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*Mixtures, quantum gases and molecules*
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Ultracold strontium and rubidium: mixtures, quantum gases and molecules

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Chapter 1

Introduction

Twenty-two years ago efforts in achieving ever colder temperatures in atomic vapours resulted in the creation of the first Bose-Einstein condensates [1, 2]. Ultracold degenerate gases became a major focus in atomic physics. Since then the research of Bose-Einstein condensates, and later degenerate Fermi gases [3], broadened our understanding of quantum phenomena. Prominent examples of these phenomena, such as formation of vortices in rotating condensates [4], the superfluid to Mott-insulator transition [5] or BEC-BCS crossover in Fermi gases [6], all rely on the supreme degree of control the experimenter has over the system. This high degree of control became evident already before the quantum degenerate regime was reached. With the help of electromagnetic fields the motional and internal degrees of freedom of atoms could be controlled precisely, especially after the development of the laser. Cooling and trapping vapours of alkali atoms allows for long interrogation times for spectroscopic measurements, which is utilized in the construction of ever more precise atomic clocks [7] or sensors of minute forces [8]. High-precision spectroscopy is also important for fundamental research: measuring the change of fundamental constants in the laboratory supports astronomical measurements in testing fundamental theories [9].

Once quantum degeneracy in atomic vapours was achieved, the technological advances and new discoveries in the field extended the ability to control the quantum matter. Firstly, the trapping potentials that are generated by optical or magnetic fields can be shaped almost arbitrarily to create low-dimensional quantum phases, even entrap the atoms in artificial light crystals resembling condensed matter systems. Secondly, the interaction between the atoms can be controlled as well. Magnetically tunable Feshbach resonances are used to change the scattering properties of the atoms across a wide range [10]. Usually, the scattering properties can be described by simple head-on collisions with a short-range, isotropic interaction potential. A different type
of interaction can be realized by using Rydberg atoms [11], atoms with a relatively large intrinsic magnetic moment [12], such as Cr, Er and Dy [13, 14, 15] or by creating samples of ultracold polar molecules [16]. In general ultracold systems are ideal platforms to study quantum physics by observing new phenomena with a supreme precision and control as well as to simulate other quantum systems.

Quantum simulation. Simulation is a way to obtain knowledge about a system that is to some degree inaccessible. Jupiter’s Great Red Spot for example is accessible through astronomic and spacecraft observation on the one hand. On the other hand the turbulent motion of the atmosphere can be simulated in a rotating tank [17]. This laboratory experiment was designed in such a way that the liquid in the tank and the atmosphere of Jupiter were subject to very similar laws. Measurements of the turbulent motion in the tank provide knowledge about the dynamics in the Great Red Spot.

In order to gain knowledge about a quantum system that is hard to access, one engineers a quantum simulator that mimicks this system [18, 19]. For a successful simulation the Hamiltonian describing the quantum simulator should be designed to abstract the Hamiltonian of the original system. Then precise measurements on the quantum simulator provide answers to how the original quantum system functions. The inaccessibility of quantum systems often lies in the fact that the Hilbert space of many-body systems grows exponentially with particle number. Numerical computation or simulation on a conventional computer is fundamentally impossible even for a relatively small number of particles. For example the numerical integration of the Schrödinger equation of an ensemble of 100 interacting spin-1/2 particles requires to store about $1.26 \times 10^{30}$ complex numbers. Assuming that the price of a 1 TB-harddrive is 50 € and one complex number is represented as a 64-bit number, the cost for the storage space alone would exceed the gross world product by four orders of magnitude [20]. Although numerical algorithms exist that treat the exponential complexity by stochastically sampling the vast Hilbert space, in many cases they fail because of the so-called sign-problem [21, 22].

It is the high degree of control and the long coherence times that make ultracold atoms a promising platform for quantum simulation of systems that are intractable on conventional computers. Firstly, the supreme control over ultracold atoms allows to prepare samples with bosonic or fermionic statistics at variable temperature and in defined quantum states. That way the quantum simulation can be initialized precisely. Secondly, the control ability ensures that the engineered simulator’s Hamiltonian can
be mapped onto the original quantum problem at hand. Thirdly, the long coherence
times allow long evolution times of the simulated Hamiltonian as well as a precise
measurement of the outcome of the simulation. As mentioned before usually ultracold
atoms interact via an isotropic, short-range contact potential. The interaction between
atoms is described by a single parameter, the scattering length. The complexity of the
system can often be reduced to a single-particle problem by mean-field theory. Emer-
gent many-body phenomena were observed in ultracold atom systems where inter-
actions were tuned using Feshbach resonances [6]. Also optical lattices can be used
to create systems in which interactions dominate the behaviour and thereby exhibit
properties beyond single-particle effects [23]. However, introducing long-range inter-
actions greatly increases the spectrum of intriguing many-body phenomena we could
study [24]. Challenges that require quantum simulation often involve particles inter-
acting with each other on a large length scale, such as high-$T_c$ superconductor exhibit-
ing strong electron correlations or quantum magnetism showing correlations in the
spin degree of freedom. Long-range interactions in ultracold matter can be introduced
by furnishing the particles with a dipole moment. Researchers have devised a few
approaches to ultracold matter interacting via long-range dipole-dipole interactions,
such as Rydberg atoms, magnetic atoms and heteronuclear ground-state molecules.
Rydberg atoms are highly excited and decay spontaneously on the ms-timescale [25].
They possess a very large dipole moment compared to molecules and magnetic atoms.
Magnetic atoms have a smaller dipole moment than molecules. Molecules are long-
lived and have a more complex structure than atoms. They possess vibrational and
rotational levels. Heteronuclear diatomic molecules in the ground state possess a large
electric dipole moment, which can lead to long-range dipole-dipole interactions by the
presence of an external electric field. Combining the ability to control ultracold atoms
with the unique properties of molecules and creating quantum matter of ground-state
molecules remains a long-standing goal in our field. The intermediate goal of our ex-
periment is the creation of polar RbSr molecules. In this thesis I will present several
steps necessary to create the molecules from ultracold Rb and Sr atomic vapours. If
we create a quantum gas of polar molecules with long-range interactions we would
be able to research novel quantum phases, such as crystalline structures, topological
phases, checker-board phases and supersolids [26, 27, 28].
1.1 Ultracold heteronuclear ground-state molecules

The long-range interactions between heteronuclear ground-state molecules are brought about by the electric dipole moment of the molecules. The electron cloud of the diatomic molecule is distributed unevenly across the two different atoms. The interaction potential decreases with $1/r^3$, much slower than the van der Waals $1/r^6$ potential of neutral atoms, and is spatially anisotropic. The interaction between particles with a dipole moment can be characterized by the dipole length $l_D = M\mu^2/\hbar^2$ [29], with $M$ being the reduced mass of the molecule and $\mu$ its dipole moment. Alkali atoms have a magnetic dipole moment of $1\mu_B$. The dipole length for an alkali atom is on the order of one Bohr radius ($a_0$), which is smaller than the typical scattering length ($\sim 100 a_0$ in $^{87}$Rb). In ultracold gases of alkali atoms the $s$-wave interaction dominates [30]. In order to observe significant long-range interactions between the particles the dipole length must be comparable to or larger than the interparticle distance. The interatomic distance in a quantum gas of density $n$ is on the order of $n^{-1/3} = 5000$ to $20000 a_0$, orders of magnitude higher than the dipole length of alkali atoms.

Atomic species, such as Cr, Er, and Dy, that have a high magnetic moment compared to alkali atoms have been brought to quantum degeneracy [13, 31, 14, 32]. Their dipole moment ranges from 6 to $10 \mu_B$ corresponding to a dipole length between 23 and $230 a_0$. Dipole-dipole interactions and their anisotropic nature were observed in dipolar Bose- and Fermi-gases [33, 34, 35]. Recently the quantum analogon to the Rosenzweig instability [36] was observed in a Dy BEC [37]. The investigation of magnetic quantum gases remains an active field of research.

Much stronger dipole moments can be achieved with ultracold, heteronuclear, ground-state molecules. The dipole length is in the range of $10^4$ to $10^6 a_0$, which is larger than the interparticle distance in quantum gases. Experimental efforts in creating high phase-space density samples of polar molecules are already in pursuit. The first ultracold sample of polar molecules consisted of the bi-alkali molecule KRb [38]. This molecule has a dipole moment of $0.566 \text{D}$ corresponding to a dipolar length of $l_D = 6000 a_0$. Dipolar spin-exchange interactions between the molecules in an optical lattice as well as many-body dynamics have been observed [39, 40]. Between 2008 and now, three additional heteronuclear, bi-alkali ground-state molecule species have been produced at ultracold temperatures, RbCs [41, 42], NaK [43] and NaRb [44]. Bi-alkali molecules in the absolute ground state have no magnetic dipole moment. The two
1.1. *Ultracold heteronuclear ground-state molecules*

electrons of the alkali atoms pair up in a singlet configuration, when the molecule is in the ro-vibronic ground state. Molecules comprised of an alkali and an alkaline-earth metal atom have an unpaired electron in the outer shell. The magnetic moment of our open-shell RbSr molecules would give us an additional degree of control not present in the bi-alkali systems. The electric dipole moment of RbSr molecules in the ground state is predicted to be about 1.6 D. The interaction between the molecules can be tuned with the help of electric as well as magnetic fields. Our molecules could be used for the quantum simulation of lattice spin models [45] in a more direct way without the need of encoding the spin in the rotational degree of freedom of the molecule [46].

1.1.1 **The quest for a ground-state molecular quantum gas**

The quantum simulation of many-body phenomena with ultracold ground-state molecules requires us to create samples that have low entropies. The ensemble of molecules should be prepared in a quantum degenerate state before the simulation. Evaporative cooling, a key technique for reaching quantum degeneracy, relies on elastic collisions. With respect to collisions the complex nature of molecules does not do us any favour: polar molecules, such as KRb and RbSr undergo chemical reactions of the type $2AB \rightarrow A_2 + B_2$ when they collide. A second collision process, “sticky collisions”, increases the chance of lossy collisions between ultracold molecules [47].

The inelastic collisions could be suppressed if the molecules did not come too close to each other during a collision. They should interact repulsively. One possible way of achieving repulsion between the molecules and thereby suppressing inelastic collisions in favour of elastic ones makes use of the anisotropic property of dipole-dipole interactions. The dipole-dipole force is repulsive when the dipoles are oriented parallel and approach each other in a plane perpendicular to the dipole axis. Forcing the dipolar molecules to approach each other in this side-by-side way would lead to a stable sample with long enough life-times for evaporative cooling. Experimentally the inelastic collision rate of KRb molecules has been decreased by polarizing the molecules with an electric field and freezing one motional degree of freedom by trapping the molecules in a pancake shaped potential [48]. Evaporative cooling to quantum degeneracy in this geometry has not yet been realized. Evaporation of the molecules is possible to the side of the pancake potential but requires a highly controlled electric field gradient [49].
Repulsive interactions between the molecules that act isotropically can be engineered with the help of external fields [50, 51]. Evaporation of cold molecules with such engineered repulsive van der Waals interactions has been demonstrated with electrostatically trapped OH-radicals [52]. Because of the additional magnetic moment of the open-shell RbSr molecules and the possibility to tune the interactions with electric and magnetic fields this technique could be employed in an easier manner than in bi-alkali systems [53]. The suppression of inelastic collisions in a bulk sample of the ultracold RbSr molecules would enable evaporative cooling, bringing the long-standing goal of a quantum gas of ground-state molecules within reach.

