, G. Lochead, F. Engel, F. Böttcher, K. M. Westphal, K. S. Kleinbach, R. Löw, S. Hofferberth, T. Pfau, J. Pérez-Ríos, and C. H. Greene. "Probing an Electron Scattering Resonance using Rydberg Molecules within a Dense and Ultracold Gas". Physical Review Letters **116** (2016), 53001 (cit. on pp. 12, 24–27, 61).

In addition, at this time two other publications are in preparation which contain knowledge of this work.

- [S2] M. Schlagmüller, T. C. Liebisch, F. Engel, K. S. Kleinbach, F. Böttcher, U. Hermann, K. M. Westphal, A. Gaj, R. Löw, S. Hofferberth, T. Pfau, J. Pérez-Ríos, and C. H. Greene. "Ultracold chemical reactions of a single Rydberg atom in a dense gas". to be published (2016) (cit. on pp. 12, 26, 27, 51, 52, 55, 60–62, 64, 65, 68).
- [S3] T. C. Liebisch, M. Schlagmüller, F. Engel, H. Nguyen, J. Balewski, G. Lochead, F. Böttcher, K. M. Westphal, K. S. Kleinbach, A. Gaj, R. Löw, S. Hofferberth, T. Pfau, J. Pérez-Ríos, and C. H. Greene. "Controlling Rydberg atom excitations in dense background gases". J. Phys. B, under review (2016) (cit. on pp. 46, 50, 67).

Zusammenfassung

In den vergangenen Jahren hat sich das Forschungsgebiet der Rydberg-Physik in vielerlei Hinsicht weiterentwickelt. Besonders interessant sind die neugewonnenen Erkenntnisse über die Wechselwirkung zwischen einem Rydberg-Atom und ultrakalten neutralen Atomen des selben Elements. Ein Rydberg-Atom besitzt mindestens ein angeregtes Elektron mit einer sehr hohen Hauptquantenzahl. Die resultierende Elektronen-Wellenfunktion des Rydberg-Atoms nimmt dabei enorme Ausmaße an im Vergleich zu einem Grundzustandsatom. Die Anregung eines Rydberg-Atoms in einer ultrakalten und dichten Atomwolke, beispielsweise in einem Bose-Einstein Kondensat (BEK) ist von besonderem Interesse, da dabei in Abhängigkeit von der Hauptquantenzahl das Rydberg-Atom mit wenigen bis vielen Atomen des Kondensats wechselwirken kann. Ein solches System ermöglicht die Untersuchung eines theoretischen Models der Vielteilchen-Wechselwirkung [4] für den Übergang von Zweiteilchen-Wechselwirkung zur Vielteilchen-Wechselwirkung. Ein Rydberg-Atom mit einer Hauptquantenzahl von n = 150, das in einem BEK mit einer maximalen Dichte von $\rho = 5.2 \times 10^{14} \, \text{atoms/cm}^3$ angeregt wird, besitzt eine Ausdehnung von einigen Mikrometern und überlappt somit mit mehreren Zehntausend Grundzustandsatome des Kondensats. Hingegen dazu befinden sich für n = 40 nur wenige Grundzustandsatome innerhalb der Elektronen-Wellenfunktion. Die Kopplung zwischen dem Rydberg-Elektron und den ultrakalten Grundzustandsatomen des Kondensats ist dabei überraschend stark. Dies lässt sich auf den enormen Massenunterschied des Rydberg-Elektrons und der Grundzustandsatome zurückführen. Diese starke Kopplung des Rydberg-Elektrons ermöglicht die Anregung von Phononen im BEK, welche eine kollektive Oszillation des Kondensats auslösen können [5]. Der zugrunde liegende Effekt hierbei beruht auf der niederenergetischen Streuung des Rydberg-Elektrons an den ultrakalten Atomen in dessen Orbit. Selbiger Effekt kann zu einer Bildung von polaren und nicht-polaren ultralang-reichweitigen Rydberg-Molekülen führen [6, 7].

List of publications

Im Rahmen dieser Masterarbeit wurde ein Model zur Beschreibung der gemessenen spektralen Linienform einer Rydberg-Anregung in einem ⁸⁷Rb Bose-Einstein Kondensats entwickelt. Dabei zeigt sich, dass die Simulation der Spektren stark von den zugrunde liegenden Potentialen abhängt. Der Vergleich der Simulation mit und ohne der *p*-Wellen Korrektur mit den experimentell gemessenen spektralen Linienformen weist auf eine Kopplung zwischen dem Butterfly-Zustand, aus der entarteten (n-3) - Mannigfaltigkeit, und dem via Zwei-Photonen Anregungsschema adressierten nS Rydberg-Zustand hin. Des Weiteren ermöglicht die auf dem entwickelten Model basierende Simulation eine Beschreibung der kompletten spektralen Linienform in Abhängigkeit von den zugrunde liegenden Potentialen. Hingegen dazu konnten die vorherigen Ansätze [8–12] lediglich eine Linienverschiebung sowie eine Linienverbreiterung in Abhängigkeit von der Hauptquantenzahl bestimmen. Zusätzlich wurde in der vorliegenden Arbeit der Einfluss unterschiedlicher Strahl-Taillen des fokussierten Anregungslasers sowie verschiedener Positionen des Fokus auf die Linienformen diskutiert. Der Vergleich der Simulation mit einer Messung für unterschiedliche Fokus-Positionen zeigt eine bemerkenswerte Übereinstimmung. In dem zweiten Teil der vorliegenden Masterarbeit wurde die Dynamik eines Rydberg-Atoms, angeregt im Bose-Einstein Kondensat, mit einem semi-klassischen Ansatz simuliert, welcher für die Kollision des ionischen Rydberg-Kerns mit den umgebenden ultrakalten Atomen geeignet ist [13]. Der Vergleich der simulierten Dynamik mit den experimentell beobachteten Reaktionsprodukten zeigt, dass die Dynamik des Systems nicht nur durch den ionischen Rydberg-Kern dominiert wird, sondern auch durch die streuungs-induzierte Wechselwirkung zwischen dem Rydberg-Elektron und den Grundzustandsatomen des Kondensats stark beeinflusst wird. Des Weiteren ergibt sich aus dem Vergleich der modellierten Lebensdauern des Rydberg-Atoms mit den gemessenen Reaktionszeiten, dass eine quantenmechanische Beschreibung benötigt wird, um die für höhere Hauptquantenzahlen $(n \gtrsim 100)$ untypische lange Lebensdauer eines Rydberg-Atoms im Kondensat zu beschreiben. Die Arbeit soll eine weiterführende Erforschung des vorliegenden Systems fördern mit Hinsicht auf eine theoretische Arbeit zur Quantenreflexion sowie zur Beschreibung der Dynamik mit einem Vielteilchen-Ansatz.

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Introduction

In recent years, the intense study of Rydberg physics has led to a large and extensive research field. In particular, the interaction of a Rydberg atom with ultracold neutral atoms is of great interest to examine the electron-atom scattering properties. A Rydberg atom possesses an electron excited to a high principle quantum number and thus extends over a large volume compared to the size of a ground-state atom. By utilizing the high density of a Bose-Einstein condensate (BEC) and the large extension of a Rydberg atom, the interaction of a Rydberg electron with many neutral atoms can be studied. In addition, such a system provides the opportunity to test theoretical models over the range of few to many-body physics for low to high principle quantum numbers [4]. For a BEC with a peak density of 5.2×10^{14} atoms/cm³ and a Rydberg atom excited with a principle quantum number of $n = 40 \dots 150$, few to several ten thousand atoms are contained by the Rydberg electron orbit. Furthermore, the coupling between the Rydberg electron and neutral atoms is surprisingly strong due to the favorable mass ratio between the interaction partners.

Recently, it has been demonstrated, that a single Rydberg electron can excite phonons in a BEC, which can trigger a collective oscillation of the condensate [5]. The underlying effect is the low-energy scattering of the Rydberg electron with many ultracold neutral atoms from the BEC, which can also lead to a formation of polar and non-polar ultralong-range Rydberg molecules [6, 7]. These scattering-induced Rydberg molecules are formed as the result of the attractive interaction of the Rydberg electron trapping a neutral atom in its wave function at a localized position, leading to molecules with an immense bond distance of several thousand Bohr radii [14], which were spectroscopically observed for ⁸⁷Rb by Bendkowsky et al. [15, 16] and via atom loss for ⁸⁴Sr by DeSalvo et al. [17]. The bound-states of the polar molecules are provided by a set of high angular momentum states (trilobite state), which in combination with the giant internuclear separation leads to a large permanent dipole moment [6]. These fairly unusual polar homonuclear molecules, were observed for Rb in 2011 by Li et al. [18] with a dipole moment D on the order of 1 Debye and for Cs by Tallant et al. [19] with D = 15 - 100 Debye, respectively. Interestingly, due to the relatively small fractional nS quantum defect of cesium, which leads to a strong non-adiabatic coupling between the photon-associated nS Rydberg state and the close-by trilobite state, blueshifted ultralong-range Cs₂ Rydberg molecules with a dipole moment one hundred times larger than for Rb, are observable [19]. In general, these kind of scattering processes are sufficiently described in terms of *s*-wave scattering, for a non-singular *p*-wave contribution [20].

The calculations of I. I. Fabrikant [10] reveals the presence of a low-energy e^{-} -Rb(5S) p-wave shape resonance for an energy of $E_r = 23 \text{ meV}$, which was predicted by A. Johnston and P. Burrow [21]. It turns out, that this shape resonance leads to an additional set of high angular momentum states (butterfly state) [6, 7], resulting in a second type of polar ultralong-range Rydberg molecules. With higher densities of ultracold atoms, the molecular bound states become unresolvable as many neutral atoms overlap with the Rydberg orbit [S1, 5, 22]. The obtained Rydberg-BEC spectral line shapes, measured by Schlagmüller et al. [S1], exhibit features, which are evidence for the predicted low-energy e^{-} -Rb(5S) p-wave shape resonance. A microscopic model, which was developed within the scope of this work, reveals a strong dependence of the observed spectral line shapes on this shape resonance.

A promising application of the strong coupling between a Rydberg electron and the surrounding cold atomic cloud is an imaging technique, which can be used to image the Rydberg electron wave function [23]. For this technique, a sufficiently long lifetime of the Rydberg atom immersed in a quantum gas is necessary for an observable impact. It has also been shown by Balewski et al. [5], that the lifetime of a Rydberg atom in a BEC is reduced, but surprisingly long lifetimes were observed for higher principle quantum numbers ($n \geq 100$) by Schlagmüller et al. [S2]. A study of these long lifetimes reveals a reaction mechanism, which relies on the coupling of the photon-associated nS state with the butterfly state of the upper-lying degenerate hydrogenic manifold [S2]. As a part of this study, the dynamics of this system has been reviewed, in the framework of this master thesis using a semi-classical approach.

2

Rydberg spectroscopy in a Bose-Einstein Condensate

In this chapter the theoretical model and simulation developed within the framework of this master thesis are discussed, to understand the spectroscopic line shape resulting from a single Rydberg atom excitation in a Bose-Einstein condensate. The theoretical background for the developed theoretical model is introduced in the first section (2.1). Following this, the experimental realization as well as the interpretation of the experimentally obtained spectral line shapes is described in section 2.2. Based on the theoretical background, the developed microscopic model and the simulation method is presented in section 2.3.1. Subsequently, the results of the simulation are tested against the experimental results for different principle quantum numbers (section 2.3.1), an enlarged excitation beam waist (section 2.3.2), and a method to probe the density distribution of a Bose-Einstein condensate (section 2.3.3).

2.1 Rydberg atom - perturber atom interaction

A Rydberg atom excited in an ultracold and dense atomic cloud, strongly interacts with the surrounding neutral atoms. Due to the large extension of the Rydberg electron wave function, a large amount of neutral atoms are present inside the Rydberg electron orbit. This mainly leads to two different effects based on the polarizability of the neutral atoms. On the one hand, the exposed ionic core of the Rydberg atom attracts the neutral atoms of the BEC, whereas on the other hand the Rydberg electron can scatter at the neutral atoms inside its electron orbit. Due to the mass ratio of the Rydberg core and the Rydberg electron, the relative nuclear motion of the Rydberg core and the neutral atoms is much slower than the electronic motion of the Rydberg electron, meaning that one can apply the Born-Oppenheimer approximation. In fact, for the system at hand this approach is appropriate [6]. This allows to treat the interaction between the Rydberg atom and the atoms of the BEC separately for the Rydberg core and the Rydberg electron.

Moreover, the Rydberg core of an alkali metal e.g. rubidium can be treated as an ion with a single positive charge due to the shielding of the core by the inner lying electronic structure. This means, for an internuclear distance larger than the extension of the inner lying electronic structure of the Rydberg core, the interaction between the Rydberg core and each neutral atom of the surrounding ultracold and dense cloud is given by the polarization potential, which in turn depends on the polarizability of each individual neutral atom. In contrast, the interaction between the Rydberg electron and the neutral atoms is described by an electron-atom scattering process, as the Rydberg electron scatters at each neutral atom inside its electron orbit. This means the neutral atoms inside the Rydberg electron orbit do perturb the electron wave function and thus this neutral atoms are denoted as perturber atoms. In order to understand the interaction between a Rydberg atom and the surrounding neutral atoms of the ultracold and dense atomic cloud, a theoretical description of the ion-atom interaction and the electron-atom scattering is presented in the following.

2.1.1 Ion-atom interaction

In comparison to the size of a neutral atom, the electron density distribution of an atom in the nS Rydberg state is spread over a huge volume due to the large orbit of the Rydberg electron. Thus, as a simple approach one can neglect the presence of the Rydberg electron to study the interaction between the ionic Rydberg core and the surrounding neutral atoms. The interaction between a single ion and a neutral atom is dominated by the polarization potential; its asymptotic limit is shown in equation 2.3 [24]. Thus, for large internuclear distances, one can describe the ion-atom interaction between a neutral atom and the ionic Rydberg core using the asymptotic limit of the polarization potential of equation 2.3. The interaction strength for large internuclear distances is proportional to the dipole polarizability of a neutral atom. A classical approach of the ion-atom interaction leads to the same result [25]. In the following this classical approach is shown.

Due to the finite spatial charge distribution of a neutral atom its polarizability α is non-zero and, therefore, an external electric field **E** induces a dipole moment **p** given by

$$\mathbf{p}(\mathbf{E}) = \alpha \cdot \mathbf{E}.\tag{2.1}$$

By using equation 2.1 one can derive the potential energy of an induced dipole moment in an external electric field as

$$V_{\rm dip} = -\int \mathbf{F} \,\mathrm{d}\mathbf{r} = -\int (\mathbf{p} \cdot \nabla) \mathbf{E} \,\mathrm{d}\mathbf{r} = -\frac{1}{2} \int \nabla (\mathbf{p} \cdot \mathbf{E}) \,\mathrm{d}\mathbf{r} = -\frac{1}{2} \alpha \left|\mathbf{E}\right|^2.$$
(2.2)

For the electric field of a single charge $(E(r) = r^{-2})$ this leads to the following polarization potential, given in atomic units by

$$V_{C_4}(R) \equiv -\frac{C_4}{R^4} = \frac{\alpha/2}{R^4},$$
(2.3)

where α is the polarizability of a neutral atom and R is the internuclear distance between the ion and a neutral atom. However, this polarization potential is only the asymptotic limit for large internuclear distances. For small internuclear distances the electron cloud of the ionic Rydberg core starts to sense the valence electron of the neutral atom. This leads to a strong repulsive interaction for very close distances, due to the Pauli exclusion principle.

The result of the calculation of an *ab initio* study of the ${}^{2}\Sigma_{g}^{+}$ of Rb₂⁺ molecule for the lowest energy state (blue), taken from [27], is shown in figure 2.1 as well as the polarization potential only (red) from equation 2.3. For the calculation of the potential energy curves, the ion-atom system of Rb₂⁺ is considered as a system with one active electron. To model the interaction of the ions and the one active electron a relativistic pseudopotential is used. The polarization of

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Fig. 2.1: Results of a *ab initio* study of the lowest potential energy curve (PEC) of ${}^{2}\Sigma_{g}^{+}$ of Rb₂⁺ and the polarization potential only. For the polarization potential a polarizability of $\alpha = 318.8(14)$ a.u. [26] of the Rb perturber atoms is used. On the left the potentials with a logarithmic axis for the internuclear distance are shown, where on the right the interaction strength of the attractive part is shown in a double-logarithmic illustration.

rubidium is given by a core polarization potential. Here we consider only the lowest potential energy curve, which asymptotically equals the 5S ground-state of the atoms surrounding the ionic Rydberg core. As is apparent from figure 2.1, the interaction strength and therefore also the potential energy curve of the ${}^{2}\Sigma_{g}^{+}$ of Rb₂⁺ and the polarization potential $V_{C_4}(R)$ starts to deviate for internuclear distances smaller than $30 a_0$. This means that for $R < 30 a_0$ the interaction between the valence electron of the perturber atom and the electron cloud of the ionic Rydberg core starts to matter. In fact, for a closer distance, the polarization potential underestimates the strength of the interaction and the relativistic pseudopotential of the *ab initio* calculated potential starts to take over.

2.1.2 Electron-atom scattering

Besides the interaction between the ionic Rydberg core and the neutral atoms, the interaction of the Rydberg electron with the neutral atoms (perturber atoms) is described by an electron-atom scattering process, as the Rydberg electron scatters with each neutral atom inside its electron orbit. Since the classical outermost turning point of the Rydberg electron scales with $(n^*)^2$ [28], where n^* corresponds to the effective principle quantum number, for a moderately high principle quantum number the extension of the Rydberg electron becomes huge compared to atomic proportions. By considering an ultracold cloud with a high density like a Bose-Einstein condensate, for a moderately high Rydberg state there is on average at least one perturber atom inside the Rydberg electron orbit. For achievable densities of the experimental setup (see section 2.2), e.g. for the peak density $\hat{\rho} = 5.2 \times 10^{14} \,\mathrm{cm}^{-3}$, a moderately high principle quantum numbers corresponds to n > 26. The investigated principle quantum numbers are $n = 40 \dots 149$, thus one has to take into account the scattering between the Rydberg electron and the perturber atoms in its orbit for the system at hand. For large distances the Coulomb interaction between the ionic Rydberg core and the Rydberg electron is weak and does not change rapidly with the position, as a consequence the momentum available for the scattering can be considered as low [7]. In particular, if the de Broglie wavelength of the electron is much larger than the interaction range, the scattering can be considered as a low-energy scattering process (s-wave dominated). For low-energy scattering the interaction potential between the scattering partners is short-ranged for r^{-s} with $s \ge 2$ [20]. Furthermore, this short-range interaction can be modeled as a contact interaction, where the interaction strength is proportional to the scattering length, leading to the well-known Fermi pseudopotential introduced by E. Fermi [8].

The pseudopotential for s-wave scattering of an electron-atom scattering process, in atomic units, is given by

$$V_{\text{pseudo}}(\mathbf{r}) = 2\pi a_s \,\delta(\mathbf{r}),\tag{2.4}$$

with the scattering length a_s of s-wave scattering and the Dirac delta function $\delta(\mathbf{r})$, which represents the contact interaction. On its part, the scattering length a_s depends on the internal states of the colliding particles. For the system at hand (see section 2.2), the appropriate scattering length is the triplet scattering

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length of e^{-} -Rb(5S), which is used in the following discussions. An important thing to note is, by using the contact interaction we assume a local perturbation of the electron wave function. In other words, due to the presence of a perturber atom inside the Rydberg electron orbit, the wave function is modified on a length scale, which varies slowly with position. As long as this assumption holds one can calculate the electron-atom scattering potential by integrating over the electron density distribution multiplied with the pseudopotential [9]. Therefore, with equation 2.4 this leads to the following potential energy resulting from the *s*-wave scattering:

$$V_{\text{scat}}(\mathbf{R}) = \int_{\mathbb{R}^3} V_{\text{pseudo}}(\mathbf{r} - \mathbf{R}) |\psi(\mathbf{r})|^2 \,\mathrm{d}\mathbf{r} = 2\pi a_s |\psi(\mathbf{R})|^2, \qquad (2.5)$$

where $|\psi(\mathbf{R})|^2$ is the electron density distribution, which is probed by its scattering partner at position \mathbf{R} .

In the case of many perturber atoms inside the Rydberg orbit this leads to a mean line shift of the Rydberg spectrum for a constant density of perturber atoms. Since the interaction range is small enough, one can use the contact interaction and thus, the perturbation of each perturber is localized and the energy contribution of all the atoms inside the Rydberg electron orbit add up, leading to a mean shift in energy $\Delta \overline{E}$. This can be shown by using equation 2.5:

$$\Delta \overline{E}(\rho) = \int_{\mathbb{R}^3} \sum_i V_{\text{pseudo}}(\mathbf{r} - \mathbf{R}_i) |\psi(\mathbf{r})|^2 \, \mathrm{d}\mathbf{r}$$

$$= \int_{\mathbb{R}^3} \rho \int_{\mathbb{R}^3} V_{\text{pseudo}}(\mathbf{r} - \mathbf{R}) |\psi(\mathbf{r})|^2 \, \mathrm{d}\mathbf{r} \mathrm{d}\mathbf{R}$$

$$= \rho \int_{\mathbb{R}^3} 2\pi a_s |\psi(\mathbf{R})|^2 \, \mathrm{d}\mathbf{R}$$

$$\Delta \overline{E}(\rho) = 2\pi a_s \rho, \qquad (2.6)$$

where ρ is the homogeneous density of the perturber atoms. The result of equation 2.6 was derived by Fermi [8] to explain the line shifts of the Rydberg absorption line explored by E. Amaldi and E. Segré in 1934 [29]. The spectroscopic line shapes, measured by Gaj et al. [22], suggest that the same mean shift approach holds in an ultracold environment.

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So far we neglected the increase of momentum of the Rydberg electron as it comes closer to the ionic Rydberg core. As the momentum increases, the kinetic energy available for the electron-atom scattering process increases as well. In a semi-classical approach, one can obtain the radial dependence of the wave number k(R) of the Rydberg electron by considering the energy of the Rydberg electron $-1/2(n^*)^2$ and the increase in energy 1/R when the scattering process takes place at the internuclear distance R between the ionic Rydberg core and the perturber atom [6]. In atomic units, this leads to the following implicit form of the wave number

$$\frac{1}{2}k^{2}(R) = -\frac{1}{2(n^{*})^{2}} + \frac{1}{R},$$
(2.7)

with the effective principle quantum number n^* . With this, we can rewrite the Fermi pseudopotential from equation 2.4 including the k-dependence of the Rydberg electron, in terms of atomic units as

$$\hat{V}_s(\mathbf{r}, \mathbf{R}) = 2\pi a_s[k(R)]\,\delta(\mathbf{r} - \mathbf{R}),\tag{2.8}$$

with the k-dependent scattering length of s-wave scattering $a_s[k(R)]$ which is defined as

$$a_s[k] \equiv -\tan \delta_s(k)/k,\tag{2.9}$$

using the corresponding scattering phase shifts $\delta_s(k)$. In analogy to equation 2.5, one can rewrite the scattering potential, including the modified scattering length $a_s[k(R)]$, in atomic units as

$$V_{\text{scat}}(\mathbf{R}) = 2\pi a_s[k(R)] |\psi(\mathbf{R})|^2.$$
 (2.10)

Furthermore, if the spin-orbit interaction is negligible, the two-body adiabatic potential energy curve (PEC) of an nS Rydberg electron and a perturber atom, can be calculated by using the Born-Oppenheimer approximation, which leads to the following result

$$U_{nS,k}(R) = -\frac{1}{2(n^*)^2} + V_{nS,k}(R)$$

= $-\frac{1}{2(n^*)^2} + 2\pi a_s [k(R)] |\psi_{nS}(R)|^2$, (2.11)

where ψ_{nS} is the wave function of the atomic nS Rydberg state, n^* is the effective principle quantum number and $V_{nS,k}(R)$ is the scattering potential.

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Fig. 2.2: Two-body adiabatic potential energy curves for the 53S + 5S state, with (red) and without (blue) the k-dependent triplet scattering length. For the k = 0 case the triplet scattering length $a_{s,\uparrow\uparrow} = -15.7(1) a_0$ from [30] is used. For the k-dependent triplet scattering length, adapted phase shifts from C. H. Greene based on calculations of I. I. Fabrikant, are used [31].

Figure 2.2 shows the two-body potential energies according to equation 2.11 for two ⁸⁷Rb atoms (one in the 53*S* Rydberg state and the other in the ground-state 5*S*) for a constant triplet scattering length (blue) and a *k*-dependent scattering length (red). The resulting PEC is proportional to the square of the absolute value of the electron wave function $|\psi_{nS}(R)|^2$ and therefore, both potential energy curves (with a constant scattering length and with a *k*-dependent scattering length) mimic the electron density distribution. This leads to the characteristic oscillatory shape of the calculated adiabatic potential energy curves. As is apparent from figure 2.2, the increasing kinetic energy of the Rydberg electron leads to shallower potential wells for smaller internuclear distances until the *k*-dependent scattering length changes its sign and thus the potential becomes repulsive ($R < 520 a_0$). In contrast, the PEC for the k = 0 case is attractive for all distances. This means by neglecting the kinetic energy, one overestimates the depth of the calculated potential energies significantly for closer distances.



Fig. 2.3: A illustration of the Rydberg electron density distribution $|\psi(\mathbf{R})|^2$ of (a) a butterfly state for an internuclear distance of $R = 1300 a_0$ and (b) a trilobite state for an internuclear distance of $R = 2700 a_0$ of the n = 50 hydrogenic manifold in Cartesian coordinate [31]. The ionic Rydberg core and perturber are aligned along the axis of symmetry. The perturber is located at the largest peaks in the density distribution.