1.1.2 Alkaline-earth like and alkali atoms

Ultracold ground-state molecules with an open shell can be created from different combinations of alkali atoms with atoms that have two electrons in the outer shell. Ytterbium is a rare-earth atom with similar properties as Sr and has been brought to quantum degeneracy. Several groups combined Yb with alkali atoms in ultracold quantum gas experiments with the goal of creating open shell ground-state molecules. The first efforts are based on Rb and Yb atoms. The dipole moment of RbYb molecules is around 1 D [54]. The creation of quantum degenerate mixtures of Yb and Rb was challenging because of the unfavourable scattering properties of most isotopic combinations [55, 56]. Nevertheless in 2015 the Bose-Fermi mixture of $^{171}\text{Yb}$ and $^{87}\text{Rb}$ was obtained [57]. Photoassociation spectroscopy of YbRb molecules providing knowledge about the scattering properties and a possible molecule association strategy were performed as well [58].

Also quantum gas mixtures of Li and Yb were produced [59, 60]. The LiYb molecule has a relatively small electric dipole moment of about 0.1 D [61]. Efforts in creating Cs-Yb mixtures are in pursuit: the simultaneous magneto-optical trapping of Cs and Yb is reported in [62]. The dipole moment of CsYb molecules is predicted to be 0.24 D [63].

1.2 Ground-state molecules from ultracold atoms

The first ultracold molecules in the vibrational ground state have been realized with Cs$_2$ and KRb in the absolute ground state, and Rb$_2$ in the triplet ground-state [64, 38, 65]. These works and the works on polar bi-alkali molecules mentioned above lead to
1.2. Ground-state molecules from ultracold atoms

A well-established “recipe” to create ultracold ground-state molecules from atoms. The ingredients are laser cooled alkali atoms mixed in a dipole trap or optical lattice. The free atoms are associated to molecules that have a high vibrational quantum number. Such weakly-bound molecules are created by so-called Feshbach association. In a second step the weakly-bound molecules are transferred into the absolute ground state by Stimulated Raman Adiabatic Passage (STIRAP).

1.2.1 Magneto-association

The association of alkali atom pairs to weakly-bound molecules is performed with the help of a Feshbach resonance. A Feshbach resonance occurs when the state of two colliding particles is equal in energy to a molecular bound state. Figure 1.1a shows the states that are resonantly coupled in a Feshbach resonance and two potentials that support these states. The two atoms scatter with a finite collision energy, which is on the order of tens of $k \text{Hz} \times h$ at ultracold temperatures. The molecular potential that is associated with the colliding atom pair is called entrance channel. The second molecular potential typically associated with a different spin-configuration of the colliding atoms is called closed channel. The closed channel has an offset in energy with respect to the

Figure 1.1: Two-channel model of a Feshbach resonance. a) The atoms collide with an energy $E$ in the open channel potential. A bound state in the closed channel can be brought into resonance with the colliding atoms with the help of a magnetic field. b) During a magnetic field sweep $B(t)$ the free-atom pair is transferred adiabatically to a weakly-bound molecule. The dashed lines are the energies of the uncoupled states. The solid lines are coupled and therefore show an avoided crossing that enables the adiabatic transfer.
entrance channel due to hyperfine interaction and supports a bound state in the proximity of the two-atom collision energy. In a magnetically tunable Feshbach resonance the offset can be tuned by an external magnetic field because the magnetic moments of the corresponding states differ. If the free-atom and the bound state energy cross and a coupling between the states is present, then a mixing between the entrance channel and closed channel occurs. The Feshbach resonance is characterized by its width, which depends on the coupling strength between the channels. The strongest coupling mechanism leading to broad Feshbach resonances is due to electron spin-dependent interactions [10].

Feshbach resonances are an integral part in the field of ultracold atoms. Most notably they are used to tune the scattering length of atoms and thereby change the interaction strength in the ultracold ensemble. Tuning the scattering length is instrumental in the creation of BECs of atomic species with background scattering lengths that are unfavourable for evaporation [66, 67]. The loading of quantum gases of Rb and K into an optical lattice in order to obtain a low-entropy Mott-insulator for optimal molecule creation [68] is assisted by tuning the interspecies scattering length. The ability to control the interaction between atoms enabled the investigation of many-body phenomena such as the BEC-BCS crossover [6]. The most relevant application of Feshbach resonances for this thesis is the magneto-association of atom pairs into weakly-bound molecules. The concept of magneto-association relies on the coupling between the free-atom state and a molecular bound state. The free atom pair can be transferred coherently into the bound state by applying a varying magnetic [69] or RF field [70] close to a Feshbach resonance. Magnetic association can be achieved by sweeping the magnetic field adiabatically across the Feshbach resonance [71]. Figure 1.1b shows the energies of the free-atom $|a\rangle$ and the weakly bound states $|m\rangle$ during the adiabatic Feshbach sweep. The dashed lines show the energies of the uncoupled states. The coupling between entrance and closed channel leads to an avoided crossing. The magnetic sweep $B(t)$ across the avoided crossing transfers the free atom pair to weakly-bound molecules, if the sweep is slow enough for the magnetic moments of the atoms to evolve adiabatically with the field.
1.2. Feshbach resonances of Rb-Sr

Broad Feshbach resonances are useful for magneto-association, because the molecule association efficiency depends on the ratio between width of the resonance and the ramp speed across it [10]. The magnetic field sweep must have a slow enough ramp to associate the molecules adiabatically. In bi-alkali mixtures interspecies Feshbach resonances are broad because of the strong electron spin-dependent interactions. For example the Feshbach resonance used for magneto-association of KRb molecules is centred at 547 G and is roughly 1 G wide [72]. The magneto-association efficiency of K-Rb atom pairs to weakly-bound molecules can be as high as 87% [73].

The ground state of the alkaline-earth metal Sr is a singlet state. The electronic spin is zero. The electron spin dependent coupling mechanisms present in the bi-alkali systems, which leads to Feshbach resonances convenient for magneto-association, are absent in the Rb-Sr mixture. Despite the fact that Sr is non-magnetic in the ground state theoretical predictions point to the existence of Feshbach resonances in Rb-Sr collisions [74]. The largest contribution to the mixing between the entrance and closed channels lies in the change of the hyperfine interaction of Rb when the Sr atom is in its vicinity. The coupling leads to Feshbach resonances that are very narrow. The largest possible resonances, with a width of 200 mG, are predicted to occur only at fields above about 1000 G. For a successful magneto-association of RbSr molecules a magnetic field control of more than $10^{-4}$ is required. Considerable experimental efforts are required to achieve this level of control over high magnetic fields and could be pursued in the future.

We chose to put the prospect of magneto-association of weakly-bound molecules aside and instead try an approach relying on optical transitions. Before explaining the plan of action to associate weakly-bound RbSr molecules with laser light, the well-established step of transferring weakly-bound molecules to the absolute ground state will be reviewed.

1.2.3 Stimulated Raman Adiabatic Passage

Once the atom pairs are associated to weakly-bound molecules, they need to be transferred to the ro-vibronic (absolute) ground-state. The binding energy of thousands of cm$^{-1}$ (tens of K) has to be transported away from the molecules without heating them up in the process. A transfer step that relies on spontaneous relaxation is out
of the question. The weakly-bound molecules must be brought to the ground-state in a coherent manner. Using an electromagnetic field that transfers the weakly-bound molecules to the ground state directly is not possible either. The internuclear distance of the weakly-bound molecule and the ground-state molecule typically differ by two orders of magnitude. The spatial form of the wavefunctions of the weakly-bound state and the absolute ground-state are very different. The overlap between the two wavefunctions is very small leading to a small transition probability. Figure 1.2 illustrates why the weakly-bound molecules cannot be transferred directly to the ground-state. While the absolute ground-state wavefunction of the molecule is a simple Gaussian centred at the potential minimum, the weakly-bound wavefunction is extended over many\(^1\) Bohr radii. The wavefunction features fast oscillating behaviour and high probability regions near the classical turning points. The transition probability between the wavefunctions is proportional to the Franck-Condon factor (FCF), which is the square of the overlap integral between the wavefunctions. Integrating the fast oscillating part of the weakly-bound wavefunction over the Gaussian ground-state averages to nearly zero.

The challenge of the negligible FCF between the weakly-bound state and the ground-state is addressed by transferring the molecules through an intermediate state. The intermediate state is a vibrational state in an electronically excited potential of the molecule. The electronically excited potential of the intermediate state is chosen such that the selection rules for electronic transitions are satisfied [76]. The intermediate state is carefully chosen such that its wavefunction overlaps with the weakly-bound as well as ground-state wave function. An ideal example of an intermediate state is shown in Fig. 1.2. The weakly-bound wavefunction overlaps well with the intermediate wavefunction at the outer turning point and the ground-state overlaps with the inner turning point of the intermediate wavefunction.

The coupling of the weakly-bound molecules to the electronically excited state and from there to the absolute ground-state is done optically, using a pump and a dump laser. The pump laser with frequency \(\nu_P\) and coupling rate \(\Omega_P\) couples the weakly-bound state \(|a\rangle\) with the intermediate state \(|m\rangle\) and the dump laser of frequency \(\nu_D\) couples \(|m\rangle\) to the ground-state \(|g\rangle\) with rate \(\Omega_D\). A possible coherent population transfer scheme consists of a \(\pi\)-pulse from the pump laser depleting the weakly-bound

\(^1\)The extent of the wavefunction of a weakly-bound molecule produced by magneto-association is ten- to hundred-fold larger than shown [75].
1.2. Ground-state molecules from ultracold atoms

molecules and populating the intermediate state. Subsequently, a $\pi$-pulse using the dump laser transfers the population of the intermediate state to the ground-state. Because of its electronically excited nature, the lifetime of the intermediate state is short. In order to avoid excessive loss, the pulses must therefore be very short, which requires high coupling rates $\Omega_P$, $\Omega_D$. Many other transfer schemes exist [77], but the lossy nature of the intermediate state compromises the transfer efficiency for most schemes.

Stimulated Raman Adiabatic Passage (STIRAP) [78] is a transfer scheme during which the intermediate state is not populated. The transfer efficiency is robust against slight changes in the pump and dump laser pulse shapes and is not compromised by the lossy nature of the intermediate state [79]. Therefore STIRAP is an ideal state transfer scheme for transferring the weakly-bound molecules to the absolute ground state. In order to avoid spontaneous emission from the intermediate state $|m\rangle$ the system is kept in the so-called dark state during the STIRAP sequence. The dark state $|D\rangle = \Omega^{-1} (\Omega_D |a\rangle - \Omega_P |g\rangle)$ with $\Omega = \sqrt{|\Omega_P|^2 + |\Omega_D|^2}$ exists when the pump and the dump lasers are resonant with the respective transitions. Spontaneous emission does not occur, because the dark state has no contribution from the intermediate state. For the STIRAP to work efficiently the system has to evolve adiabatically from $|a\rangle$ to $|g\rangle$ through $|D\rangle$. Adiabaticity requires phase coherence between the pump and dump lasers. The initial state of STIRAP is $|a\rangle$ dressed with the dump laser photons. The state $|a\rangle$ does not absorb the dump light. This dark state in the initial configuration evolves adiabatically into a superposition of $|a\rangle$ and $|g\rangle$ when the intensity of the pump laser is ramped up. Simultaneously the intensity of the dump laser is ramped down. When the dump intensity is zero, the dark state evolved into $|g\rangle$ dressed with the pump laser photons. Here again, $|g\rangle$ does not scatter photons of the only laser beam present, the pump laser. Switching the pump laser off leaves the system in $|g\rangle$ and terminates the STIRAP sequence. During the coherent evolution the admixture of the intermediate state to the dark state remains zero. This is why the transfer efficiency is not compromised by the finite lifetime of the intermediate state.
1.2.4 Creation of weakly-bound molecules by STIRAP

STIRAP also can be used to associate atom pairs into weakly-bound molecules. When the conventional way of creating weakly-bound molecules, namely magneto-association, is not in reach, STIRAP becomes especially useful. The association of ultracold atoms to weakly-bound molecules by STIRAP was demonstrated in our experiment using $^{84}$Sr atoms [80]. The Sr atoms were brought to quantum degeneracy and subsequently transferred into an optical lattice. Populating optical lattice sites with two Sr atoms enhanced the local density, which increases the coupling between the free atom state and the molecular state during STIRAP. The two STIRAP lasers were derived from the laser addressing the narrow-linewidth transition $^1S_0-^3P_1$ of Sr. The STIRAP laser frequencies were tuned to molecular resonances. The pump beam was resonant with the $\nu' = -3$ vibrational level bound in the excited $0_u$ potential. The dump laser coupled this excited molecular state to the $\nu = -2$ vibrational state in the electronic ground state. The binding energy of the resulting molecule was 645 MHz. The STIRAP sequence produced $4 \times 10^4$ Sr$_2$ molecules with an efficiency of 30%.