For the potential energy curves calculated by the approach of equation 2.11, Greene et al. [6] predict the presence of two different types of molecular Rydberg states. On the one hand, the adiabatic potential energy curve of a Rydberg state with a low angular momentum (for Rb l < 3) supports bound-states for non-polar ultralong-range Rydberg molecules. On the other hand, a class of high-l states, which splits off of the degenerate hydrogenic manifold, leads to a second type of polar ultralong-range Rydberg molecules. In addition to the class of states, which leads to the polar molecules, Greene et al. [6] also predict a second class of high-l states, which splits off of the degenerate hydrogenic manifold, due to the existence of a shape resonance with a ${}^{3}\!P^{o}$ symmetry of the e^{-} -Rb(5S) scattering. Accordingly, the second class of states appears only if the *p*-wave scattering is included in the calculations. For closer internuclear distances the electron density distribution of a Rydberg atom in the state, which arises from the *p*-wave contribution, looks like a butterfly and thus is called butterfly state (see figure 2.3a) [7]. For the same reason the other class of states is denoted as trilobite state (see figure 2.3b) [6]. In contrast to the butterfly state, the trilobite state occurs for s-wave scattering only.

This shape resonance occurs, as a resonant coupling to a quasi bound state behind the centrifugal barrier, which arises for a non-zero angular momentum, is possible for a certain kinetic energy available for the electron-atom scattering process. Following the method of Omont [9], including a p-wave correction, one can obtain the interaction potential for the p-wave contribution as

$$\hat{V}_p(\mathbf{r}, \mathbf{R}) = -\frac{6\pi \tan \delta_p(k(R))}{k(R)^3} \,\delta(\mathbf{r} - \mathbf{R}) \overleftarrow{\nabla} \cdot \overrightarrow{\nabla}, \qquad (2.12)$$

with the corresponding p-wave scattering phase shifts $\delta_p(k(R))$. The arrows on top of the nabla operator in equation 2.12 indicate the acting direction, meaning that one operator acts on the wave function on the left hand side and the other on the wave function on the right hand side, for calculating the expectation value. The *p*-wave scattering term of equation 2.12 becomes dominant as $\tan \delta_p$ diverges for $\delta_p \to \pi/2$, meaning that one has to consider the *p*-wave contribution for the calculations. A proper interpretation of the consequences implementing the interaction potential \hat{V}_s and \hat{V}_p is given by Hamilton et al. [7]. Namely, the interaction part given by \hat{V}_s selects a linear combination of atomic states which maximizes the wave function at the perturber position, where \hat{V}_p selects a linear combination maximizing the derivative of the wave function at the perturber position. Therefore, the trilobite state is composed of a set of states, which maximizes the wave function at the perturber position, whereas the butterfly state consists of states, which maximizes either the wave function or its derivative at the perturber position, depending on the internuclear distance. In the following, figure 2.4 shows the used phase shifts for s- and p-wave scattering from I. I. Fabrikant, which are adapted by C. H. Greene [31], depending on the kinetic energy. Regarding equation 2.7 it is clear that the kinetic energy of the Rydberg electron depends on the internuclear distance R and the effective principle quantum number n^* . The phase shifts depicted in figure 2.4 are plotted versus the kinetic energy of a 53S Rydberg electron.

By recalling, that the *p*-wave shape resonance occurs at a certain kinetic energy of the Rydberg electron. It becomes clear that, as the perturber atom gets closer to the ionic Rydberg core, at a certain internuclear distance $R_{\rm res}$ the kinetic energy of the Rydberg electron meet the resonance condition, which in turn leads to a resonant coupling to a quasi bound state. At this certain distance the triplet *p*-wave phase shift δ_p reaches $\pi/2$ and due to the tangent in the interaction potential the energy seems to diverge. This divergence in energy



Fig. 2.4: Adapted triplet s- and p-wave phase shifts from C. H. Greene [31] based on calculations of I. I. Fabrikant for low-energy scattering of e^- -Rb(5S), as a function of the kinetic energy of the Rydberg electron and therefore as a function of the internuclear distance R between the ionic Rydberg core and the perturber atom, for a 53S Rydberg electron.

is non-physical, but it demonstrates that one can not neglect higher-order wave corrections for the scattering, if a shape resonance can occur. Nevertheless, due to level repulsion, in the case of low-energy electron-atom scattering the resulting energies of eigenstates are limited by the upper n + 1 and lower n - 1 lying hydrogenic manifold. Therefore, even without energy renormalization, applying a diagonalization method close to this resonance is reasonably accurate [7]. The p-wave phase shifts displayed in figure 2.4 reach $\delta_p = \pi/2$ for $E_{\rm kin} \approx 29$ meV, which corresponds to an internuclear distance of $R_{\rm res} \approx 800 a_0$ for a 53S Rydberg electron.

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Fig. 2.5: Two-body adiabatic potential energy curves between the hydrogenic manifolds of the principle quantum number n = 49 and n = 50, including the polarization potential V_{C_4} of the ion-atom interaction, with a polarizability of $\alpha = 318.8(14)$ a.u. [26] of the Rb perturber atoms. The asymptotic two-body interaction energy of the Rydberg state and the perturber atom (53S + 5S) is set to zero energy.

In order to obtain the PEC of ⁸⁷Rb for the interaction between the Rydberg electron and a perturber atom, based on the Born-Oppenheimer approximation and including the s-wave scattering $\hat{V}_s(\mathbf{r}, \mathbf{R})$ as well as the p-wave correction $\hat{V}_p(\mathbf{r}, \mathbf{R})$, a degenerate perturbation theory can be applied [S1]. Figure 2.5 shows the calculated adiabatic potential energy curves between the n = 49 and n = 50 hydrogenic manifold. For a large internuclear distance, the interaction between the Rydberg atom and the perturber atom vanishes and the potential energy levels split into four bare states. Namely, a class of high-*l* states with $l \geq 3$ (the so called hydrogenic manifold) and the low-*l* states 53*S*, 51*D* and 52*P*. The degeneracy of the energy levels of the low-*l* bare states is lifted from the corresponding manifold, as for a low angular momentum the probability to find the Rydberg electron close to the Rydberg core is enhanced and thus the shielding of the Rydberg core through the inner lying electronic structure becomes insufficient leading to a shift in energy. To take into account this so called quantum defect, one can introduce an effective principle quantum number n^* [28]. As one can see from figure 2.5, from each hydrogenic manifold both classes of high-l states (trilobite state and butterfly state) split off. In the vicinity of the *p*-wave shape resonance, the potential energy curve corresponding to the butterfly state crosses the two-body states 53S + 5S, 51D + 5S and 52P + 5S, where 5S denotes the perturber atom and nl the corresponding Rydberg state. The butterfly state couples to the low-*l* two-body states and thus an avoided crossing occurs at the crossing position. Based on the energy level repulsion of the avoided crossings one can see from figure 2.5 that the coupling of the butterfly state and the 53S + 5S state is weaker compared to the other two avoided crossings along the butterfly state. This is the case, as the coupling between the high-*l* states increases with higher angular momentum. The shape resonance position is reflected by the calculation (see figure 2.5), as close to the resonance the *p*-wave correction \hat{V}_p from equation 2.12 takes over and thus the PEC of the butterfly state exhibits a maximum gradient. In the following, it turns out that this *p*-wave correction is essential in order to understand the spectral line shape of a single Rydberg atom excited in an ultracold dense cloud like a Bose-Einstein condensate [S1]. The modeling of these spectra is one of the main subjects of this work and is presented in chapter 2.3.



2.2 Experimental realization

Fig. 2.6: Illustration of the realization of a localized Rydberg excitation in a BEC. For the excitation a collimated 420 nm laser beam aligned with the y-axis and a focused infrared laser with a minimal waist of 2.1(3) µm aligned with the z-axis is used. The quantization axis is given by an offset magnetic field in the y-direction. For a 111*S* Rydberg state the classical extension of the Rydberg electron orbit (cyan) is shown in the center of the BEC. A sketch in the top right corner shows the two-photon Rydberg excitation scheme as well as the involved energy levels.

The measured Rydberg spectra of [S1] and the studied reaction dynamics in [S2], which are modeled in section 2.3 and chapter 3, are performed in a pure spin polarized Bose-Einstein condensate (BEC) of approximately 1.4×10^{6} ⁸⁷Rb atoms, which is magnetically trapped in an elongated QUIC trap [32]. For this system the magnetically trapped spin polarized ground-state is given as $|5S_{1/2}, F = 2, m_F = 2\rangle$. The radial trapping frequency $\omega_r = 2\pi \times 200$ Hz and the axial trapping frequency $\omega_{ax} = 2\pi \times 15$ Hz of the QUIC trap lead to a Thomas-Fermi radius of 5 µm in radial and 66 µm in axial direction, which describe the spatial extension of a trapped BEC [33], as it is depicted in figure 2.6. For the experimental setup at hand, the number of trapped atoms and the used trap frequencies result in a peak density of $\hat{\rho} = 5.2 \times 10^{14}$ atoms/cm³ of the BEC. The Rydberg excitation is achieved by a two-photon excitation scheme using a collimated blue laser beam with a wavelength of 420 nm for the lower transition and an infrared focused laser beam with a minimal waist of 2.1(3) µm for the upper transition, as is illustrated in figure 2.6. For the twophoton excitation the ground-state is coupled to the $|nS_{1/2}, ms = 1/2\rangle$ Rydberg state by the intermediate state $|6P_{3/2}\rangle$. The intermediate state detuning is chosen such that the absorption and heating of the BEC, caused by the 420 nm excitation laser light, is low. Therefore, the intermediate state detuning is kept constant at $\Delta = 80 \text{ MHz} \approx 56 \Gamma$ with respect to the state $|6P_{3/2}\rangle$. The wavelength of the infrared laser light, which drives the upper transition, is tuned (1011 nm - 1019 nm) in order to address the corresponding nS Rydberg state (n = 40...149). The blue excitation laser light is circularly polarized (σ^+) , whereas the linear polarization of the infrared excitation laser is selected in a way ensuring that it is perpendicular to the magnetic offset field. In the schematic of figure 2.6 the magnetic offset field, which defines the quantization axis, is chosen parallel to the y-axis, and therefore the linear polarization of the infrared excitation laser light is parallel to the x-axis. For the spectroscopy measurements both excitation lasers are simultaneously pulsed for 2 µs, with a repetition rate of 2 kHz. For each pulse the created Rydberg atom is electric field ionized 400 ns after the excitation light is switched off. After the Rydberg atom is ionized the ion is detected by a microchannel plate. The used electric field strength is three times higher than the ionization threshold electric field for the corresponding nS Rydberg state. This is done to also detect Rydberg atoms, which are subject to an inelastic state-changing collision [S2, 34]. For the performed Rydberg-BEC spectra, which are subject of this work, the singleatom Rabi-frequency is kept constant at 250 kHz by adjusting the power of the infrared excitation laser for different principle quantum numbers, except the spectra taken for the n = 40, where the used power was doubled. This relatively small single-atom Rabi-frequency of the Rydberg excitation ensures that the probability of multiple Rydberg excitations in the BEC is negligibly low. For a typical experiment the same BEC can be used for up to 500 single Rydberg excitations. The repeated measurements cause a reduction in atom number and density, and thus all obtained data used for the evaluation of the Rydberg-BEC spectra [S1] and the reaction dynamics [S2] are tested to be independent of this atom loss. A more detailed explanation of the experimental setup can be found in the PhD thesis of M. Schlagmüller [34].

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Fig. 2.7: Typical spectrum of an nS Rydberg excitation in a Bose-Einstein condensate. Here for a principle quantum number of n = 53. The signal in region (1) is due to a Rydberg excitation with one or more perturber atoms near the avoided crossing between the 53S Rydberg state and the butterfly state from the upper lying manifold. In region (2) the Rydberg excitation takes place in an environment of high \rightarrow low density. Region (3) shows signal from the thermal cloud as well as Rydberg molecule lines from the low density region of the BEC and the thermal cloud. The signal in region (4) is provided by the avoided crossing, where the potential energy is above the atomic resonance.

2.3 Spectrum of a single Rydberg atom in a BEC

The experimental realization explained in the previous section 2.2 allows the measurement of a precise spectrum of a single Rydberg excitation in a Bose-Einstein condensate or in a dense thermal cloud. For the achievable principle quantum numbers ($n = 40 \dots 149$), the number of perturber atoms inside the Rydberg orbit ranges from 6 to 20,000 for the peak density ($5.2 \times 10^{14} \text{ atoms/cm}^3$) of the BEC. In figure 2.7 a typical Rydberg spectrum for a Rydberg state with principle quantum number of n = 53 is depicted. Due to the relatively high density in the BEC, a broad spectral line shape is observed, which extends

over more than 100 MHz. In contrast, a Rydberg spectrum of a thermal cloud exhibits discrete lines (due to the formation of Rydberg molecules [15]) or for a larger principle quantum number (e.g. n = 111) a broad line shape of the order of several MHz [22]. The measurement and evaluation of the presented spectra is expounded in the PhD thesis of M. Schlagmüller [34]. The resulting spectrum of a single Rydberg excitation in a BEC can be separated in four different regions. For a large red detuning from the atomic resonance (region (1)) the Rydberg atom is excited in a high density environment. The mean field approach introduced by Fermi, which leads to the mean density shift (equation 2.6) for a Rydberg atom excited in a very dense environment, can not explain the signal for a large red detuning ($\delta \ll -50$ MHz for a density of $\rho \approx 5 \times 10^{14}$ atoms/cm³).

The purpose of this section is to point out that the signal in region (1) can only be explained by considering one or more perturber atoms in the vicinity of the avoided crossing between the excited nS Rydberg state and the butterfly state from the upper lying (n-3) manifold. For the spectrum of a 53S Rydberg state the relevant potential energies are depicted in figure 2.8. In region (2) the Rydberg atoms are excited in an area of high or low density depending on the detuning δ . The signal in region (3) arises from the thermal cloud around the BEC, where the sharp peaks occur due to the formation of Rydberg molecules (dimer, trimer,...) [22] created in both the thermal cloud and in the low density region of the BEC. In region (4) one would expect a signal of the high density region of the BEC, since the avoided crossing provides potential energies ($\delta > 0$ MHz) larger than the energy of the bare 53S Rydberg state. This can be seen from the potential energies displayed in the following in figure 2.8b (red). Figure 2.8 shows in addition to the potential energies (b) the nearest neighbor distribution of three different densities in (a). For example in blue the distribution for the peak density of the BEC is displayed. The nearest neighbor distribution clarifies the importance of the *p*-wave shape resonance for the modeling of the spectral line shapes, as the probability to find the nearest neighbor peaks in the vicinity of the avoided crossing of the 53S Rydberg state and the butterfly state. This means that at least the nearest neighbor is strongly affected by the existence of the *p*-wave shape resonance. This also reveals that in the region (1) and (2) of a typical spectrum (see figure 2.7), one would expect a deviation to the mean density shift, since even for lower densities, e.g. 1×10^{14} atoms/cm³, the nearest neighbor experiences a lowered potential energy due to the avoided crossing of the Rydberg state with the butterfly state.



Fig. 2.8: (a) Nearest neighbor distributions of three relevant densities. In blue the nearest neighbor distribution for the peak density of the BEC. (b) A zoom in of the calculated potential energy curves, which is relevant for the modeling of the spectral line shape, with (red) and without (blue) taking into account the *p*-wave shape resonance. The avoided crossing, which arises from the *p*-wave contribution, appears at a distance of $R \approx 1420 a_0$ (red curve). The bare 53S Rydberg state is set to zero energy.

2.3.1 Microscopic model and simulation of a Rydberg spectrum

In the following, a model to explain the spectral line shape of a single Rydberg excitation in a Bose-Einstein condensate is presented. For this model the perturber atoms are treated as uncorrelated, point-like perturbers with zero velocity, where the binding energies are associated with the internuclear distance between the perturber atom and the ionic Rydberg core. Therefore, it is assumed that on the one hand all perturber atoms are affected by the same two-body potential and on the other hand the perturbation on the Rydberg electron wave function is small enough.

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Fig. 2.9: Positions corresponding to the scattering energy $E_{\rm res} = 29$ meV of the *p*-wave shape resonance and the avoided crossing of the *nS* Rydberg state and the butterfly state (BF) for different principle quantum numbers. The avoided crossing position is obtained from the calculation of the corresponding adiabatic potential energy curve. The dashed line shows the asymptotic limit $(n \to \infty)$ of the *p*-wave shape resonance position.

This assumption does not hold for few or more perturbers excited deeply in a potential region, which is associated with the butterfly state from the upper lying hydrogenic manifold, because in this area a single perturber influences strongly the Rydberg electron wave function leading to a strongly anisotropic electron density distribution with a shape of a butterfly (see figure 2.3a). Figure 2.9 shows the position corresponding to the scattering energy of the *p*-wave shape resonance as well as the position of the avoided crossing between the *nS* Rydberg state and the butterfly state for different principle quantum numbers. Both positions saturate for higher principle quantum numbers. This is not surprising, as the kinetic energy available for the scattering process in the limit of $n \to \infty$ is limited by the Coulomb interaction which does not depend on the principle quantum number. From equation 2.7 one can deduce, that the position of the *p*-wave shape resonance for $E_{\rm res} = 29 \,{\rm meV}$ saturates at an internuclear distance of around 940 a_0 , which is in accordance with the positions shown in figure 2.9.

For a principle quantum number of $n \ge 160$ a slight step occurs, due to the oscillatory shape of the PEC and thus for the obtained crossing positions. The avoided crossing position of the nS Rydberg state with the butterfly state seems to saturate for an internuclear distance of $R \approx 1900 a_0$. The avoided crossing positions in figure 2.9 are determined by the position at which the S-character along the adiabatic state is equal to 0.5. The S-character can be obtained from the calculations of the adiabatic potential energy curves by calculating the overlap of the bare Rydberg state with the eigenstate ψ_i as

$$s_i(R) = \left| \langle \psi_{nS}(\infty) | \psi_i(R) \rangle \right|^2, \qquad (2.13)$$

with the bare Rydberg state $\psi_{nS}(\infty)$ and the resulting S-character s_i depending on the internuclear separation of the perturber atom and the ionic Rydberg core. Regarding the excitation scheme (see section 2.2), which only allows to excite into a S-state, it follows that for a perturber placed at a distance R from the Rydberg core, the S-character $s_i(R)$ gives the probability to find this perturber atom in the eigenstate $\psi_i(R)$.

So far it is not motivated that applying the two-body potential for many particles inside the Rydberg electron orbit leads to a physical result at all. For a proper motivation one can define a so called *p*-wave shell to quantify the number of strongly influenced perturber atoms by the presence of the *p*-wave shape resonance. The shell is approximated by the volume of a spherical shell, where the inner and outer radii are given by the distance at which the S-character is below a certain value. In the following, by means of the p-wave shell, the mean number of perturber atoms in the vicinity of the *p*-wave shape resonance for an excitation of the nS Rydberg state is displayed in figure 2.10. From figure 2.10, it becomes clear that for a Rydberg atom excited in the center of the BEC ($\hat{\rho} = 5.2 \times 10^{14} \, \text{atoms/cm}^3$) on average less than one perturber atom is found in the vicinity of the *p*-wave shape resonance. Only for a S-character larger than 0.99 on average more than one perturber atoms are located in the *p*-wave shell. However, by considering the inset of figure 2.10, which illustrates the width of the shell for different S-character, it is easy to understand that this additional perturber atom is not located very close to the avoided crossing of the nS Rydberg state and the butterfly state, as in comparison to s < 0.75(orange) and s < 0.9 (red) the p-wave shell for s < 0.99 (blue) is significantly larger. Thus, this verifies for our system the treatment of many perturber atoms in the Rydberg orbit with a two-body potential.

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Fig. 2.10: Number of perturber atoms inside a *p*-wave shell for different *S*-character s(R) near the avoided crossing for a homogeneous density of $5.2 \times 10^{14} \text{ atoms/cm}^3$. The inset shows the *S*-character of the adiabatic states ψ_S and ψ_{BF} , which correspond asymptotically to the butterfly state and the 100*S* Rydberg state, respectively. The line color along the curve in the inset indicates the width of the *p*-wave shell, where the narrower shell is always included.

Finally, one has to note that the jumpy behavior for a increasing principle quantum number n occurs due to the oscillatory shape of the potential energy curve, which leads to an oscillation with R of the S-character. By considering the saturation of the p-wave shape resonance position, it follows that the mean number of perturber atoms inside the p-wave shell has to saturate as well for high principle quantum numbers.

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For the system at hand it is also important to verify that the de Broglie wavelength λ_{dB} of the electron is much larger than the interaction range r^* , as it was mentioned in section 2.1.2. The interaction range can be obtained from the stationary radial Schrödinger equation of a moving particle in the asymptotic potential V_{C_4} by introducing reduced units $x \equiv R/\sigma$ and $K \equiv k/\sigma$. Thus, in atomic units it follows

$$0 = \left(\frac{1}{2\mu}\frac{d^2}{dR^2} + \frac{C_4}{R^4} - E\right)\psi(R)$$

= $\left(\frac{d^2}{dx^2} + \frac{1}{x^4} - K^2\right)y(x),$ (2.14)

where $E = k^2/2\mu$ is the energy of a quasi free particle. One can easily verify the relationship between σ and C_4 as $\sigma = (2\mu C_4)^{1/2}$ by inserting the reduced units in equation 2.14. With this, the introduced length scale σ can be used to define the interaction range r^* as:

$$r^* \equiv \sigma = (2\mu C_4)^{1/2} \stackrel{[\alpha=2C_4]}{=} \sqrt{\mu\alpha}.$$
 (2.15)

The interaction range r^* can be understood as the distance at which the particle starts to feel the interaction potential. The de Broglie wavelength of the Rydberg electron is approximated with the semi-classical approach from equation 2.7, as $\lambda_{\rm dB} = 2\pi/k(R)$. The ratio of the de Broglie wavelength $\lambda_{\rm dB}$ and the interaction range of the electron-atom interaction r^* leads to the family of curves displayed in the following in figure 2.11, where the polarizability of the perturber atoms $\alpha = 318.8(14)$ a.u. [26] and the reduced mass $\mu = m_{\rm Rb}/2$ of ⁸⁷Rb is used. For the peak density of the BEC (5.2×10^{14} atoms/cm³) the nearest neighbor distribution peaks around $r_{\rm peak} \simeq 1300 a_0$. In this region, the ratio of the de Broglie wavelength and the interaction range is $\lambda_{\rm dB}/r^* \approx 10$ for the achievable principle quantum numbers. For larger distances the ratio increases in a monotonous way. The good agreement of the modeled spectral line shape, which is shown in the following, suggests that even for $\lambda_{\rm dB}/r^* \lesssim 10$ the presented theoretical approach is appropriate.

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Fig. 2.11: Ratio of the de Broglie wavelength of the Rydberg electron and the interaction range r^* defined by equation 2.15, where $\lambda_{dB} = 2\pi/k(R)$ depends on the internuclear distance (see equation 2.7), for different principle quantum numbers n = 40...200 of a nS Rydberg state.

In the following an approach to generate configurations of perturber atoms distributed inside the Rydberg electron orbit is presented. Thus, first of all the configurations are generated based on the density distribution of the Bose-Einstein condensate. Secondly, the spatial Rydberg excitation probability is taken into account. Afterwards the perturber atoms are placed in the already introduced potential energy curves. The interaction energy of the perturber atoms with the Rydberg atom leads to an energy shift. This means to excite a Rydberg atom with a specific configuration of perturber atoms inside its Rydberg orbit, one needs to match the energy shift, given by the interaction, with the detuning from the atomic resonance. Finally, the resulting configurations are used to obtain the spectral line shape of a nS Rydberg atom excited in a Bose-Einstein condensate.





Fig. 2.12: Density plot of the yz-plane of a simulated BEC with 1.4×10^{6} ⁸⁷Rb atoms. The upper color plot shows the BEC density distribution using the Thomas-Fermi approximation. The lower color plot illustrates the difference between the BEC with (left) and without (right) the additional atoms of the thermal distribution. Therefore, the maximum value of the column density on the left side is set as ten times smaller than on the right side. The adopted atom number and temperature of the thermal distribution (Gaussian) is 0.3×10^{6} and 300 nK, respectively.

Figure 2.12 shows the simulated density distribution of a magnetically trapped BEC as a false color image as well as the pixel sum along the y- and z-axis. To obtain the density distribution the Thomas-Fermi approximation is applied, which leads to a parabolic density profile for trapped BEC [33]. The radial and axial expansion is determined by the trapping frequencies $\omega_{\rm r} = 2\pi \times 200$ Hz and $\omega_{\rm ax} = 2\pi \times 15$ Hz according to the experimental setup. In the lower color plot of figure 2.12 the difference between the BEC with (left) and without (right) the additional atoms of the thermal cloud (Gaussian distribution) is illustrated. The number of atoms and the temperature of the thermal cloud is obtained by a bimodal fit of a time-of-flight measurement, performed with the experimental
setup presented in section 2.2. The Thomas-Fermi approximation breaks down at the edge of the BEC, since at the edge the atom number is lower and the kinetic energy term in the Gross-Pitaevskii is no longer negligible. Therefore, the density distribution at the edge of the BEC is enlarged on a typical length scale. In the literature, this length is called the coherence length or healing length. For the peak density of the BEC one can estimate the healing length by $\xi = \sqrt{4\pi \hat{\rho} a} \approx 0.17 \,\mu\text{m}$, where $\hat{\rho} = 5.2 \times 10^{14} \,\text{cm}^{-3}$ is the peak density and $a = 5.45 \,\text{nm}$ [35] is the corresponding scattering length of the ⁸⁷Rb atoms in the BEC. However, by comparing the density distributions of figure 2.12 (lower plot), it becomes clear that the added Gaussian distribution shows the most effect on the overall density distribution at the edge of the BEC. This means the effect of the added Gaussian distribution outweighs the widened BEC density near the edge. Thus, for the problem at hand it is appropriate to neglect the deviation from the parabolic density distribution of the BEC.