The technique can be employed as the intermediate step towards the creation of polar RbSr molecules in the ro-vibronic ground state. Other molecules consisting of an alkali and alkaline-earth like atom could be associated with the help of STIRAP. The example of Sr$_2$ association shows the requirements for the creation of weakly-bound RbSr molecules by STIRAP. Firstly, the atoms need to be brought to quantum degeneracy and transferred into an optical lattice. Secondly molecular resonances in the excited

\[ \text{Figure 1.2: Illustration of the Franck-Condon principle.} \]
1.3 Thesis overview

The achievements on the way towards the realization of ultracold RbSr molecules will be described in the following five chapters. In Chap. 2 I will give an overview of the experimental apparatus and explain the changes made, which were necessary to produce quantum gas mixtures of Rb and Sr. These efforts lead to the production of degenerate quantum gas mixtures of $^{87}\text{Rb-}^{88}\text{Sr}$ and $^{87}\text{Rb-}^{84}\text{Sr}$, which I will present in the subsequent Chap. 3. Then I will describe the experimental realization of a double Mott insulator of $^{87}\text{Rb-}^{84}\text{Sr}$ in Chap. 4. The Mott-insulator is an ideal starting point for associating ultracold atoms into molecules.

In order to make the association of atoms into molecules successful, spectroscopic information about the molecules must be retrieved first. Chapter 5 contains the description of our work on photoassociation spectroscopy of RbSr molecules. In the last chapter (Chap. 6) I will present a frequency-shifted feedback laser based on a tapered amplifier. This opto-electronic device emits laser light with a spectrum consisting of equidistant, discrete modes spanning 370 GHz, which could be employed as a frequency reference tool in further spectroscopic studies of RbSr molecules or to frequency-stabilize STIRAP lasers that could transfer the molecules to deeper vibrational states.
Chapter 2

Experimental Setup

Our experimental setup was initially designed and built to create degenerate quantum gases of Sr and to use the quantum gases as a platform for quantum simulation experiments. On the machine the first successful Bose-Einstein condensation of $^{84}\text{Sr}$ [81] and $^{86}\text{Sr}$ [82] as well as reaching Fermi degeneracy in a cloud of $^{87}\text{Sr}$ [83] was achieved between 2009 and 2010. Since 2012 the machine was extended to produce Rb-Sr quantum gas mixtures.

Because the experimental details of the Sr machine can be found in the thesis of S. Stellmer [84], I will give only a brief overview of it in Sec. 2.1. In Sec. 2.2 the additions to the machine that were necessary for the creation of Rb-Sr mixtures will be explained. The explanation is to be read as a manual for future researchers who use and maintain the Rb subsystems.

2.1 SrBEC machine

The SrBEC machine consists of three essential subsystems, which will be explained in the following. The first subsystem is the vacuum assembly, in which the atomic vapour of Sr is cooled and trapped. Magnetic fields necessary for cooling and trapping are produced by coils attached to the vacuum assembly. The second subsystem contains near-resonant laser systems used to address the optical transitions of Sr, thereby exerting optical forces on the atoms or probing them. In addition infrared lasers are used for dipole trapping. The third system is the control system, which allows the experimentalist to control the SrBEC machine with the help of the computer. The intensities and frequencies of the laser beams and the magnetic field strengths produced by the current carrying coils can be controlled. The control system is described in detail elsewhere [85].
Chapter 2. Experimental Setup

2.1 Vacuum chamber

In the vacuum assembly the atomic beam is produced, the atoms are slowed down, trapped, and cooled down to quantum degeneracy. Figure 2.1 shows the compartments of the assembly where the steps towards quantum degeneracy occur.

The production of the atomic beam takes place in the oven, where a solid Sr sample is heated up to a temperature of 530°C. The atomic vapour exits the beam source through a nozzle consisting of about 110 tubes with 200 µm inner diameter. This microtube array emits a collimated Sr beam flying at a mean velocity of 530 m/s towards the glass cell [86, 87]. The heated metal leads to higher pressures in the oven chamber, which would cause short lifetimes of the trapped atoms in the glass cell. In order to maintain low pressure in the glass cell a differential pumping tube separates the oven chamber and UHV chamber that consists of the Zeeman slower and the glass cell. Before the atoms enter the UHV chamber through the differential pumping tube, they fly through the transverse cooling section. There the atomic beam transversal velocity spread is decreased by a pair of crossed molasses beams [88]. This transverse cooling stage increases the brightness of the atomic beam, which is especially important when operating the machine with the low-abundance (0.6%) $^{84}$Sr isotope. The number of $^{84}$Sr atoms in the magneto-optical trap (MOT) can be enhanced threefold by means of the transverse cooling. The transversally cooled atoms enter the ZS, where they are
2.1. SrBEC machine

decelerated with the help of a combination of a counter-propagating laser beam and a magnetic field. The current through the three magnetic coils (i-iii) generating the magnetic field can be controlled independently allowing for a adaptation of the ZS to operate for different atomic species. The magnets i and ii are operated in reverse to iii leading to a spin-flip region at two-thirds of the length of the ZS. Upon arrival at the glass cell, the atoms are cooled and trapped in the MOT. After reaching a low enough temperature the Sr is captured in the science dipole trap, where evaporative cooling or further experimental steps are performed. The ZS magnetic field significantly shifts the centre of the the MOT quadrupole field. To counteract that shift, coil (v) on the side of the glass cell opposite to the ZS is used. Particles that are neither decelerated by the ZS nor captured in the MOT hit the back-end window. If the particles stuck to that window, the ZS beam would be obstructed leading to a malfunction of the ZS. The window is made of sapphire, which has a smaller affinity to bind particles on its surface and reacts less with alkali and alkaline-earth metals. In addition the window is heated to 180°C to detach the particles from the surface.

**Figure 2.2**: Energy level scheme for Sr with relevant transitions for laser cooling.
2.1.2 Strontium laser systems

We use three laser systems for laser cooling of Sr, each addressing a specific transition. The three relevant transitions are shown in Fig. 2.2. Initial laser cooling to 1 mK and deceleration in the ZS is performed on $^1S_0-^1P_1$ at 461 nm. A laser on $^3P_2-^3D_1$ at 497 nm is used to repump atoms from the $^3P_2$ meta-stable state back into the ground state. A great feature of Sr is to allow laser cooling to less than 400 nK and even to quantum degeneracy [89] on the $^1S_0-^3P_1$ transition. The three laser sources providing light to address these three transitions are now briefly described.

The 461 nm laser light is created via doubling of semiconductor laser light at 922 nm. The 922 nm master oscillator output is amplified to 1 W by a tapered amplifier and sent through a doubling cavity based on an LBO crystal. The conversion efficiency from infrared to blue laser light is about 30%. A small part of the 461 nm-light is used for Sr vapor cell spectroscopy, providing a signal for locking the master laser. The rest of the 461 nm light is split into MOT, ZS, transverse cooling\textsuperscript{1} and imaging beam path. The frequency of the laser in each path is adjusted by AOMs.

The blue transition is not entirely cyclic. The initial blue MOT loses atoms to the metastable $^3P_2$ state, which is beneficial for accumulation of the least abundant isotope $^{84}$Sr in a magnetic trap - a key artifice in creating the first Sr BEC. Once enough atoms are accumulated in this metastable reservoir a repump is needed to bring the atoms from the metastable state to the ground state for further laser cooling on the intercombination line $^1S_0-^3P_1$. Many repumping transitions exist [91]. Our system works with the $^3P_2-^3D_1$ transition at 497 nm. The laser accessing this transition consists of a laser diode with 300 mW output power at 994 nm, which is doubled in a cavity. The laser diode is stabilized to an ultra-low expansion (ULE) reference cavity. One of the cavity mirrors is mounted on a piezo-actuator. The cavity length can be tuned by an applied high voltage to adjust the laser frequency.

The laser accessing the intercombination line $^1S_0-^3P_1$ with 7.4 kHz linewidth at 689 nm is referenced to a high-finesse cavity in order to achieve a laser-linewidth below the natural linewidth of the atomic transition. Because the cavity resonance drifts by about 50 kHz/h, which is more than the transition linewidth after just a few minutes, long-term stabilization is necessary. One mirror of the cavity is piezo-driven so that the length can be locked to the Doppler-free saturated absorption feature from a Sr laser diode [90] became commercially available, C.-C. Chen installed an injection-locked slave diode for the transverse cooling beam.
2.2 Rubidium subsystems

The additions to the SrBEC machine that lead to the creation of ultracold mixtures of Rb and Sr will be described. The oven was extended to produce a Rb and Sr beam. Two coils were added to the ZS slower so that the Rb could be decelerated. For cooling and trapping the Rb a near-resonant laser system addressing the D\textsubscript{2} line of Rb was built. We installed additional imaging system to be able to detect the Rb atoms. The dipole trapping of Rb (storage ODT) was realized by a high-power infrared laser system. We built a bichromatic dipole trap, which is used increase the overlap between Rb and Sr BECs. Finally an optical lattice was installed to be able to trap individual atom-pairs in the lattice.

2.2.1 Double species oven

The creation of a Rb-Sr quantum gas mixture requires us to add a source of Rb atoms to the SrBEC machine. The vacuum assembly contains one ZS, which initially produced a slow Sr beam, but was already designed to be also usable for slowing Rb. Therefore by extending the Sr atomic beam source with the capability of emitting also a Rb atomic beam and by slightly adapting the ZS a cold Rb and Sr beam can be created.