For the next step of generating configurations of perturber atoms, the spatial intensity profile of the excitation beams are implemented. Since, the blue excitation laser ($\lambda = 420 \,\mathrm{nm}$) is collimated and illuminates the entire cloud, only the focused infrared excitation laser ($\lambda = 1020 \,\mathrm{nm}$) is involved in the further calculations. In the following in figure 2.13 the atom distribution before and after the weighting of the spatial intensity profile along the elongated axis is illustrated in a false color. Here, a waist of 2.1 µm is assumed, which was experimentally determined using a test setup identical to the optics setup in the experiment, except for the glass cell of the vacuum chamber [36]. Since the beam waist is determined by a test setup, it may be that the waist in the actual experimental setup deviates a little from the assumed waist. Since, the laser intensity falls off exponentially with the square of the distance from the beam propagation axis (here z-axis), a cutoff distance is introduced, which defines an excitation volume $V_{\text{ex}}(z,n)$. All atoms outside this excitation volume are taken out of the atom distribution. A proper cutoff can be set at the distance $\tilde{w}(z,n) \equiv 2 \cdot w(z) + r_{\text{Ryd}}(n)$ from the propagation axis (z-axis), where $r_{\text{Ryd}}(n)$ is the classical outer most turning point of the nS Rydberg electron and w(z) is the waist of the Gaussian beam. The cutoff distance is enlarged by the expansion of the Rydberg electron of the corresponding Rydberg state to make sure that all perturber atoms inside the Rydberg orbit are present in the generated atom distribution.



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Fig. 2.13: Density plot of the yz-plane of a simulated BEC with 1.4×10^{6} ⁸⁷Rb atoms surrounded by a thermal cloud with a Gaussian distribution of 0.3×10^{6} atoms and a temperature of $T_{\text{thermal}} = 300 \text{ nK}$. The upper plot shows the density distribution before a weighting with the spatial profile of the excitation beam, where the lower plot shows the density distribution after the excitation beam is weighted. The excitation volume $V_{\text{ex}}(z, n = 53)$ is displayed in red.

For the illustration shown in figure 2.13 each atom *i* inside the excitation volume is assigned with an uniformly random number $rand_i$. Each random number $rand_i$ is compared to the beam intensity $I(r_i, z_i)$ at the position of the corresponding atom. All atoms with $rand_i < I(r_i, z_i)/I_0$, where I_0 is the peak intensity in the center of the beam, are taken out. This procedure corresponds to a convolution of the density distribution with the spatial beam intensity of the excitation lasers. After the spatial intensity profile is weighted, only 2% of the total atom number turn out to be relevant, due to the tightly focused excitation beam. For the actual calculation of the spectra the same definition of an excitation volume is utilized. However, the beam intensity is implemented in a different way, which is explained in the following paragraph.



Fig. 2.14: Two-body potential energy curves, with (red) and without (blue) including the *p*-wave shape resonance, for the 53*S* Rydberg state. The filled circles represent one of the simulated perturber atom distributions inside the Rydberg electron orbit at the center of the atom cloud. For this particular configuration of perturbers the resulting energy shift of *s*-wave scattering only is equal to -54.4 MHz, whereas the resulting energy shift of the *s*- and *p*-wave scattering is -81.8 MHz.

The next step in the modeling process is to take into account the interaction between the Rydberg atom and the perturber atoms inside the wave function of the Rydberg electron. Therefore, each atom inside the excitation volume $V_{\text{ex}}(z,n)$ of the generated atom distribution is treated as a possible Rydberg excitation. The energy levels of the *i*-th Rydberg atom, of the atom distribution, at the position \mathbf{r}_i are shifted by

$$\delta_i = \sum_{i \neq j} V(|\mathbf{r}_i - \mathbf{R}_j|), \tag{2.16}$$

with respect to the atomic resonance ($\delta = 0$), where \mathbf{R}_j denotes the *j*-th perturber atom inside the Rydberg electron orbit and V(r) represents the calculated two-body potential energy curves introduced in section 2.1. One has to note that the approach from equation 2.16 is only valid for a negligible back-action of a single perturber atom on the Rydberg electron wave function.

As an example, figure 2.14 shows a possible perturber atom distribution in the PEC, which includes the *p*-wave shape resonance (red) and in the PEC, which considers only s-wave scattering (blue). The energy shift for the potential energies in figure 2.14 are -54.4 MHz and -81.8 MHz for s-wave scattering only and s- and p-wave scattering, respectively. This example demonstrates, that the potential energy calculation, which includes the *p*-wave shape resonance, can lead to a much larger energy shift for the same perturber atom distribution. Finally, for the actual spectrum, the excitation bandwidth of the Rydberg excitation as well as the spatial Gaussian beam intensity has to be considered. For each atom i the energy shift δ_i is calculated. Afterwards, the spectrum is obtained by summing up the Lorentzian contribution of each atom i, which is weighted with the beam intensity at its position. In other words, every atom in the simulated atom distribution is treated as a possible Rydberg atom weighted with the intensity and the available energy given by the detuning of the excitation laser. Another perspective of this approach is that a specific configuration of perturber atoms is selected by the excitation laser. This means a certain detuning δ selects a specific configuration of atoms out of the present atom distribution in the excitation volume. Based on the density distribution of the BEC one can map this specific configuration on a density shell. This is shown in the following section 2.3.3. The actual spectrum is calculated as follows

$$S(\delta) = A \sum_{i} \frac{1}{\pi} \frac{\left(\frac{1}{2}\Gamma\right)^2}{\left(\delta - \delta_i\right)^2 + \left(\frac{1}{2}\Gamma\right)^2} \times I_i,$$
(2.17)

with the Rydberg excitation bandwidth Γ , the detuning from the atomic resonance δ of the addressed Rydberg state, the energy shift δ_i , the Gaussian beam intensity $I_i = I(\mathbf{R}_i)$ of the *i*-th atom in the created atom distribution and a normalization factor A. In order to increase the sample number, the procedure of calculating the atom distribution is repeated fifty times before the actual spectrum is obtained. In the following, the simulated spectral line shape of the 53S Rydberg state, with (solid red) and without (dashed yellow) including the *p*-wave shape resonance is depicted in figure 2.15. The total average ion count of the experimentally obtained spectrum is used to determine the normalization factor A in equation 2.17. Thus, the spectral line shapes are obtained without any free parameters. The large deviation of the line shapes with and without the *p*-wave contribution in figure 2.15, demonstrate a strong dependence on the applied potential energy curves of the presented model. Especially, for a

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Fig. 2.15: Spectrum of a Rydberg excitation in a Bose-Einstein condensate and the resulting line shapes calculated by equation 2.17, with (solid red) and without (dashed yellow) including the *p*-wave shape resonance, here for the 53*S* Rydberg state.

large red detuning $\delta \lesssim -50$ MHz only by considering the *p*-wave contribution a significant signal can be reproduced by the theoretical model. Therefore, the presence of a signal in this region for both, the measured as well as the simulated spectrum, indicates the importance of the *p*-wave shape resonance of the e^{-} -Rb(5S) scattering for the system at hand. Since, the presented model is based on a semi-classical approach it does not reproduce the discrete lines of the Rydberg molecules ($-6 \leq \delta < 0 \text{ MHz}$). Furthermore, the overall discrepancy for a small red detuning $(-20 \leq \delta < 0 \text{ MHz})$ is a possible indication that the correlation between the perturber atoms, which is neglected for the applied model, matters in this region. In the following the results for principle quantum numbers $n = \{40, 53, 71, 90, 111\}$ are depicted in figure 2.16. As one can clearly see from figure 2.16, even for higher principle quantum numbers, the signal for a large red detuning is only reproducible if the *p*-wave shape resonance is taken into account. In contrast, the calculations for s-wave scattering only and for s- and p-wave scattering show a more similar broadening for higher principle quantum numbers. Furthermore, the spectra displayed in figure 2.16reveal a strong *n*-dependence of the broadening of the spectral line shapes.



Fig. 2.16: Spectra of a Rydberg excitation in a Bose-Einstein condensate and the resulting line shapes calculated by equation 2.17, with (solid red) and without (dashed yellow) including the *p*-wave contribution.

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The volume of a nS Rydberg electron orbit scales with $V_{\rm Ryd} \propto r_{\rm Ryd}^3 \propto (n^*)^6$, therefore the number of total atoms N inside the electron orbit ranges from few to several thousands for the studied principle quantum numbers. Thus, compared to a high principle quantum number (n = 111), the standard deviation $\sqrt{N/N}$ is much larger for lower n, which leads to an additional broadening mechanism in contrast to high n. Moreover, the depth of the potential energies scale similar to the energy difference of adjacent energy levels ($\propto n^{-3}$ [28]). Regarding the broadening of the spectral line shapes, this means on average the energy shift from each perturber atom inside the Rydberg electron orbit decreases with an increasing principle quantum number, leading to a narrower spectrum for higher n. Therefore, it is not surprising that for increasing principle quantum numbers the broadening for both, s-wave scattering only and s- and *p*-wave scattering, exhibit a similar trend. For a high principle quantum number, the *p*-wave contribution leads to an overall red shift of the simulated line shape in addition to the wing on the red side of the overall peak in the spectra. As a consequence, the broadening of the spectrum depends strongly on the principle quantum number, however, the influence of the *p*-wave shape resonance on the additional broadening plays a subordinate role.

Finally, it is important to mention that, since all perturber atoms in the Rydberg orbit are treated by means of a pair-interaction with the Rydberg atom, the presented model is only valid as long the perturbation of each perturber atom on the Rydberg electron wave function is localized. This can happen if a perturber atom is placed in the butterfly state. However, the probability to excite such a configuration is strongly suppressed, due to the limited available energy given by the Rydberg excitation detuning, the low S-character far away from the addressed Rydberg state and the uncorrelated spatial atom distribution. Thus, for the investigated principle quantum numbers and densities, one can claim that the presented simulation method is self-consistent, because the restrictions of the model can not be exceeded by the created configurations based on the introduced method.



2.3.2 Variation of the excitation beam waist

Fig. 2.17: Density plot of the yz-plane of a simulated BEC with 1.4×10^{6} ⁸⁷Rb atoms surrounded by a thermal cloud with a Gaussian distribution of 0.3×10^{6} atoms and a temperature of $T_{\text{thermal}} = 300 \text{ nK}$, which is weighted with the spatial profile of the excitation beam. The simulated distributions are shown, from top to bottom for a beam waist of $w_0 = \{2.1, 4.2, 10.5\}\mu\text{m}$ of the focused infrared Rydberg excitation laser. The beam waist along the z-axis is displayed in red.

The developed microscopic model and the presented simulation method allows a variation of several experimental parameters. As an example, the influence of an increased beam waist is discussed in this section, whereas in the next section 2.3.3, different positions of the excitation laser focus in BEC are examined. As already mentioned the actual beam waist of the infrared excitation laser can be slightly different from the beam size measured in the test setup. Therefore in this section, the simulation is used to test the influence on the spectral line shape for a different beam waist of the infrared excitation laser. In figure 2.17, the result of the simulated atom distribution convoluted with the intensity profile of the infrared Gaussian beam for three different waists $w_0 = \{2.1, 4.2, 10.5\}\mu m$, is depicted. With an increased beam waist, the

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Fig. 2.18: Spectra of a Rydberg excitation in a Bose-Einstein condensate and the resulting line shapes calculated by equation 2.17, where the *p*-wave shape resonance is taken into account. Here for the 53S Rydberg state and three different beam waists.

region of possible Rydberg excitation extends over a larger volume inside the BEC. Therefore, more configurations are involved in the simulation. This can lead to a redistribution of the probability to find specific configurations in the sample and thereby to a different line shape of the spectrum. Especially, in comparison to a tightly focused Gaussian beam, for a large beam waist lower density configurations are more significant, since the probability to excite further away from the center of the BEC is enhanced (see figure 2.17). In fact, the results of the simulated line shapes, displayed in figure 2.18, show an enhanced contribution from lower density regions for an increased beam waist. This can be understood, as a lower density leads to a reduced number of perturber atoms inside the Rydberg electron orbit, which in turn leads to a reduced resulting energy shift. Therefore, the overall shift of the signal towards a smaller red detuning can be assigned to an increased contribution of lower density configurations. The simulated line shapes in figure 2.18 match the experimental data for a waist $w_0 = 10.5 \,\mu$ m very well. Even for a higher principle quantum

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Fig. 2.19: Spectra of a Rydberg excitation in a Bose-Einstein condensate and the resulting line shapes calculated by equation 2.17, where the *p*-wave shape resonance is taken into account. Here for the 90S Rydberg state and three different beam waists.

number, e.g. n = 90, the simulation match with the experimental data for the same beam waist (see figure 2.19) in a very convincing way. This suggests that the assumed minimal beam waist of the infrared Rydberg excitation laser is different in the actual experimental setup. However, it turns out in the following section, that the measured value of the beam waist ($w_0 = 2.1 \,\mu\text{m}$) is relatively close to the actual beam waist in the experimental.

2.3.3 Variation of the excitation position

In this section the simulation method is compared to the experimental results, which are presented and discussed in detail by Cubel Liebisch et al. [S3], for a different overlap of the tightly focused infrared excitation laser with the Bose-Einstein condensate. In order to probe different regions in the BEC, the infrared excitation beam is moved along the axis perpendicular to the propagation direction and the axial axis of the BEC (x-axis see figure 2.6). For each position



Fig. 2.20: Density plot of the xz-plane of a simulated spatial excitation distribution for a 90*S* Rydberg state in a BEC with 1.4×10^{6} ⁸⁷Rb atoms surrounded by a thermal cloud with a Gaussian distribution of 0.3×10^{6} atoms and a temperature of $T_{\text{thermal}} = 300 \text{ nK}$. From left to right the resulting distribution for a detuning of $\delta = \{-55, -24, -12\}$ MHz from the atomic resonance of the addressed Rydberg state is displayed. The beam waist along the z-axis is displayed in red.

the ion signal is evaluated for three different detunings from the atomic resonance of the addressed nS Rydberg state. With the microscopic model, introduced in section 2.3.1, it is clear that for different detunings the excitation laser is resonant with configurations of a specific region in the BEC. This means that, for a given detuning a specific density region is probed, which allows a mapping between detuning and density. In the following this detuning-density mapping is demonstrated using the developed simulation method.

Figure 2.20 shows the spatial distribution of possible Rydberg excitations simulated for a detuning of $\delta = \{-55, -24, -12\}$ MHz from the atomic resonance of the 90S Rydberg state, as a density plot of the xz-plane (elongated axis perpendicular to the drawing plane). The excitation distribution indicates that for a large red detuning the excitation probability is enhanced in the center and thus in a more dense region of the BEC. Whereas, for a small red detuning the Rydberg excitation probability is increased in a shell further away from the center of the BEC. As already predicted, it follows that for a specific detuning from the atomic resonance, a specific density region is addressed. Regarding the symmetry of the density distribution of the BEC, it is clear that different density shells are assigned by the corresponding detunings δ from the atomic resonance of the addressed Rydberg state.



Fig. 2.21: Density plot of the xz-plane of a simulated BEC with 1.4×10^{6} ⁸⁷Rb atoms surrounded by a thermal cloud with a Gaussian distribution of 0.3×10^{6} atoms and a temperature of $T_{\text{thermal}} = 300 \text{ nK}$, which is weighted with the spatial profile of the excitation beam. The resulting distributions are displayed from left to right with a focus position of $x_{\text{focus}} = \{0, 2.5, 5\} \mu \text{m}$ of the infrared Rydberg excitation laser, with respect to the center of the BEC. The beam waist along the z-axis is displayed in red. The maximum values of the projection (x-axis) on top are rescaled to one.

In order to probe the density distribution, one can measure the ion count for a certain detuning and move the tightly focused excitation laser beam with respect to the center of the BEC. In figure 2.21 a created atom distribution weighted with the spatial intensity profile of the infrared excitation laser, for a focus position of $x_{\text{focus}} = \{0, 2.5, 5\}\mu m$, is depicted. The tightly focused beam $(w_0 = 2.1 \,\mu m)$ cuts out a specific region of the created atom cloud. For a focus position close to the edge of the BEC (e.g. $x_{\text{focus}} = 5 \,\mu m$), the thermal cloud becomes apparent as a background offset. Whereas, for a focus position at the center of the BEC, the contribution of the thermal atoms is imperceptible. Additionally, the z-projection of the atom distribution shows the density profile of a BEC, whereas the x-projection mimics the Gaussian intensity profile of the excitation beam, for a focus position at the center. The tight focus of the infrared excitation laser allows one to measure the excitation probability of an nS Rydberg state for a specific detuning at different positions in the BEC and thus for different density regions.



Fig. 2.22: (a) Experimental results (dots) as well as the outcome of the simulation for a waist $w_0 = 2.1 \,\mu\text{m}$ (lines) and $w_0 = 3.0 \,\mu\text{m}$ (dashed lines), for a detuning of $\delta = \{-55, -24, -12\}$ MHz from the atomic resonance of the addressed 90S Rydberg state. (b) Shows the parabolic density profile of the BEC, where in (c) the different addressed density shells corresponding to the detuning δ are shown. The different colors in (b) and (c) are associated with the different detunings in (a).

In figure 2.22a the experimental results as well as the simulation is depicted for three different detunings, where the illustrations in (b) and (c) demonstrate the mapping from a laser detuning onto a density shell of the BEC. As one can clearly see from figure 2.22a the simulation and the experimental results do match very well, especially, for a small red detuning. However, for a large red detuning of $\delta = -55$ MHz, the flank of the extracted experimental data and simulated data (figure 2.22a lines) show a small discrepancy. By considering, that the simulated spectral line shapes suggest an underestimated beam waist (e.g. $w_0 = 4.2 \,\mu\text{m}$ in figure 2.18 and figure 2.19), one could argue that this small discrepancy is due to the same reason. Since, with a larger beam waist the signal from the corresponding density region is probed for larger distances of the focus from the center of the BEC, which leads to a broader curve in figure 2.22a. The effect of a slightly increased beam waist (e.g. $w_0 = 3 \,\mu\text{m}$) is depicted in figure 2.22a (dashed lines). The dip in the middle for smaller red detuning (green and blue curve) indicates a beam waist w_0 of about 2 - 3 µm, as the addressed density regions are well resolved for a focus position close to the center ($|x| \leq 3$ µm in figure 2.22a). The good agreement in figure 2.22a also demonstrates the validity of the detuning-density mapping for a large principle quantum number (n = 90). However, one has to note that the additional broadening mechanism of the spectra, for low principle quantum numbers, which is discussed at the end of section 2.3.1, has a strong influence on this detuning-density mapping. This becomes clear, by recalling that for low n the number of perturber atoms inside the Rydberg electron orbit is of the order of 10 and thus the standard deviation is rather large and leads to an enlarged interval of densities associated with the same detuning. A more detailed discussion of the detuning-density mapping can be found in [S3]. Overall, the good agreement of the experimental data and simulated data in figure 2.22a for the 90S Rydberg state shows again the validity of the developed microscopic model from section 2.3.1 for the system at hand and the investigated principle quantum numbers.

3

Simulation of Rydberg dynamics in a Bose-Einstein condensate

In the previous chapter the static properties of a Rydberg excitation in a dense ultracold gas were discussed. Whereas in this chapter, the resulting dynamics of a Rydberg atom immersed in a BEC is studied. It turns out, that the resulting collisional lifetimes are not only dominated by the ionic Rydberg core, but also are strongly affected by the scattering induced interaction of the Rydberg electron and the surrounding atomic cloud.

The experimental realization presented in section 2.2 not only allows to measure the spectral line shape of a Rydberg excitation in an ultracold and dense gas, but also enables one to study the dynamics of such a system. For this purpose, after the Rydberg excitation an additional delay time is implemented before the ionization pulse is applied. The collisional lifetime of a Rydberg atom can be extracted from the measured ion-signal for the different delay times, while the flight time of the detected ions can be used to obtain the kinetic energy released by the collision of a Rydberg atom and a neutral atom. Evaluating the collisional lifetime and energy release, one can conclude, that the collision leads to two reaction products, which are observed and studied by Schlagmüller et al. [S2]. In figure 3.1 the two corresponding reaction channels are depicted. The Rydberg atom is initially prepared in the (n, l) state and can react with the surrounding neutral atoms of the BEC, resulting in either a deeply bound molecular ion (Rb₂⁺) or an *l*-mixing reaction. For the latter one the Rydberg atom changes its internal state during the reaction mechanism. This is due to the



Fig. 3.1: The two observed reaction channels for a Rydberg atom (Rb^{*}) excited in an ultracold and dense environment. For the upper sketched reaction channel (1), the excited Rb^{*} atom and the neutral atom (Rb) form a deeply bound Rb⁺₂ ion due to chemi-ionization [37]. The lower channel (2) results in a *l*-mixing reaction, which leads to a change of the angular momentum of the excited Rydberg atom. For this reaction, the energy release of the collision is equally shared by both, the Rydberg atom and the neutral atom. The two reaction mechanisms exhibit two reaction times τ_1 and τ_2 .

presence of the *p*-wave shape resonance with a ${}^{3}P^{o}$ symmetry of the e^{-} -Rb(5S) scattering, which is introduced in section 2.1. The shape resonance gives rise to a butterfly state, which in turn couples to the trilobite state of the lower lying hydrogenic manifold and thus provides a possible path through the energy landscape for the observed final (n-4, l > 2) state of the *l*-mixing reaction [S2].

A simple theory to describe the collision of the ionic Rydberg core and a neutral atom is given by the Langevin model [38], which is based on the polarization potential discussed in section 2.1.1. In principle, the Langevin model describes the ability of the ion to capture the neutral atom due to the long-range polarization interaction potential as the ion and the atom move towards each other along classical trajectories. A theoretical study of ultracold atom-ion collisions for alkali atoms [13] reveals, that the semi-classical approach of the Langevin model is valid for a collision energy down to $E \sim k_{\rm B} \times 10$ nK. For the Rydberg electron-neutral atom scattering, one has to consider the orbital period of the electron $(T_n = n^3 1.5 \times 10^{-16} \text{ s} [39])$ and the relative motion of

the Rydberg atom and the neutral atom. The resulting collisional lifetimes indicates, that the dynamics of the system is on the order of few microseconds. By considering the orbital period $T_{n=150} \sim 500 \,\mathrm{ps}$, it becomes clear that the electron motion is much faster than the nuclear motion and thus the electron can slowly adjust its wave function by scattering several times at the nearly frozen neutral atom. Therefore, the nuclear motion and the electronic motion can be treaded separately, meaning that the Born-Oppenheimer approximation is valid. This suggest, that for both, the ion-atom interaction and the electronatom interaction, a classical dynamics simulation can be applied to model the collision times. However, with this approach, quantum dynamical effects such as quantum reflection are neglected. In order to simulate the collisional lifetimes, the microscopic model of section 2.3.1 is applied to create configurations of neutral atoms distributed inside the Rydberg electron orbit. The interaction potential for the dynamics simulation is modeled by taking into account only the ion-atom interaction in section 3.2 (polarization potential approach). Whereas in section 3.3, the electron-atom interaction potential is modeled using an average potential approach and both, the ion-atom interaction and the electron-atom interaction is taken into account for the dynamics. The resulting collision times of each potential approach are compared to the measured Rb_2^+ formation times and the *l*-mixing collision times for different principle quantum numbers.

3.1 Dynamics simulation method

Within the framework of the microscopic model (section 2.3.1), a method is presented, which allows to determine configurations of neutral atoms distributed inside the excited Rydberg atom, depending on the addressed nS Rydberg state and the detuning from the atomic resonance. For the dynamics simulation, the initial positioning of the neutral atoms inside the Rydberg orbit is obtained from these configurations. Furthermore, the excitation process and the subsequent time evolution of the system, are assumed to be independent of each other. This approximation is necessary, since the excitation process is not taken into account for the calculation of the applied potential energy curves. For a first rough estimate of the dynamics, one could argue to consider only the nearest neighbor atom of the excited Rydberg atom. However, by regarding the interaction range of the ionic Rydberg core interacting with the surrounding neutral atoms, it becomes clear that this simple approach is insufficient. With equation 2.15 the interaction range for the polarization potential of the ionic Rydberg core is

$$r^* = \sqrt{\mu\alpha} \approx 5000 \, a_0,\tag{3.1}$$

where the polarizability of the neutral atoms $\alpha = 318.8(14)$ a.u. [26] and the reduced mass $\mu = m_{\rm Rb}/2$ of two ⁸⁷Rb is used. This means, for the system at hand, with a peak density of up to $\rho \approx 5.2 \times 10^{14} \, {\rm atoms/cm^3}$, on average the ionic Rydberg core interacts simultaneously with more than 40 neutral atoms. Thus, for the dynamics simulation all neutral atoms in the interaction range are taken into account.