An atomic beam source emitting multiple species at the same time benefits from a design where the temperatures of the species can be controlled independently. Mixing different atomic species in one beam source oven would lead to vastly different fluxes, because of the different vapour pressures of the species. Stan and Ketterle [93] propose to combine the atomic vapours from separate species reservoirs at different temperatures in a mixing chamber. The mixed vapour is then emitted from a nozzle in the mixing chamber. Figure 2.3 shows the oven used in the Rb-Sr machine. We use two separate reservoirs containing Rb and Sr. The temperatures of the reservoirs containing
Chapter 2. Experimental Setup

Figure 2.3: Double species atom source adapted from [92]. The top image is a cross-section schematic of the Sr reservoir and oven and part of the Rb oven. The red boxes and the red wires wound around vacuum parts, labeled in bold, are heating wires. The crossed green boxes with non-bold lower-case labelling are thermocouple positions for temperature monitoring. The bottom image shows the Rb oven section with reservoir and valves.

the metals can be adjusted individually to set the atom fluxes. The atomic beams are kept separate and overlap at the output of the nozzle. The Sr oven part and the procedure to fill it with 10 g of Sr is described in [84]. The new Rb oven is mounted upstream from the Sr oven, with the Rb atomic beam passing through a flythrough section in the Sr oven. The two ovens are separated from each other by a CF40 T-connector housing a Ti-sublimation pump. The long tube of the T-connector housing the Ti-sub pump and is cooled by a series of Peltier-elements to below 5°C. Residual Rb vapour freezes onto the cold wall of the T-connector. Instead of directly mounting the Rb reservoir to the oven section, a valve and a CF16 T-connector is attached between the two. The third port of the T-connector is attached to another valve, which enables us to connect a vacuum pump to the Rb reservoir. The installed valves and an attachment for the vacuum pump make a future reloading of the Rb reservoir possible without filling the
2.2. Rubidium subsystems

<table>
<thead>
<tr>
<th>Thermocouple label</th>
<th>Name</th>
<th>Operating temperature (°C)</th>
<th>Stand-by temperature (°C)</th>
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<tbody>
<tr>
<td>rb 1</td>
<td>Back flange</td>
<td>215</td>
<td>175</td>
</tr>
<tr>
<td>rb 2</td>
<td>Oven back</td>
<td>235</td>
<td>191</td>
</tr>
<tr>
<td>rb 3</td>
<td>T connector</td>
<td>270</td>
<td>94</td>
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<td>rb 4</td>
<td>Reservoir top</td>
<td>245</td>
<td>103</td>
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<tr>
<td>rb 5</td>
<td>Reservoir bottom</td>
<td>120</td>
<td>68</td>
</tr>
<tr>
<td>rb 6</td>
<td>Oven front</td>
<td>240</td>
<td>198</td>
</tr>
<tr>
<td>rb 7</td>
<td>Front flange</td>
<td>225</td>
<td>194</td>
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<td>Big T connector</td>
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<td>Back flange</td>
<td>580</td>
<td>574</td>
</tr>
<tr>
<td>sr 2</td>
<td>Oven back</td>
<td>610</td>
<td>602</td>
</tr>
<tr>
<td>sr 3</td>
<td>Reservoir</td>
<td>530</td>
<td>527</td>
</tr>
<tr>
<td>sr 4</td>
<td>Oven front</td>
<td>600</td>
<td>599</td>
</tr>
<tr>
<td>sr 5</td>
<td>Front flange</td>
<td>590</td>
<td>587</td>
</tr>
</tbody>
</table>

Table 2.1: Oven temperature settings for the experiment.

The whole oven section with gas at 1 atmosphere, which would require baking that section after reloading the reservoir. Similarly the Rb sample can remain under vacuum when the Sr reservoir is replenished, reducing the risk of loosing Rb by oxidation. The refilling procedure is described in detail in [92]. In short, a glass ampoule containing 5 g of Rb is placed in the reservoir tube. After mounting the reservoir to the oven section and establishing a low enough pressure the reservoir tube is compressed with pliers until the ampule cracks, allowing Rb vapour to enter the vacuum chamber. To establish the desired temperatures across the entire oven assembly 13 heating wires are installed shown as dark red squares and lines in Fig. 2.3. The currents through the heating wires are carefully adjusted with the help of current controlled DC-power supplies. The vicinity of the micro-tube arrays is set to the highest temperature in order to prevent the atoms from forming deposits in the micro-tubes leading to a clogged beam source. The Rb reservoir temperature is ramped down when the machine is not operating for several days in order to prolong the reservoir lifetime. The temperatures are monitored with the help of thermocouples installed at different points of the oven assembly (green crossed boxes). The settings of the temperatures at each measurement point in the active as well as stand-by state of the experiment are tabulated in Tab. 2.1.
2.2.2 Zeeman slower

In the ZS atoms are decelerated by a counter-propagating near-resonant laser beam. The frequency of the laser beam is red-detuned with respect to the atomic transition matching the Doppler-shift of the fast moving atoms. As the atoms are decelerated and become slower, they experience a smaller Doppler-shift and eventually become invisible to the slowing light. To achieve a constant deceleration the resonance frequency of the atoms is adjusted along the flight path. This adjustment is realized by a spatially varying magnetic field making use of the Zeeman effect. The profile of the

<table>
<thead>
<tr>
<th>Name</th>
<th>ZS coil</th>
<th>Rb loading current (A)</th>
<th>Sr loading current (A)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1st ZS</td>
<td>i</td>
<td>3</td>
<td>10</td>
</tr>
<tr>
<td>2nd ZS low</td>
<td>ii</td>
<td>1.9</td>
<td>6.7</td>
</tr>
<tr>
<td>2nd ZS high</td>
<td>iii</td>
<td>45</td>
<td>125</td>
</tr>
<tr>
<td>Suppl. coil 1</td>
<td>a₁</td>
<td>1</td>
<td>0</td>
</tr>
<tr>
<td>Suppl. coil 2</td>
<td>a₂</td>
<td>10.5</td>
<td>0</td>
</tr>
<tr>
<td>Compensation</td>
<td>v</td>
<td>56</td>
<td>85</td>
</tr>
<tr>
<td>MOT coil</td>
<td>iv</td>
<td>17</td>
<td>90</td>
</tr>
</tbody>
</table>

Table 2.2: Optimal currents of the ZS and MOT coils.
magnetic field strength along the flight path takes the form of a square root function, if constant deceleration is desired [88]. The dashed line in Fig. 2.4a shows the magnetic field strength along the flight path of the atoms we calculated for a ZS with 110 m/s² deceleration and capture velocity of our MOT of 50 m/s. The mean velocity of the atoms from the oven is 300 m/s. The $\Delta = -170$ MHz red detuned ZS beam decelerates the atoms to 38 m/s, which is below the capture velocity of the MOT. The difference between the optimal B-field shape for Rb and Sr is small and most pronounced towards the end of the ZS. To accommodate the difference, we have installed two coils at the end of the ZS that are only used when slowing Rb. The magnetic field profile at 0.7 m is smoothed out by two additional coils of 13 windings (supplementary coils $a_1$, $a_2$) installed close to the glass cell. The currents for the ZS coils yielding a high flux of Rb are found by maximizing the fluorescence signal of the Rb MOT and are tabulated in Tab 2.2. The solid line in Fig. 2.4a shows the calculated magnetic field by using the law of Biot-Savart for each turn of the ZS coils carrying the found current. The profile at 0.55m is steeper than intended, likely limiting the flux of Rb slowed by the ZS. In the future another supplementary coil similar to $a_1$ could be installed there, which would allow us to increase all ZS currents without exceeding the maximum possible deceleration. The correspondingly higher capture velocity would result in a higher flux of Rb.

With the adapted ZS we can load $7.5 \times 10^7$ Rb atoms in 10 s into the MOT. Fig. 2.4b shows the loading curve of the Rb atoms captured in the MOT over time. Usually we load the Rb MOT for 5 s, which provides us with enough atoms to create Rb-Sr quantum gas mixtures.

### 2.2.3 Rubidium laser system

To laser cool Rb we use the $5s^2 S_{1/2} \rightarrow 5p^2 P_{3/2}$ transition at 780 nm. The hyperfine structure of the relevant states is shown in Fig. 2.5a. The trapping and cooling laser for MOT, ZS and imaging addresses the $(F = 2) \rightarrow (F' = 3)$ transition. Due to the finite linewidth of the transition the atoms can be excited to the $(F' = 2)$ hyperfine state and subsequently decay to the $(F = 1)$ ground-state. Atoms in the $(F = 1)$ state are dark with respect to the trapping and cooling light. To guarantee cyclic operation of the MOT and ZS as well as to allow imaging of $F = 1$ atoms, we repump the atoms to the $(F = 2)$ state. Repumping is achieved with laser light resonant with the $(F = 1) \rightarrow (F' = 2$ or 1) transitions. The Rb laser system produces the cooling light for MOT, ZS, imaging and the
Chapter 2. Experimental Setup

Figure 2.5: a) Hyperfine structure of the D_2 line in Rb at 780 nm, which we use for cooling and trapping (adapted from [94]). b) Frequency level scheme of the repump and cooling laser systems.

Figure 2.6: Doppler-free saturated absorption (dashed) and error signals (solid) of $^{87}$Rb in the vapour cell. a) spectroscopy signal from the cooling laser addressing the $F = 2$ and b) repump laser addressing $F = 1$ hyperfine state. The locking points are marked by the arrows.
2.2. Rubidium subsystems

repump light with the help of two external cavity diode lasers (ECDLs). The frequency of the ECDLs is stabilized using laser locking to a Rb spectroscopy reference. AOMs are used to generate various beams with the correct frequency for MOT, ZS, etc. Each beam is transported by optical fibres to the vacuum assembly.

Figure 2.5b depicts the frequency level scheme of the cooling and repump laser systems with respect to the hyperfine structure of the $5s^2S_{1/2}$ and $5p^2P_{3/2}$ states. The cooling laser source (Toptica TA-pro) is red detuned from all hyperfine states of $5p^2P_{3/2}$. The AOMs shift the laser beams to higher frequency. The laser frequency is stabilized to the cross-over resonance ($F = 2 \rightarrow F' = 3, 2\omega$) of the Doppler-free saturated absorption spectrum shown in Fig. 2.6a. To derive the various required beams, the output of the master laser is split up and the AOMs for ZS, MOT and imaging beams are shifted accordingly to achieve the desired detuning from the addressed level.

The frequency of the repump laser (Toptica DL 100) is stabilized to the ($F = 1 \rightarrow F' = 0$) spectroscopy line of a Doppler-free saturated absorption spectrum shown in Fig. 2.6b. The MOT AOM generates a beam resonant with the ($F = 1 \rightarrow F' = 1$) transition and the ZS repump AOM generates a beam with -176 MHz detuning from the ($F = 1 \rightarrow F' = 2$) transition.

The full schematic of the two laser systems is shown in Fig. 2.7. The TA-pro cooling laser features a tapered amplifier (TA) in addition to the ECDL. The TA amplifies the master light to 900 mW, which satisfies the intensity requirements for the MOT and ZS beams. Each MOT beam requires roughly 25 mW and the ZS requires 75 mW of laser power at the optical fibre input of the laser system. After the frequency shift by the respective AOMs the light is coupled into polarization maintaining optical fibres\(^2\) transporting the light to the vacuum assembly.

The power output of the repump ECDL is 40 mW. This repump light is distributed to the spectroscopy cell, the ZS and the MOT. The spectroscopy cell requires only 200µW. The ZS repump light is superimposed with the ZS cooling light on a 50/50 beamsplitter, such that 5 mW of repump light enter the ZS optical fibre. Two MOT repump beams of several 100 µW of power each are sent to the glass cell via optical fibres. One repump beam is used for standard optical pumping during the MOT loading sequence and during absorption imaging. The other “dark-spot” repump beam

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\(^2\)The polarization stability of some of the beams is ensured by placement of $\lambda/2$-retardation plates before fibre couplers. The angle of the retardation plate is set to minimize the fluctuations in the fibre output polarization. In some cases tilting the plates away from orthogonal to the optical axis increased the stability further.
is employed only for loading of the dipole trap. Close to the glass cell a wire casts a shadow into the dark-spot repump beam. The shadow is superimposed with the storage ODT (see Sec. 2.2.6), such that atoms in the ODT are optically pumped into the dark state and do not scatter resonant light coming from the cooling laser. This dark spot technique increases the density of atoms in the ODT.