For the classical dynamics simulation the initial velocity of the neutral atoms is assumed to be zero, as for a single atom of the BEC no individual velocity is defined. Whereas, the initial velocity of the Rydberg atom is given by the photon recoil of the Rydberg excitation laser as: $E_{\text{recoil}} \approx h \times 15 \text{ kHz}$. Furthermore, a Bose-Einstein condensate can be considered as a dilute gas, if the scattering length of the neutral atoms is much smaller than the mean inter-particle distance [33]. This is the case for the system at hand, as the scattering length of the neutral ⁸⁷Rb-atoms ($a_{\rm Rb} = 103 a_0$ [35]) is much smaller than the mean inter-particle distance $(r_{\text{mean}} \approx 1500 a_0)$ for the peak density $\hat{\rho} = 5.2 \times 10^{14} \,\mathrm{cm}^{-3}$ of the BEC. In turn, the interaction between the neutral atoms of a dilute gas is dominated by the long-range tail of the dipole-dipole interaction, which is more commonly known as the van der Waals interaction [33]. By regarding equation 2.14, using the van der Waals interaction (C_6/R^6) instead of the polarization potential it follows that the interaction range for the van der Waals interaction is: $r_{\rm vdW}^* = (\mu C_6)^{1/4} \approx 140 a_0$, with the reduced mass $\mu = m_{\rm Rb}/2$ of two ⁸⁷Rb and the C₆-coefficient $C_6 = 4691$ a.u. calculated by Derevianko et al. [40]. Therefore, the mean inter-particle distance is much larger than the range of the van der Waals interaction: $r_{\text{mean}} \gg r_{\text{vdW}}$. Consequently, the neutral atoms are sufficiently separated and one can neglect the neutralneutral interaction for the dynamics simulation. This applies at least until the first collision of the Rydberg atom with a neutral atom occurs, meaning that the internuclear separation of the Rydberg core and the neutral atom falls below $10 a_0$ and the atoms repel each other due to the Pauli exclusion principle

(see section 2.1.1). After the first collision, the initial photon recoil is shared between the collision partners and the collided neutral atom can scatter with another neutral atom. However, a simple estimation of a geometric cross-section ($\sigma_{\text{geo}} = \pi a_{\text{Rb}}^2 \approx 100 \,\text{nm}^2$), using the scattering length of ⁸⁷Rb, indicates that this is improbable for the system at hand, as the peak density of the BEC leads to a mean free path of about 20 µm.

The classical dynamics simulation for the system of a Rydberg atom interacting with the surrounding neutral atoms is realized with a symplectic leapfrog integration method, which can be found in every standard text book about molecular dynamics simulation (e.g. in [41]). For each simulation the energy conservation and the angular momentum conservation as well as the center of mass motion of the system was reviewed. Since the aim of the dynamics simulation is to estimate the time that elapses until the first collision occurs, the simulation can be stopped shortly before the actual collision happens, in order to avoid unnecessarily small integration time steps.

In the following sections (3.2 and 3.3) the results of two different approaches are presented and compared to the collision times observed by Schlagmüller et al. [S2]. For a first approximation of the collision times, one must at least consider the interaction between the ionic Rydberg core and the surrounding neutral atoms. The results of this estimate are shown and discussed in the following section 3.2. However, for this first estimation, the interaction of the Rydberg electron and the neutral atoms (perturber atoms) inside its electron orbit, is neglected. Especially, the avoided crossing of the potential energies of the butterfly state and the addressed nS Rydberg state, which is necessary to explain the observed l-mixing collision [S2], is not considered. In order to take into account the coupling of the butterfly state with the addressed nS Rydberg state, the electron-atom interaction is also included in section 3.3 by using the two-body potential energies, which are introduced in section 2.1 and afterwards discussed in section 2.3.1. However, for the dynamics simulation the neutral atoms are considered as point like particles with zero initial momentum and thus, by taking into account the full potential energy curve, this results in a non-physical quasi bound state, as most of the perturber atoms are classically trapped in the oscillatory shape of the potential. In order to avoid this nonphysical effect, for the dynamics simulation, the PECs are radially averaged such that the oscillatory shape vanishes. This means, for the electron-atom interaction an instantaneous tunneling of the perturber atoms through the potential wells is assumed.



Fig. 3.2: (a) Histogram of the nearest neighbor distribution for a detuning of $\delta = -48$ MHz from the atomic resonance of the 50S Rydberg state (blue), as well as the nearest neighbor distribution for a constant density corresponding to the addressed density (red). (b) Potential energy curves over the internuclear distance R, which are relevant for the dynamics simulation in section 3.2 and 3.3.

Figure 3.2b shows the polarization potential of the ionic core of a Rydberg atom (orange), as well as the two-body potential energy curve for the 50S + 5Sstate (blue) and the corresponding average potential (red). The average potential is obtained from the 50S + 5S potential by using a cubic spline function through the inflection points with a negative slope, while the outer most potential well stays unchanged. For both electron-atom interaction potentials, the polarization potential is added. The same procedure is applied for all average potentials using the corresponding nS + 5S two-body potential energy curve. As one can see from figure 3.2b, the polarization potential takes over for an internuclear distance smaller than $\sim 500 a_0$. In (a) the nearest neighbor distribution of the corresponding simulated configurations for a detuning of $\delta = -48$ MHz (50S Rydberg state), is depicted. The nearest neighbor distribution reflects the influence of the crossing between the butterfly state and the addressed Rydberg state. Since, the configurations are chosen with respect to the detuning from the atomic resonance, the probability to observe a perturber atom in the crossing region is reduced. Especially, for a negative detuning the probability to find a perturber atom in the upper branch is suppressed. This becomes clear, by comparing the nearest neighbor distribution for a constant density, which corresponds to an energy mean shift according to equation 2.6, in figure 3.2a. The simulated distribution shows an enhanced probability close to the crossing in the lower branch, whereas the probability in the upper branch is reduced next to the crossing. Far away from the crossing, the simulated nearest neighbor distribution and the constant density distribution are equivalent. Since the back-action of a single perturber atom on the Rydberg electron wave function is neglected, the dynamics simulation of the average potential approach is stopped when the first perturber atom enters the region of the avoided crossing. Specifically, this means that the simulation ends, once a perturber atom enters the region, where the S-character of the lower potential branch falls below 0.5. With this, the potential resulting from the electron-atom interaction can be treated as spherical symmetric, since the perturbation of the electron wave function is small and thus the arising anisotropic shape of the butterfly state can be neglected. This stop criteria leads to a systematic offset for the determined collision times. However, the increase of momentum of the neutral atoms, which are subject to a collision, is large enough to neglect this offset. For the system at hand, the apparent systematic offset is on the order of few nanoseconds. Nevertheless, the resulting collision time of each dynamics simulation has been checked to be unaffected by this stop criteria. The lower potential branch denotes the outer part of the potential, down to the crossing of the butterfly state, which is at about $R \approx 1400 a_0$ in figure 3.2b. For the upper potential branch, denoting the inner part of the potential, the dynamics simulation ends for an internuclear distance $R < 100 a_0$. The second condition is also applied for the polarization potential approach in section 3.2. The probability to find a perturber atom either close to the ionic Rydberg core $(R < 100 a_0)$ or close to the center of the crossing is suppressed, due to the energy limitation given by the detuning from the atomic resonance. This is also apparent in figure 3.2a, where the nearest neighbor distribution of the calculated configurations shows a dip near the avoided crossing $(R \approx 1400 a_0)$. Finding a perturber atom initially placed deep in the butterfly potential is most likely for a large red detuning.

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Fig. 3.3: Minimal S-character of the perturber atoms inside the Rydberg orbit for a detuning of $\delta = -48$ MHz from the atomic resonance of the addressed nS Rydberg state.

In figure 3.3 the minimal S-character of the perturber atoms of 5000 possible Rydberg excitations, for a detuning of $\delta = -48$ MHz, is shown. By considering the minimal S-character, depicted in figure 3.3, and the mean number of perturber atoms inside the *p*-wave shell (see figure 2.10 in section 2.3.1), it follows that, not only the mean number of perturber atoms close to the crossing is below one, but also that the minimal S-character of the initially occupied state is on average much larger than 0.5. Thus, the number of configurations, which lead to an immediate collision due to the stop criteria of a present S-character below 0.5, is negligibly low. In the following sections 3.2 and 3.3 the cumulative collision probability is used to define the lifetime τ for different principle quantum numbers and detunings from atomic resonance of the addressed nS Rydberg state. For each obtained lifetime more than 1000 trajectories are calculated for the Rydberg atom-neutral atom system. In turn, for each trajectory the initial distribution of neutral atoms inside the Rydberg orbit is randomly picked out of a larger sample of the simulated configurations for the corresponding detuning of the addressed nS Rydberg state.

3.2 Polarization potential approach

For the system at hand, the interaction range of the polarization potential is larger than the mean inter-particle distance. Consequently, for a first estimate of the collision time, only the interaction of the ionic Rydberg core with the surrounding neutral atoms, is considered for the classical dynamics simulation, which is displayed in figure 3.2b (orange). This means, in particular, that the electron-atom interaction between the Rydberg electron and a neutral atom is neglected for the simulated time evolution of the system. However, for the initial positioning of the neutral atoms and the single ionic Rydberg core, the atom distributions based on the microscopic model are utilized. Therefore, it is expected that the resulting collision times depend on the detuning from the atomic resonance of the addressed nS Rydberg state.



Fig. 3.4: Collision probability and the resulting cumulative collision probability, obtained from the classical dynamics simulation of 1000 different starting positions for a detuning of $\delta = -48$ MHz from the atomic resonance of the addressed 60S Rydberg state. The cumulative collision probability is modeled with $p_{\rm csum}(t) = (1 - e^{-t/\tau})$ in order to obtain the lifetime τ of the Rydberg atom excited in the BEC.

As an exemplary evaluation, the normalized collision probability and the resulting cumulative collision probability of 1000 simulated collision times, for a 60S Rydberg atom excited with a detuning of $\delta = -48$ MHz, is depicted in figure 3.4. The cumulative collision probability is calculated by the cumulative sum of the time-dependent collision probability and gives the probability of a

collision after the system has evolved a certain time t. In order to obtain a typical lifetime τ of the Rydberg atom for a specific detuning δ and a certain principle quantum number n, the cumulative collision probability of the simulated collision times is modeled with $p_{\text{csum}}(t) = (1 - e^{-t/\tau})$. This means, each obtained lifetime corresponds to an evolved time $t = \tau$, for which the probability of collision is $1 - 1/e \approx 0.63$.

In figure 3.4 a step is apparent in the cumulative collision probability at the time $t = 0.8 \,\mu\text{s}$. This step has its origin in the simulated initial neutral atom distributions. The reduced probability to excite a Rydberg atom with a neutral atom close to the avoided crossing of the butterfly state and the nS Rydberg state leads to a dip in the nearest neighbor distribution of the simulated neutral atom distributions. Since, the polarization potential is monotonically decreasing for a decreasing internuclear distance, in most cases the Rydberg atom will collide with the nearest neighbor of the initial atom distribution. In some cases the photon recoil can lead to a collision with the next nearest neighbor. However, the latter case is very unlikely, as the displacement of the Rydberg atom due to photon recoil is negligible. Consequently, the step occurs due to a lowered probability of a Rydberg atom excited with a neutral atom close to the avoided crossing of the butterfly state and the addressed Rydberg state. For the following evaluation, all the obtained lifetimes were checked to be mainly unaffected by this step (see appendix A.1).

In figure 3.5 the obtained lifetimes of the classical dynamics simulation, using the polarization potential approach, are compared with the experimentally determined *l*-changing collision time and the Rb₂⁺ formation time measured by Schlagmüller et al. [S2] in a double logarithmic plot. The results of the classical dynamics simulation are slightly above the experimentally obtained lifetimes of the initially prepared Rydberg state for a principle quantum number of $n \leq 100$. For $n \geq 100$ the observed *l*-changing collision times are largely increased. So far the reason of the increased *l*-changing collision time is unexplained. For the estimated lifetimes of the classical dynamics simulation no threshold behavior is observed. Thus, one can conclude, that the experimentally observed threshold is not a direct consequence of the detuning selected neutral atom distributions. The resulting lifetimes depend not only on the detuning, but also on the principle quantum number n. This is not surprising, since the starting configurations used for the dynamics simulation depend on both, the detuning and the principle quantum number n.



Fig. 3.5: Results of the classical dynamics simulation using the polarization potential (lines), as well as the *l*-changing collision time and the Rb_2^+ formation time measured by Schlagmüller et al. [S2] (triangles) for three different detunings from the addressed nS Rydberg state. The cumulative collision probability as well as the corresponding fit for each data point of the simulation is shown in section A.1.

However, to explain the observed l-changing collision, the coupling of the Rydberg S-state to the butterfly state is essential [S2]. Therefore in the following section, the electron-atom interaction of the Rydberg electron and the neutral atoms is also taken into account.

3.3 Average potential approach

The observed *l*-changing collision [S2] as well as the specific spectral line shape [S1] reveals the importance of the *p*-wave shape resonance for a Rydberg excitation in a ⁸⁷Rb Bose-Einstein condensate. In order to take into account the *p*-wave contribution of the electron-atom scattering, in addition to the interaction of the ionic Rydberg core with the neutral atoms, the interaction of

the Rydberg electron with the neutral atoms inside the Rydberg orbit (perturber atoms), is considered. However, for a proper classical dynamics simulation, an average potential of the actual potential energy curve (PEC) is used. In figure 3.2 the PEC and the corresponding average potential is depicted. The introduction as well as the motivation of the used average potentials is given in section 3.1. Basically, an average of the oscillatory shape of the PEC is applied with the aim to avoid a non-physical trapping of the perturber atoms in the potential wells. The purpose of this section is the presentation of the results of the classical dynamics simulation for the average potential approach, as well as the comparison with the results of the previous section and the measured collision times [S2]. By using the average potential, it is expected that the resulting lifetimes are reduced in comparison to the simulated collision times of the polarization potential approach and to the experimentally observed collision times. This is the case, as on the one hand the polarization potential is applied in both cases and on the other hand the average potential approach is equivalent to an instantaneous tunneling of the perturber atoms through the oscillatory shape of the actual PEC towards the ionic Rydberg core. Consequently, the simulated dynamics of the system is accelerated and thus the resulting lifetimes of the simulation can be understood as a lower limit for a Rydberg atom in the Bose-Einstein condensate. In general, the configurations of neutral atoms inside the Rydberg orbit are determined by detuning and not by density (see chapter 2). As the mean energy shift for a high density is increased due to a large number of perturber atoms inside the Rydberg orbit, for a large red detuning the probability is enhanced to excite a Rydberg atom in a region of high density. Rarely, even for a small red detuning a configuration in the high density region of the BEC can be addressed. Since, this configuration is unlikely to occur, the probability to excite a Rydberg atom in a high density region with a small red detuning is reduced. Thus, for a given detuning, the Rydberg excitation is more probable in a specific density region of the BEC. However, this specific region can be quite large. Especially for the system at hand, for a detuning of $\delta \gtrsim -6$ MHz the addressed density region extends over the complete illuminated area of the focused excitation laser in the BEC. For a small red detuning the Rydberg excitation at the edge of the BEC is more probable. However, the excitation in the center is increased due to the tightly focused excitation laser. Thus, for a small red detuning $\delta \rightarrow 0$, the probability to excite in the focus and to excite in a low density region becomes equivalent and as a result the density shell approach (see section 2.3.3) is no longer valid.



In this case, different lifetimes of a Rydberg atom can occur simultaneously in the system.

Fig. 3.6: Collision probability and the resulting cumulative collision probability, obtained from the classical dynamics simulation of 1000 different starting positions for a detuning of $\delta = \{-6, -48\}$ MHz from the atomic resonance of the addressed 60S Rydberg state. The cumulative collision probability is modeled with $p_{\text{csum}}(t) = (1 - e^{-t/\tau})$ in order to obtain the lifetime τ of the Rydberg atom excited in the BEC.

The collision probability for a detuning of $\delta = -6$ MHz (60S Rydberg state), depicted in figure 3.6, reflects that the dynamic of the system is dominated by two lifetimes. The Rydberg atoms excited in the center of the BEC lead to a fast time scale, whereas the Rydberg atoms close to the edge of the BEC provide a slow time scale for the dynamics of the system. This is also reflected by the fraction of the neutral atoms, which are subjected to a collision, starting in the inner region ($R < R_{crossing}$) of the averaged potential. In the case of a Rydberg excitation with a low red detuning in a high density region, only few neutral atoms in the inner region of the potential have to compensate the large mean shift of the neutral atoms in the outer region of the potential. Thus,

for these specific configurations the probability of neutral atoms close to the avoided crossing in the inner potential is increased. For the dynamics simulation, this results in an enhanced probability of colliding neutral atoms starting in the upper branch of the potential due to the steep gradient in this region (see figure 3.2 800 $a_0 < R < 1400 a_0$). A comparison of the fraction of inner neutral atoms and outer neutral atoms for the collision probability is depicted in figure 3.6, for a large red detuning (-48 MHz) and a small red detuning (-6 MHz). As predicted, for the case of a small red detuning, the fraction of the colliding neutral atoms, starting in the inner potential region, is increased in comparison to a large red detuning. At this point, it should be noted, that the step in the cumulative collision probability in figure 3.4 (polarization potential approach) occurs due to the reduced probability of perturber atoms close to the avoided crossing, whereas the step in figure 3.6 (average potential approach) for $\delta = -6$ MHz mainly results from addressing different density regions. As is apparent from figure 3.6, the modeling with $p_{\rm csum}(t) = (1 - e^{-t/\tau})$ of the cumulative collision probability is insufficient for a small red detuning, since two different time scales occur in the system. In general, for the examined principle quantum numbers, no lifetimes are extracted for a detuning of $\delta \gtrsim -10 \,\mathrm{MHz}$ due to the appearance of different time scales.

Figure 3.7 shows the resulting lifetimes of the polarization potential approach (lines) of the previous section and the average potential approach (dashed lines) of the current section, as well as the experimentally observed *l*-changing collision times and the Rb_2^+ formation times, observed by Schlagmüller et al. [S2] for different principle quantum numbers. As predicted, the average potential approach leads to a shorter lifetime in comparison to the polarization potential approach and the experimentally observed formation and collision times. From figure 3.7, it is apparent, that the polarization potential approach provide a fairly good estimation. However, for this approach the electron-atom interaction is neglected for the dynamics of the system, which in turn is essential to explain the observed l-changing collision times [S2]. In general, the simulated nearest neighbor distributions show an enhanced or reduced probability close to the Rydberg core, depending on whether the crossing position is closer or further away in comparison to the peak of the nearest neighbor distribution of the corresponding constant density (see figure 3.2b). This results in an enhanced or reduced lifetime of the simulated collisions for the polarization potential approach, as the neutral atom, which is subject to the first collision, is equal to the nearest neighbor.



Fig. 3.7: Results of the classical dynamics simulation using the polarization potential (lines) and the average potential approach (dashed lines), as well as the *l*-changing collision time and the Rb_2^+ formation time measured by Schlagmüller et al. [S2] (triangles) for three different detunings from the addressed *nS* Rydberg state. The cumulative collision probability as well as the corresponding fit for each data point of the simulation is shown in section A.1.

Thus, the simulated lifetimes of the polarization potential approach in figure 3.7, show an enhanced lifetime for an increasing principle quantum number for a small red detuning. For a large red detuning ($\delta = -48$ MHz) the crossing position is closer to the Rydberg core than the peak of the nearest neighbor distribution for a constant density, which results in a faster collision for smaller principle quantum numbers ($n \leq 70$). A comparison of the simulated lifetimes of the polarization potential approach and the average potential approach, reveals a strong influence of the avoided crossing of the butterfly state and the addressed

Chapter 3 Simulation of Rydberg dynamics in a Bose-Einstein condensate

Rydberg state on the dynamics of the simulated system. Especially, the latter approach leads to lifetimes, which show the same trend as the experimentally observed collision and formation times for principle quantum numbers $n \leq 100$ (see figure 3.7).

To conclude, the threshold behavior of the *l*-changing collision time indicates the importance of a quantum mechanical treatment of the dynamics. In fact, two main effects are not covered by this semi-classical approach of the dynamic of the system, which can increase the collision time significantly and withal depend on the principle quantum number. On the one hand, the finite tunneling rate for the actual oscillatory shape of the potential energy curve slows down the dynamics of the system in comparison to the assumption of an instantaneous tunneling in the average potential approach. On the other hand, for a steep potential, as it is the case close to the avoided crossing, quantum reflection can arise due to the wave nature of the ultracold neutral atoms, leading to an increased lifetime of a Rydberg atom in a Bose-Einstein condensate. Both effects can be covered with a quantum dynamics simulation. However, for a system of many neutral atoms coupled through a Rydberg atom, this can be extremely complex and computationally intensive. The realization and discussion of such a simulation is beyond the scope of this work, but a further investigation, using e.g. a full wave packet simulation, can give a deeper understanding of the threshold phenomena, which in turn could be important for future studies on the system under consideration.

4

Conclusion and Outlook

Within the framework of this master thesis, a microscopic model has been developed to simulate the experimentally observed spectral line shapes of a single Rydberg atom excited in a ⁸⁷Rb Bose-Einstein condensate. The simulation reveals a strong dependence on the calculated potential energy curves, in particular, the PECs neglecting the *p*-wave contribution, describe the overall line shape of the measured spectra, but by including the *p*-wave contribution the resulting line shapes are improved significantly. The simulated spectra, with and without the *p*-wave contribution, in comparison to the measured spectra, gives evidence for the butterfly state coupling to the addressed Rydberg state. Prior methods [8-12] were only able to predict the broadening and an overall line shift for different principal quantum numbers. The presented simulation method, based on the microscopic model, gives access to the full spectral line shape and can be used to test predicted potential energy curves. Furthermore, with the developed model, the influence of an increased beam waist and a different focus position of the excitation lasers has been tested against experimental results. The former one suggests an enlarged overlap of the spatial excitation beam profile with the BEC, but the latter limits the beam waist of the focused excitation laser to $w_0 = 2 - 3 \,\mu\text{m}$, which is in good agreement with the measured beam waist [36]. Moreover, the developed model gives the perspective to predict either the density profile for a given spatial intensity distribution of the excitation laser or by knowledge of the density profile, the opportunity to review the beam waist and thus can be used as a theoretical model for the density probe method presented by Cubel Liebisch et al. [S3].

In the second part of this master thesis, the dynamics of the presented system has been reviewed using a semi-classical approach, which is expected to be appropriate for the collision of the ionic Rydberg core and the surrounding neutral atoms [13]. Regarding the experimentally observed reaction channels [S2], it turns out that the collisional lifetime of a Rydberg atom, excited in an ultracold and dense environment, is not only dominated by the ionic Rydberg core-neutral atom interaction, but also is strongly affected by the scattering induced interaction of the Rydberg electron and the surrounding atomic cloud. The experimentally observed collisional lifetimes and the here presented results of the dynamics simulation, indicates that a quantum mechanical treatment is necessary in order to describe the measured threshold behavior of the collisional lifetimes for larger principle quantum numbers $(n \gtrsim 100)$ [S2], which in turn could be important for future studies. A promising application of the presented system is an imaging technique, which can be used to image the Rydberg electron wave function [23]. The scattering induced attractive interaction of the Rydberg electron results in an atom flow into the scattering potential and thus the wave function gets imprinted onto the atom distribution of the surrounding atomic cloud. However, for this technique, a sufficiently long lifetime of the Rydberg atom in the quantum gas is necessary for an observable impact and thus the enhanced lifetime for larger principle quantum numbers [S2] is very promising. Furthermore, the presented study of the dynamics can be seen as a motivation for a theoretical work about quantum reflection, which has been already observed for excited states of Rydberg molecules [16], and to not only describe the static properties [4], but also the resulting dynamical properties of this system, using a theoretical tool-set for many-body physics.

A

Classical dynamics simulation

A.1 Curve fitting of collisional lifetimes

In this section the cumulative collision probability, which is obtained from the classical dynamics simulation, using either the polarization potential approach or the average potential approach, of 1000 different starting positions for a detuning of $\delta = \{-48, -24 - 12\}$ MHz from the atomic resonance of the addressed nS Rydberg state, are shown. The cumulative collision probability of each set of trajectories is modeled with $p_{\text{csum}}(t) = (1 - e^{-t/\tau})$ in order to obtain the lifetime τ of the Rydberg atom excited in the BEC. The results of the obtained lifetimes are presented in section 3.2 (figure 3.5) and 3.3 (figure 3.7).



Fig. A.1: Cumulative collision probability, obtained from the classical dynamics simulation (polarization potential approach) of 1000 different starting positions for a detuning of $\delta = -48$ MHz from the atomic resonance of the addressed nS Rydberg state. The cumulative collision probability is modeled with $p_{\text{csum}}(t) = (1 - e^{-t/\tau})$ in order to obtain the lifetime τ of the Rydberg atom excited in the BEC.



Fig. A.2: Cumulative collision probability, obtained from the classical dynamics simulation (polarization potential approach) of 1000 different starting positions for a detuning of $\delta = -24$ MHz from the atomic resonance of the addressed nS Rydberg state. The cumulative collision probability is modeled with $p_{\text{csum}}(t) = (1 - e^{-t/\tau})$ in order to obtain the lifetime τ of the Rydberg atom excited in the BEC.



Fig. A.3: Cumulative collision probability, obtained from the classical dynamics simulation (polarization potential approach) of 1000 different starting positions for a detuning of $\delta = -12$ MHz from the atomic resonance of the addressed nS Rydberg state. The cumulative collision probability is modeled with $p_{\text{csum}}(t) = (1 - e^{-t/\tau})$ in order to obtain the lifetime τ of the Rydberg atom excited in the BEC.


Fig. A.4: Cumulative collision probability, obtained from the classical dynamics simulation (average potential approach) of 1000 different starting positions for a detuning of $\delta = -48$ MHz from the atomic resonance of the addressed nS Rydberg state. The cumulative collision probability is modeled with $p_{\rm csum}(t) = (1 - e^{-t/\tau})$ in order to obtain the lifetime τ of the Rydberg atom excited in the BEC.



Fig. A.5: Cumulative collision probability, obtained from the classical dynamics simulation (average potential approach) of 1000 different starting positions for a detuning of $\delta = -24$ MHz from the atomic resonance of the addressed nS Rydberg state. The cumulative collision probability is modeled with $p_{\rm csum}(t) = (1 - e^{-t/\tau})$ in order to obtain the lifetime τ of the Rydberg atom excited in the BEC.



Fig. A.6: Cumulative collision probability, obtained from the classical dynamics simulation (average potential approach) of 1000 different starting positions for a detuning of $\delta = -12$ MHz from the atomic resonance of the addressed nS Rydberg state. The cumulative collision probability is modeled with $p_{\rm csum}(t) = (1 - e^{-t/\tau})$ in order to obtain the lifetime τ of the Rydberg atom excited in the BEC.

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