2.2.4 Combined MOT and imaging optics

The starting point of the production of Rb-Sr quantum gas mixtures is trapping the Rb atoms in a MOT and mixing them with the Sr atoms. The Sr atoms are trapped in two consecutive MOT stages. First the broad-linewidth transition $^1S_0-^1P_1$ at 461 nm is employed to Zeeman slow and capture many atoms. In a second stage, the narrow-linewidth transition $^1S_0-^3P_1$ at 689 nm is used to laser cool the Sr gas to temperatures below 1 µK. Because the optical access of the glass cell is limited, the blue and red MOT beams are superimposed by dichroic mirrors and sent into the glass cell along the same axis. In order to save space around the glass cell for ODT, spectroscopy and other beams the Rb MOT beams were superimposed with the Sr MOT beams as well. Sr blue, red and Rb light are well separated in wavelength, allowing us to use dichroic mirrors to combine the beams.

**MOT beams.** Figure 2.8 shows a schematic of the optics setup of one axis of the MOT in the horizontal direction. One dichroic mirror transmitting 780 nm and reflecting 689 nm was used to combine the Rb MOT with the Sr red MOT beam about one meter away from the glass cell. Closer to the glass cell a second dichroic, transmitting 780 and 689 nm and reflecting 461 nm, was installed to overlap the Sr blue MOT beam with the other two beams. The blue MOT beam is superimposed later in the beam path because the polarizing beam splitters used to combine the imaging beams with MOT beams are not optimal for all three wavelengths. At the other side of the glass cell the blue beam is separated again from the red and 780 nm beams to be sent onto retro-reflection mirrors back into the glass cell. The angles of the achromatic λ/4-wave plates placed around the glass cell are adjusted for optimal polarization of the red MOT, because the red MOT is most sensitive to an imbalance in polarizations of the counter-propagating MOT beams. The combination of the Rb and Sr beams for the second horizontal and the vertical MOT beams is achieved with the help of dichroic mirrors in a similar manner (not shown).
2.2. Rubidium subsystems

Imaging. The optical axis used for the horizontal imaging is shared with the MOT beam axis. The MOT and imaging beams have orthogonal polarizations and are separated by polarizing beam splitters (PBS). The Rb (green in Fig. 2.8) and Sr red imaging beams (red dashed) are first overlapped by a dichroic mirror and then superimposed onto the combined MOT beam of Rb and Sr red by the PBS. The PBS at the glass cell combines the Sr blue imaging beam (purple) with the Sr blue MOT beam and both are reflect off the dichroic mirror into the glass cell. The atoms are imaged by a gradient-index lens with a focal length of 70 mm. Imaged light is separated by the dichroic mirror. The Sr blue light is reflected off the dichroic, transmitted through the imaging PBS, and focused onto the CCD-camera (Luca-R, Andor) by a lens with 140 mm focal length. The Rb and Sr red light is transmitted through the dichroic and another imaging PBS. The 150 mm focal length lens focuses the light onto the second CCD-camera. The magnification of the Rb imaging system (3.45 µm/pixel) is similar to the Sr system (3.96 µm/pix). The camera calibration is achieved by measuring the parabolic trajectory of a cloud of Rb in the non-magnetic \( F = 1, m_F = 0 \) state in free fall for different time-of-flights.

2.2.5 Wide-angle imaging system

As mentioned before the MOT and imaging beams are superimposed in our setup. This is no problem for images taken when the MOT beams are off, e.g. for absorption images of a quantum gas. But imaging the MOT itself is hard, since the MOT beams saturated the camera. During the Rb subsystem installation phase it was desirable to observe the Rb atoms trapped in the MOT for direct feedback in the MOT for in-situ, real-time optimization. Moreover during the installation phase the Sr optical components around the glass cell had to be rearranged and therefore the Sr MOT had to be reoptimized as well. Therefore an additional imaging system for in-situ observation of the MOT was installed off-axis from the MOT beams. The optics schematic is depicted in Fig. 2.10. The magnification of
Chapter 2. Experimental Setup

The system (9.46 \( \mu \text{m/pixel} \)) is smaller than the one of the MOT-axis imaging system, resulting in a larger field of view. Rb or Sr atoms can be imaged in fluorescence as well as time-of-flight absorption. The latter is used to obtain more precise atom number measurements, important for MOT atom number optimization. The absorption imaging beam has to be adapted to Rb or Sr by placing a fibre providing the corresponding light into the imaging beam collimator. If Sr is imaged, we simply use the fibre that usually provides the vertical Sr imaging light.

Figure 2.9 shows a fluorescence image of our first Rb MOT with an estimated atom number of \( 10^4 \). With the help of the wide-angle imaging system we could optimize this initially miniscule MOT to obtain enough atoms for our needs.

2.2.6 Rb dipole trap

The standard path to a quantum degenerate gas of atoms starts with laser cooling and then proceeds with evaporative cooling. Evaporative cooling requires us to trap the gas in a conservative potential. In our experiment the Sr is trapped in an infrared dipole trap. By lowering the laser intensity of the trapping laser we can perform evaporation and reach quantum degeneracy of the Sr. Dipole trapping of Rb atoms requires a higher laser intensity than available from the Sr ODT laser, because the temperature of laser cooled Rb is higher than the temperature achieved with narrow-linewidth laser cooling of Sr.

A new dipole trap system is installed, which is an important tool for the creation of ultracold mixtures of Rb and Sr. Figure 2.11 shows the optical setup for this storage dipole trap. A 100-W fibre laser at 1070 nm (YLR-100-LP-AC-Y12 from IPG) is chosen as the light source for the storage ODT. The output collimator of the fibre laser is mounted on the optical table close to the main experimental chamber with a customized mount. The mount enclosed the fibre collimator and an additional \( f = 200 \text{ mm} \) lens. The lens is part of a telescope to reduce the waist size of the beam, but serves also as a protective window for the fibre collimator lens against dust. The second lens of the telescope has a focal length of 75 mm. The waist of the beam is reduced from 1 mm to 375 \( \mu \text{m} \) by the telescope in order to fit the aperture of an AOM used to control the dipole trap power. The maximum power is manually adjusted using a \( \lambda/2 \) waveplate combined with a Brewster plate. The beam transmitted through the Brewster-plate

\footnote{The size of a pixel of the Luca-R camera is 8 \( \mu \text{m} \), therefore the magnification is close to 1:1.}
is dissipated on a beam dump. The reflected beam is sent through the beam power control AOM. The 0th order transmission of the AOM is safely dissipated on a water-cooled beam dump. The 1st order diffraction is sent to a three-lens telescope and a final focusing lens of $f = 150\,\text{mm}$ focal length in front of the glass cell. The three-lens telescope together with the final lens are used to define the waist of the ODT at the position of the atomic cloud. Changing the trapping volume of the ODT can be changed by adapting the three-lens telescope configuration. Initially we set the waist of the storage trap to $40\,\mu\text{m}$ at the position of the atoms. The fraction of atoms loaded from the laser cooled cloud into the ODT was 10%. In Amsterdam we changed the lens configuration to obtain a waist of $110\,\mu\text{m}$ and the loading efficiency increased to 80%. To maintain the same trap depth as before, higher intensities of the laser were required. The high intensity laser beam damaged the PBS of the Faraday isolater. A compromise between good loading efficiency and optics lifetime was obtained by using a waist of $60\,\mu\text{m}$ resulting in a loading efficiency of 40% and long lifetime of the optics.

Leaving the main experimental chamber the dipole trap beam is sent through a lens and reflected into a water cooled beam dump by two adjacent mirrors. The beam transmitted through the last mirror is sent onto a monitoring camera. The camera serves two purposes. Firstly the camera is used for the alignment of the dipole trap onto the Rb cloud. In order to align the dipole trap onto the Rb MOT, the Rb MOT is imaged onto the camera using the lens after the glass cell. The dipole trap beam focus is also imaged onto the camera, and aligned with the MOT image. After this rough alignment loading of the ODT with a small number of Rb can be observed by absorption imaging. Secondly after optimizing the dipole trap loading, the imaging system is focused onto the focus of the dipole trap beam and serves for daily surveillance of beam position and profile.

### 2.2.7 Bichromatic dipole trap

All necessary components for the creation of quantum gas mixtures of Rb and Sr have been described and in the next chapter I will present how we produce Rb-Sr BECs. The first version of the beam dumps was made of aluminium. After a year of operation in Amsterdam, water had corroded through them, which never happened using the cooling water in Innsbruck. We speculate that galvanic erosion between the aluminium and the material of the cooling water pipes is to blame. In Innsbruck steel cooling water pipes were used, whereas in Amsterdam copper pipes are used. The problem was overcome by using water cooled copper blocks in thermal contact with aluminium beam dumps.
creation of a double BEC is an important step on the pathway to ultracold molecules. The next step involves transferring the double BEC into an optical lattice. We must ensure that during the transfer as many sites of the optical lattice as possible are filled with one Sr and one Rb atom. Having many doubly occupied sites is an excellent starting point for the creation of an ensemble of ultracold ground-state molecules. The number of doubly occupied sites depends on how well the BECs overlap spatially in the trapping potential. At the wavelength of the science ODT (1065 nm), where the double BECs are created, Rb and Sr have a threefold difference in polarizability. The trap depth and trapping frequencies of Rb are higher. Together with gravity, the difference of the dipole trapping potentials leads to the spatial separation of the BECs (see Chap. 4).

In order to overcome the spatial separation of Rb and Sr condensates, a second dipole trap beam is installed to adjust the trapping frequencies of the atomic species independently. The wavelength of the additional dipole potential laser is 532 nm, which is far-detuned from the Rb as well as Sr optical transitions. While the green, 532-nm ODT is attractive for Sr, it is repulsive for Rb. With the correct choice of relative intensity of the infrared and green dipole beams an equal trapping potential can be achieved for Rb and Sr, leading to overlapping condensates.

The optical scheme for power distribution of the green laser light between vertical and horizontal ODT is shown in Fig. 2.12a. The source for the green ODT is a 532-nm laser (Verdi-V8, Coherent) with a maximum output power of 8 W. The waist of the output beam is adjusted by the telescope to fit the beam into the apertures of the AOMs. The AOMs control the intensity of the vertical and horizontal beams. Then the beams are transported close to the glass cell by optical fibres. The laser distribution setup is placed close to the cell to avoid the use of long optical fibres. The spectral quality of the light is known to be compromised by stimulated Brillouin scattering in long optical fibres [95]. Fig. 2.12b shows the optical path of the green ODT near the glass cell. The green beam is combined with the existing infrared, 1065-nm ODT with the help of dichroic mirrors.

Careful overlap of the infrared and green science dipole traps is needed. The first step in doing so is to align the foci of the corresponding two dipole trap beams along their beam direction by the telescope close to the fibre-collimator of the green beam. The lens “a” is mounted on a translation stage driven by a micrometer screw. The position of the lens is precisely adjusted so that a Sr cloud is trapped by the green and
2.2. Rubidium subsystems

Infrared ODT at the same location. Because the waists in gravity direction (z) of the green and infrared horizontal trapping beams are quite small, less than 20 µm, the most critical alignment parameter is their relative z-position. The difference of z-positions is minimized with the following, precise method. After creating a Sr BEC in the infrared trap, the horizontal green ODT is switched on rapidly. At the same time the infrared trap is switched off. If the two traps have a difference in the z-alignment, the Sr BEC is accelerated towards the centre of the green ODT after toggling the beams. The BEC then oscillates in the green trap with an amplitude proportional to the difference in the z-positions of the two traps. We minimize the amplitude of the BEC’s center-of-mass motion by changing the z-position of either the horizontal infrared or the green beam. The relative positioning of the trapping beams in the other direction as well as the alignment of the vertical beams is not as sensitive and can be done by measuring the position of the clouds on in-situ absorption images of the Sr cloud.

2.2.8 Optical Lattice

When the beam of an ODT is reflected back onto itself, a standing wave is created. Atoms experience a modulated potential inside the standing wave. With a three-dimensional arrangement of such optical lattice beams an artificial light crystal can be created. Whereas in a shallow lattice the atoms can move easily from lattice well to lattice well, for a deep enough lattice they will localize on lattice sites. A BEC can be “loaded into a lattice” by exposing the BEC to a lattice with slowly increasing depth. If a sufficient depth is reached the BEC will undergo a transition to a Mott insulator. The demonstration of this quantum phase transition showed that ultracold atoms combined with optical lattices have a great potential in simulating solid-state like systems [5]. Efforts to simulate Bose-Hubbard models with long-range interactions with ultracold dipolar molecules in optical lattices are already pursued [39]. Moreover the optical lattice is an excellent tool to enhance the efficiency of ultracold molecule association [96] and to prevent molecule loss through chemical reactions [97].

For our initial lattice experiments, performed only with Sr, an optical lattice with a wavelength of 532 nm was installed and a Mott-insulator of Sr atoms was prepared [84]. Because of the polarizability having opposite sign, Sr atoms are trapped in the antinodes of the standing wave of this lattice, whereas Rb atoms are trapped in the
nodes. The spatial offset of half a lattice wavelength would make RbSr molecule association in such a sample impossible.

We therefore installed a new 3D optical lattice setup in our experiment based on a laser source emitting 1064 nm light (Coherent, Mephisto MOPA, 48 W). An optical lattice with 1064-nm wavelength traps both atomic species on the anti-nodes of the standing wave enabling the population of sites with one atom of each species, which is the starting point for molecule association. The optical power distribution setup of the lattice laser is shown in Fig. 2.13a. The laser source has to be protected from the retro-reflected beams by a Faraday isolator. The laser beam is divided up into three beams by a sequence of $\lambda/2$ waveplates and polarizing beam splitters to provide power for the three lattice arms. The intensity of the beams is controlled by AOMs. In order to prevent interference between the arms, the AOMs shift the lattice beam frequencies by 162 MHz ($f_{AOM0} - f_{AOM3}$), 158 MHz ($f_{AOM1} - f_{AOM3}$) and 4 MHz ($f_{AOM1} - f_{AOM0}$) relative to each other. The three lattice beams are transported by polarization maintaining optical fibres to the glass cell. The optical lattice consists of one horizontal and two diagonal arms focused into the glass cell. The horizontal arm and the horizontal science beam cross at an angle of roughly 20° and lie in the horizontal plane. The diagonal arms are at an angle close to 45° in respect to the horizontal plane. Figure 2.13b shows the setup of the horizontal and one diagonal arm around the glass cell. The setup of the second diagonal arm is similar not shown for clarity. Each arm consists of the output fibre collimator, a PBS, two lenses, a pair of waveplates and retro-reflection mirrors. The PBS is used for conditioning the polarization and the waveplates are used to adjust the polarization precisely. The lenses focus the beam on the atoms.

For the alignment of the lattice beams on the atoms, the following procedure is recommended. The first step consists of aligning the lattice beam without retro-reflection. The focused lattice beam will have the same trapping effect as an ODT, therefore the rough alignment can be achieved following the procedure described in section 2.2.6. Once the lattice beam is in close proximity with the atomic cloud, the trapping action will appear as an increased atom density in absorption images. After overlapping the lattice beam with the atomic cloud in the science trap, the retro-reflecting mirror can be installed. For optimal alignment of the lattice beam with the retro-reflectored beam, the coupling of the retro-reflectored beam back into the fibre has to be optimized. If the retro-reflectored beam is coupled back into the fibre, the intensity of the Faraday isolator reflection marked in purple in Fig. 2.13a will increase. With the help of a power meter
placed at the marked location (star) one can optimize the fibre coupling. After the described procedure one can employ the Raman-Nath diffraction method [98] on a $^{84}$Sr BEC to further optimize the lattice depth.
Figure 2.7: Optical layout of the Rb laser system. The symbols of the optics components will be used throughout the thesis.
2.2. Rubidium subsystems

**Figure 2.8:** Optical setup of one horizontal MOT and imaging beam of the MOT for Rb and Sr.

**Figure 2.10:** Optical setup of the wide-angle imaging system.

**Figure 2.11:** Optical setup of the high-power storage dipole trap.
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Figure 2.12: a) Power distribution setup for the 532 nm Rb anti-trap. b) Schematic of the bichromatic science dipole trap at the glass cell.

Figure 2.13: a) Power distribution setup for the optical lattice. The purple beam is the retro-reflection of the lattice arms. For retro-reflection alignment a power meter is to be installed at the marked position (star). b) Schematic of the lattice at the glass cell.
Chapter 3

Double Bose-Einstein condensates of Sr and Rb

We report on the realization of quantum degenerate gas mixtures of the alkaline-earth element strontium with the alkali element rubidium. A key ingredient of our scheme is sympathetic cooling of Rb by Sr atoms that are continuously laser cooled on a narrow linewidth transition. This versatile technique allows us to produce ultracold gas mixtures with a phase-space density of up to 0.06 for both elements. By further evaporative cooling we create double Bose-Einstein condensates of $^{87}$Rb with either $^{88}$Sr or $^{84}$Sr, reaching more than $10^5$ condensed atoms per element for the $^{84}$Sr-$^{87}$Rb mixture. These quantum gas mixtures constitute an important step towards the production of a quantum gas of polar, open-shell RbSr molecules.

3.1 Introduction

Quantum degenerate gas mixtures of different chemical elements have opened up important new areas for the study of interacting quantum systems. The possibility to apply species-specific optical potentials [99] is a versatile tool and has for example been used to study the exchange of entropy between two gases [100] or mixed-dimensional systems [101]. The mass difference between the constituents of the mixture can lead to new few- and many-body phenomena [6, 102], such as novel trimer states [103, 104, 105] or crystalline quantum phases [106]. Quantum gas mixtures of two elements have also attracted a great deal of attention because they are an ideal starting point for the coherent production of heteronuclear ground-state molecules, which can have large electric dipole moments [38, 49, 16]. The dipole interaction can dominate the behavior
of a quantum gas of these molecules and lead to intriguing many-body phenomena [24, 107, 108, 109]. Ultracold polar molecules also provide insights into chemistry at the quantum level and have the potential to be used as sensitive probes for variations of fundamental constants or as the basis of quantum computation schemes [49, 110].

Most experimentally investigated quantum gas mixtures of two elements consist of two alkali metals [111, 112, 113, 114, 115, 116, 117]. Advances in producing quantum degenerate samples of Yb [118] and alkaline-earth elements [119, 81, 120] have led to efforts towards mixtures containing these elements and recently quantum degenerate Yb-Li mixtures were obtained [60, 59]. A driving force behind these efforts is the interest in quantum gases of polar molecules beyond alkali dimers, such as RbYb [58], LiYb [60, 59], or RbSr [121]. Contrary to alkali dimers, these open-shell molecules possess an unpaired electron, which provides them with a rich spin structure and a magnetic dipole moment. This property will enable new ways to design and control few- and many-body systems and could prove very useful to implement lattice-spin models [45], to suppress inelastic collisions [52], to imprint geometrical phases [122], or to study collective spin excitations [123].

In this Article, we present the realization of quantum gas mixtures composed of the alkali metal $^{87}\text{Rb}$ and the alkaline-earth metal $^{88}\text{Sr}$ or $^{84}\text{Sr}$. An essential ingredient of our experimental strategy is the use of sympathetic laser cooling [124, 125, 126] on a narrow linewidth transition, which allows us to reach a high phase-space density (PSD) for both elements before evaporative cooling. Strontium atoms are laser cooled on the narrow $^1S_0 \rightarrow ^3P_1$ intercombination line and act as a refrigerant for Rb confined in an optical dipole trap. During this sympathetic laser cooling stage, the PSD of Rb increases by a factor of more than 200, with only a 20% reduction of the Rb atom number. In less than 400 ms the PSD of both elements can reach 0.06. These very favorable conditions allow us to efficiently reach quantum degeneracy for both species by evaporative cooling and to create BECs with more than $10^5$ atoms per element. The ease of producing large quantum degenerate samples enabled by sympathetic narrow-line laser cooling, together with the large electric dipole moment of RbSr ground-state molecules of 1.5 Debye [127], make Sr-Rb quantum gas mixtures an ideal stepping stone towards the exploration of dipolar physics with open-shell molecules.

The organization of this paper is as follows. In Sec. 3.2, we present an overview of our scheme. Section 3.3 describes the loading of a cloud of Rb into an optical dipole trap, followed by the loading of Sr atoms into a narrow-line magneto-optical
3.2 Overview of the experimental strategy

To reach quantum degeneracy, most ultracold atom experiments rely on laser cooling followed by evaporative cooling. The latter process intrinsically leads to a loss of atoms. To minimize this loss, it is beneficial to develop laser cooling methods that are able to reach high PSDs. The main atomic transition for laser cooling of alkalis, such as Rb, is broad, on the order of several MHz. To achieve high PSDs by laser cooling of such species, one can apply sub-Doppler cooling techniques, such as polarization gradient cooling in optical molasses [128, 129, 130, 131, 132, 133], velocity selective coherent population trapping [134], Raman cooling [135], Raman sideband cooling [136], or narrow-line cooling on transitions to higher electronic states, which have linewidths down to the 100 kHz range [137, 138]. The best PSD achieved so far by laser cooling of an alkali is to our knowledge 0.03 [139]. As a member of the alkaline-earth family, Sr has a singlet/triplet electronic structure. Its $^1S_0 \rightarrow ^3P_1$ intercombination line has a width of only 7.4 kHz. It has been shown that simple Doppler cooling using this narrow line can reach temperatures as low as 250 nK [140]. A PSD of 0.1 has been reached by transferring Sr clouds from a narrow-line MOT into an optical dipole trap [141, 142]. Starting from such samples, evaporative cooling can produce BECs that contain 25% of the initial atoms, much more than the 1% typical for alkali BEC experiments. It is therefore tempting to transfer those excellent properties to other species, by using Sr as a cooling agent. Here we demonstrate the efficiency of sympathetic cooling of Rb with Sr atoms laser cooled on a narrow line and the formation of dual-species BECs by a consecutive evaporative cooling stage. Our procedure to achieve quantum degeneracy can be divided into three main stages, see Fig. 3.1(a). During the first, “preparation” stage (Sec. 3.3) we use well-established cooling and trapping techniques to prepare ultracold samples of Rb and Sr. We accumulate Rb atoms in a magneto-optical trap and transfer them into a single beam optical dipole trap, henceforth referred to as the “storage” trap. After having stored Rb, we operate a “blue” Sr MOT, thereby accumulating metastable Sr atoms in a magnetic trap. We then optically pump Sr back to the
Figure 3.1: Timing of the experimental sequence and trap configurations used to produce a $^{88}\text{Sr}-^{87}\text{Rb}$ double BEC. (a) Timing sequence. The central sympathetic laser cooling of $^{87}\text{Rb}$ by $^{88}\text{Sr}$ shaded in gray is characterized in Fig. 3.2. (b,c) Dipole trap configurations and atomic clouds at the end of the preparation stage (b) and during the evaporation stage (c) (not to scale).
3.3 Preparation of an ultracold sample of rubidium and strontium

In this Section, we describe our experimental setup and the preparation of an ultracold mixture consisting of Rb contained in the storage trap and Sr stored in a narrow-line MOT, see Fig. 3.1(b). Our experimental setup is based on our Sr BEC apparatus, which has been described in detail in [142, 84]. The basic principle of the apparatus is to capture a Zeeman slowed atomic beam in a MOT and to cool the gas to quantum degeneracy by evaporation out of a dipole trap. Here we will focus on the upgrades carried out to also trap and cool Rb with the apparatus. Copropagating atomic beams of each element are produced by two independent ovens, heated to 550°C for Sr and 200°C for Rb [92, 84]. The same Zeeman slower (ZS) is used to slow both beams. Since the magnetic fields required to slow Rb and Sr differ significantly, we perform a time-sequential loading scheme. The Rb ZS laser beam is superposed with the Sr ZS beam by a dichroic mirror, and uses 14 mW of light detuned by $-165$ MHz from the $^2S_{1/2} \rightarrow ^2P_{3/2}$, $|F = 2\rangle \rightarrow |F' = 3\rangle$ transition. A “repumping” beam with 4.5 mW of power, addressing the $|F = 1\rangle \rightarrow |F' = 1\rangle$ transition is overlapped with the ZS beam. All three laser beams have a waist of about 8 mm at the MOT position. Slowed atoms are captured by a 3D MOT. Producing an ultracold mixture of Rb and Sr requires the use of MOTs of three wavelengths, one for Rb and two for Sr. The Rb MOT uses 780-nm light, the blue Sr MOT is operated on the broad $^1S_0 \rightarrow ^1P_1$ transition at 461 nm, and the red MOT on the $^1S_0 \rightarrow ^3P_1$ intercombination line at 689 nm. MOT beams of the three wavelengths are overlapped by dichroic mirrors on each of the three retro-reflected MOT beam paths. The Rb MOT beams have a waist of 9.8 mm, a power of 18 mW.
(25 mW) in the horizontal (vertical) direction, and are detuned by $-16$ MHz from the $^{2}\text{S}_{1/2} \rightarrow ^{2}\text{P}_{3/2}, |F = 2\rangle \rightarrow |F' = 3\rangle$ transition. A repumping beam with a waist of 6 mm and a peak intensity of $130 \mu W/cm^2$, on resonance with the $|F = 1\rangle \rightarrow |F' = 2\rangle$ transition, is shone onto the MOT. The quadrupole magnetic field of the Rb MOT has a gradient of 12 G/cm along the vertically oriented coil axis.

The experimental sequence starts by operating the Rb MOT during 20 s to accumulate a cloud of $2 \times 10^7$ atoms at a temperature of $175 \mu K$. We then compress the cloud in 140 ms by raising the gradient of the quadrupole field to 50 G/cm and increasing the MOT laser detuning to $-30$ MHz. After compression the $1/e$ cloud radius is $\sim 250 \mu m$. To decrease the temperature we use polarization gradient cooling in an optical molasses. After switching off the magnetic field, the MOT laser detuning is set to $-110$ MHz and the beam power is halved. After 3 ms of molasses, we obtain $1.8 \times 10^7$ atoms cooled to $15 \mu K$ with a peak density of $3 \times 10^{10} \text{cm}^{-3}$.

We then transfer the Rb atoms into the storage trap. This trap consists of a horizontal beam with a waist of $40 \mu m$ propagating at a small angle to the x-direction, see Fig. 3.1(b). The beam is derived from a 100-W multimode fiber laser operating at a wavelength of 1070 nm (YLR-100-LP-AC-Y12 from IPG). It is linearly polarized in the vertical direction to minimize the light shift induced on the red Sr laser cooling transition [142]. Initially we use a power of 14 W for the storage trap, which results in a potential depth of $k_B \times 830 \mu K$ and trap frequencies of $f_{\text{rad}} = 2.2$ kHz and $f_{\text{ax}} = 13$ Hz in the radial and axial directions respectively. To improve loading of the storage trap, we toggle the repumping beam to a path were a wire is imaged onto the trap region, creating a dark spot. The repumping beam power is reduced to a peak intensity of $8 \mu W/cm^2$ and in 500 ms we transfer 10% of the molasses atoms into the trap, pumping them at the same time into the $F = 1$ manifold. Up to $1.7 \times 10^6$ atoms are stored in the storage trap at a density of $2 \times 10^{13} \text{cm}^{-3}$ and a temperature of $\sim 20 \mu K$.

Having stored Rb, we now capture Sr atoms. The loading and cooling of Sr is done in a manner similar to our previous work [81, 142]. We operate a blue MOT on the broad transition at 461 nm, which has a leak towards the metastable $^3\text{P}_2$ state. We accumulate $^3\text{P}_2$ atoms in the magnetic trap formed by the quadrupole field of the MOT. We typically load this reservoir in a few seconds with several million $^{88}\text{Sr}$ atoms. For

Since the data presented here was taken, we have improved the atom number of the Rb MOT to $2 \times 10^8$. A further increase of the atom number would most likely be possible by using a six-beam Rb MOT.
the data presented in Sec. 3.3 and 3.4 we load for 0.5 s. The atoms are subsequently optically pumped back to the ground state using a flash of light on the $^3\text{P}_2 \rightarrow ^3\text{D}_2$ transition at 497 nm. The atoms are captured by a red MOT operating on the $^3\text{S}_0 \rightarrow ^3\text{P}_1$ intercombination transition and using a magnetic field gradient of 1.8 G/cm. The narrow, 7.4 kHz-linewidth intercombination transition allows us to cool Sr to less than 1 µK while keeping millions of atoms. The atoms settle in the lower part of an ellipsoid of constant magnetic field magnitude, see Fig. 3.1(b). The size and position of this cloud can be influenced by the magnetic field and the detuning of the MOT light. Varying these parameters facilitates the transfer of Sr into the optical dipole trap.

At the end of the preparation stage, we obtain $7 \times 10^6$ $^{88}\text{Sr}$ atoms at a temperature of 2.5 µK in a red MOT. The storage trap contains $1.6 \times 10^6$ Rb atoms at a temperature of 30 µK and a phase-space density of $2.5 \times 10^{-4}$, which is slightly worse than before loading the Sr atoms. The $1/e$ lifetime of the Rb cloud in the storage trap in presence of the blue $^{88}\text{Sr}$ MOT is 5.0(5) s. To detect the influence of Sr atoms on the Rb cloud in the storage trap, we perform the same experimental sequence, but shutter the atomic beam off after loading the Rb MOT. Under these conditions, the lifetime of the Rb sample in the storage trap is 30(1) s. This value does not change if also the Sr laser cooling beams are off.

### 3.4 Sympathetic narrow-line laser cooling

The following, sympathetic laser cooling stage is crucial in our approach to obtain a Sr-Rb double BEC. We make use of laser cooled Sr to further cool Rb and increase its PSD. To this aim, we overlap the cloud of $^{88}\text{Sr}$ atoms with the focus of the storage trap. This overlap is achieved by moving the magnetic field center upwards, while keeping the MOT laser frequency red detuned by 200 kHz from resonance. At this point, the Sr density is so low that the influence on the Rb cloud is negligible on a timescale of 500 ms. To increase the density of the Sr cloud, the magnetic field together with the MOT detuning and intensity are changed over 200 ms such that the cloud is compressed while remaining at the same position. The Sr atoms are loaded into the storage trap during this compression phase. The loading process is strongly influenced by the light shift induced on the red MOT transition by this trap. At the center of the trap, the light shift is $\sim +500$ kHz, which is almost 70 times the linewidth. In order to laser cool Sr atoms in the storage trap we tune the MOT laser frequency 350 kHz to the
FIGURE 3.2: Sympathetic laser cooling of $^{87}$Rb by $^{88}$Sr. During the compression phase the density of the red Sr MOT is increased and the Sr atoms are loaded into the storage dipole trap by changing magnetic field and both the MOT laser detuning and intensity. During the hold phase these parameters are held constant. (a,b) Evolution of the Rb and Sr atom numbers (a) and temperatures (b) as determined from time-of-flight absorption images. During a part of the compression phase the Sr cloud separates into two components, one being heated out of the system, the other remaining trapped and being transferred into the storage trap. We only show the number of trapped Sr atoms and the Sr temperature if we can determine them reliably from bimodal fits to the absorption images. (c) PSD of Rb, calculated from trap parameters and Rb atom number and temperature.
3.4. Sympathetic narrow-line laser cooling

blue of the unshifted atomic transition in a 200 ms ramp, while reducing the peak beam intensity to 6 µW/cm². The part of the Sr cloud that does not spatially overlap with the dipole trap is thereby expelled from the MOT, leading to a loss of 60% of the Sr atoms. This loss could be reduced by canceling the light shift of the transition, for example by inducing a light shift on the excited state of the laser cooling transition [89]. Since ⁸⁸Sr has a high natural abundance, leading to a high atom number in the MOT, we here simply tolerate the loss. The temperature of the Sr cloud in the storage trap increases to 15 µK before reducing to below 5 µK during the compression phase and a subsequent 200 ms hold phase. This temperature increase results from MOT dynamics in presence of the light shift induced by the dipole trap and would also occur in absence of Rb atoms. The Rb cloud thermalizes with laser cooled Sr by elastic collisions. During this process, the Rb PSD increases dramatically, by a factor of more than 200, reaching 0.062(6), while only 20(5)% of the Rb atoms are lost ². From the thermalization behavior we can deduce a minimum absolute value of the interspecies scattering length of 30 a₀ between ⁸⁷Rb and ⁸⁸Sr assuming pure s-wave scattering³ [145]. After the hold phase the Sr laser cooling beams are switched off. We observe that complete thermal equilibrium between Rb and Sr can only be reached in absence of the cooling light.

To prepare for the creation of a quantum degenerate sample, we transfer both elements into the science dipole trap, which has been described in detail in Ref. [142] and will be especially important for the creation of a large ⁸⁴Sr-⁸⁷Rb double BEC (see Sec. 3.5.2). This crossed-beam dipole trap is composed of an elliptical beam propagating in the x-direction with waists of \( w_z = 17 \mu m \) and \( w_x = 300 \mu m \), crossed by a nearly vertical beam with a waist of 90 µm, see Fig. 3.1(c). The center of this dipole trap is overlapped with the center of the storage trap, and the horizontal beams of the two traps intersect at an angle of 17°. Individual laser sources are used for each science trap beam (5-W, 1065-nm multimode fiber lasers, YLD-5-LP from IPG). At the wavelength of 1065 nm the polarizability of Rb is 2.9 times the polarizability of Sr [146, 147]. The ratio of trap depths is even higher because of gravitational sagging. Initially the science trap has a depth of \( k_B \times 25 \mu K \) for Rb and \( k_B \times 7 \mu K \) for Sr. The reduction of the Rb cloud volume and temperature by sympathetic laser cooling allows us to transfer Rb from the storage trap into this much shallower trap in 500 ms with a nearly perfect

²We estimate that the fraction of Sr atoms in the optically excited state is \( \sim 10\% \).
³The p-wave centrifugal barrier has a height of \( k_B \times 90 \mu K \) [143], rendering p-wave and higher order partial wave scattering negligible at our temperatures if shape resonances are absent [144].
transfer efficiency (more than 95% of the atoms). The $^{88}\text{Sr}$ transfer efficiency is 30%. After transfer we typically obtain $1.25 \times 10^6$ Rb atoms and $1.6 \times 10^6$ Sr atoms at a temperature of $1.2 \mu\text{K}$. The Rb PSD is 0.5(2) and the Sr PSD is 0.10(3). We attribute the increase in PSD to evaporation of Sr during the transfer process.

3.5 Evaporation to alkali/alkaline-earth double BECs

In this Section we present the creation of quantum degenerate mixtures of $^{87}\text{Rb}$ with either $^{88}\text{Sr}$ (Sec. 3.5.1) or $^{84}\text{Sr}$ (Sec. 3.5.2). These two Sr isotopes have markedly different properties, which we take into account in our experimental strategy. The $^{88}\text{Sr}$ isotope has a high natural abundance and provides us with an ideal coolant for sympathetic narrow-line laser cooling of $^{87}\text{Rb}$. Since the $^{88}\text{Sr}$ scattering length is negative ($-2a_0$), the $^{88}\text{Sr}$ BEC atom number is limited to a few thousand atoms. By contrast, $^{84}\text{Sr}$ has a convenient scattering length of $+123a_0$, allowing us to create BECs with high atom number. Unfortunately this isotope has a low natural abundance (only 0.56%), which renders it less favorable for sympathetic laser cooling. To overcome this drawback, we employ both isotopes in the production of a $^{84}\text{Sr}-^{87}\text{Rb}$ double BEC, using $^{88}\text{Sr}$ for sympathetic laser cooling of $^{87}\text{Rb}$ and $^{84}\text{Sr}$ for evaporative cooling to quantum degeneracy.

3.5.1 $^{88}\text{Sr}-^{87}\text{Rb}$ double BEC

To obtain a $^{88}\text{Sr}-^{87}\text{Rb}$ double BEC, we prepare as before a $^{88}\text{Sr}-^{87}\text{Rb}$ mixture in the science trap. Compared to the previous sections, we here increase the number of ultracold $^{88}\text{Sr}$ atoms by increasing the Sr MOT loading time to 5 s. Then we perform forced evaporative cooling by lowering the trap depth exponentially over 11 s to 0.5 $\mu\text{K}$ for Sr. As demonstrated in Sec. 3.4, the interspecies scattering cross section is sufficient for interspecies thermalization. Note that by itself $^{88}\text{Sr}$ does barely thermalize because of its small scattering length of $-2a_0$ [148, 149] and only the presence of Rb ensures proper thermalization. Since the trap depth is more than three times deeper for Rb than for Sr, Rb is sympathetically cooled by evaporating Sr. At the end of the evaporation stage, we obtain Rb and Sr BECs with low atom number immersed in thermal clouds (see Fig. 3.3). The Sr cloud contains $2.3 \times 10^3$ condensed atoms and $6.5 \times 10^4$ thermal atoms. The Rb cloud consists of a mixture of all three $m_F$
3.5. Evaporation to alkali/alkaline-earth double BECs

Figure 3.3: (Color online) $^{88}\text{Sr}^{87}\text{Rb}$ double BEC. The absorption images have been recorded after an expansion time of 26 ms. The lower panels show density profiles obtained by vertical integration of the absorption images. Bimodal fits consisting of a Gaussian and a Thomas-Fermi distribution are shown as red, solid lines. The Gaussian part of the fit corresponds to the thermal fraction of the cloud and is shown as blue, dotted line.
states of the $F = 1$ manifold, as confirmed by Stern-Gerlach measurements. The spin state distribution can be influenced by applying a magnetic field offset and gradient during the evaporation stage [150]. The distribution for zero offset and gradient is 38%, 25%, and 37% for the $m_F = -1, 0, +1$ states after evaporation. In total the Rb cloud consists of $1.3 \times 10^4$ atoms in the spinor BEC and $5 \times 10^4$ thermal atoms. The temperature of both elements is 190(30) nK. The low $^{88}\text{Sr}$ BEC atom number is expected since the negative scattering length of $^{88}\text{Sr}$ leads to a collapse of the BEC for higher atom numbers [151, 152, 153, 154, 142].

3.5.2 $^{84}\text{Sr}^{87}\text{Rb}$ double BEC

To obtain quantum degenerate samples with higher atom numbers, we now turn to the $^{84}\text{Sr}$ isotope. This isotope has a scattering length of $+123 \ a_0$ [148, 149], which is well-suited for evaporative cooling. Despite the small $^{84}\text{Sr}$ natural abundance of only 0.56% the production of large $^{84}\text{Sr}$ BECs with $10^7$ atoms has been demonstrated [142]. The low abundance can be compensated for by a longer blue MOT duration compared to the one used for the highly abundant $^{88}\text{Sr}$ isotope, leading to nearly the same atom number accumulated in the metastable state reservoir. By simply replacing $^{88}\text{Sr}$ with $^{84}\text{Sr}$ and operating the blue MOT for 20 s, we can prepare a $^{84}\text{Sr}^{87}\text{Rb}$ double BEC with essentially the same scheme as the one used for the production of a $^{88}\text{Sr}^{87}\text{Rb}$ double BEC. Nonetheless we have developed an improved strategy, which requires less time, makes optimal use of the precious $^{84}\text{Sr}$ atoms, and leads to much larger numbers of condensed atoms. Because of its high natural abundance, we use $^{88}\text{Sr}$ as the refrigerant for sympathetic laser cooling of Rb. We then use $^{84}\text{Sr}$ during the evaporation stage to obtain a $^{84}\text{Sr}^{87}\text{Rb}$ BEC.

We adapt the scheme of our experiment to this new strategy, see Fig. 3.4(a). During the Sr blue MOT stage we load both Sr isotopes, $^{88}\text{Sr}$ and $^{84}\text{Sr}$. This double-isotope loading is achieved by accumulating one isotope after the other in the metastable state reservoir. In between, the frequency of the 461-nm cooling laser source is changed by the isotope shift [155, 142]. We first accumulate $^{88}\text{Sr}$ for 500 ms and then add $^{84}\text{Sr}$ during 10 s. The lifetime of Rb atoms in the storage trap in presence of the $^{84}\text{Sr}$ blue MOT is 23(1) s. Afterwards both isotopes are optically repumped into the electronic ground state. The two isotopic clouds are captured simultaneously by two narrow-line red
3.5. Evaporation to alkali/alkaline-earth double BECs

MOTs, which contain $4 \times 10^6$ atoms of $^{88}\text{Sr}$ and $9 \times 10^6$ atoms of $^{84}\text{Sr}$ respectively, see Fig. 3.4(b) [155, 142]. The different isotopes are addressed independently by using two frequencies for the red MOT laser beams, separated by the isotope shift. Changing one of the MOT laser frequencies, vertically displaces the corresponding isotopic cloud and changes its radial size.

We proceed as described in Sec. 3.4 with the sympathetic laser cooling of Rb by $^{88}\text{Sr}$ and the transfer of the $^{88}\text{Sr}^{87}\text{Rb}$ mixture into the science trap. In the meantime we keep the $^{84}\text{Sr}$ cloud about 0.7 mm below the center of the dipole traps. At this position the red MOT has a radius of about 1 mm and a low density, which reduces light-assisted collisions. The lifetime of the MOT is independent of the various operations on the other species and we lose only 10% of the $^{84}\text{Sr}$ atoms during the sympathetic laser cooling stage (b) and after removing $^{88}\text{Sr}$ (c) (not to scale).
Figure 3.5: (Color online) Formation of a $^{84}\text{Sr} - ^{87}\text{Rb}$ double BEC while Sr is evaporated, sympathetically cooling Rb. (a) Absorption images of the Rb and Sr clouds at time $t$ of the evaporative cooling ramp recorded after 24 ms of expansion. Strontium condenses at $t = 4.3\, s$ and Rb at $t = 6\, s$. The temperature $T$ of Sr is given. (b) Density profiles obtained from the absorption images at $t = 6.8\, s$ by integration along the vertical direction. The solid, red lines are bimodal fits to the data by a Gaussian plus a Thomas Fermi distribution. The blue, dotted line shows the Gaussian part of the fit corresponding to the thermal part of the cloud.

cooling stage. Reciprocally, the presence of the $^{84}\text{Sr}$ red MOT does not affect the sympathetic laser cooling of Rb. It is advantageous to expel the refrigerant $^{88}\text{Sr}$ after it has fulfilled its role. Without the removal of $^{88}\text{Sr}$, the large scattering length between $^{84}\text{Sr}$ and $^{88}\text{Sr}$ of about $1700\, a_0$ [148, 149] would lead to strong three-body loss as soon as $^{84}\text{Sr}$ is loaded into the science trap. We expel $^{88}\text{Sr}$ by adiabatically lowering the science trap depth in 300 ms. Including gravitational sagging the trap is about ten times shallower for Sr than for Rb at the end of the ramp, leading to a removal of all $^{88}\text{Sr}$ atoms without affecting the Rb atom number. We then raise the trap back to its former depth in 100 ms. At this point we load $^{84}\text{Sr}$ into the science trap, see Fig. 3.4(c). We shift the red MOT upwards by changing the cooling laser frequency until the MOT overlaps with the science trap. Now 70% of the $^{84}\text{Sr}$ atoms are loaded into the dipole trap, compared to 40% when transferring into the storage trap as in Sec. 3.4. The reasons for this increased transfer efficiency are that the much shallower science trap induces a negligible light shift on the red Sr laser cooling transition and that the horizontally extended science trap is well adapted to the pancake shape of the red MOT. The $^{84}\text{Sr}$
cloud is slightly colder than the Rb sample, which again leads to sympathetic laser cooling of Rb. When choosing the final red MOT parameters, we have to compromise between attainable temperature and remaining Rb and Sr atom numbers [140, 126]. We chose a temperature of $1 \mu K$, for which we obtain $4.0(1) \times 10^6$ Sr atoms and $5.2(1) \times 10^5$ Rb atoms, both elements at a PSD of 0.4(1). At this point the cooling laser beams are switched off.

Starting with these excellent conditions, we perform evaporative cooling by lowering the science trap depth exponentially over 8.8 s to $k_B \times 150 \text{nK}$ for Sr. Strontium is evaporated, sympathetically cooling Rb, and a double BEC is formed (see Fig. 3.5). At the end of evaporation we obtain a pure $^{84}\text{Sr}$ BEC of $2.3 \times 10^5$ atoms and $1.3 \times 10^5$ quantum degenerate Rb atoms accompanied by $6.5 \times 10^4$ thermal Rb atoms at a temperature of $\sim 70 \text{nK}$. The Rb cloud again contains a nearly equal mixture of the three $F = 1 \ m_F$ states. The trapping frequencies in the x-, y-, and z-direction are at this point 40 Hz, 37 Hz, and 190 Hz for Sr and 67 Hz, 63 Hz, and 400 Hz for Rb. For the lowest values of the science trap depth, the gravitational sagging of the Sr cloud is $\sim 3.5 \mu \text{m}$ larger than the gravitational sagging of the Rb cloud. For comparison, the Thomas-Fermi radii, calculated neglecting the interspecies mean-field, is $2.5 \mu \text{m}$ for the Sr BEC in the vertical direction and $2 \mu \text{m}$ for the Rb BEC. The two elements are barely overlapping, which reduces interspecies thermalization. The differential gravitational sag between Sr and Rb could be compensated for the magnetic Rb $F = 1, \ m_F = +1$ or $m_F = -1$ state by using a magnetic field gradient in the vertical direction, which does not influence the non-magnetic Sr. The peak density of the Sr BEC and of each $m_F$-state component of the Rb BEC is $1.5 \times 10^{14} \text{cm}^{-3}$. The $1/e$ lifetime of the BECs is about 10 s. The atom number of the Sr BEC can be increased at the expense of the Rb BEC atom number by reducing the MOT loading time of Rb. On absorption images of $^{84}\text{Sr}-^{87}\text{Rb}$ double BECs taken after 24 ms of expansion, we observe that the position of the Sr BEC is shifting downwards for an increasing Rb BEC atom number. This observation hints at a positive mean-field interaction and therefore a positive interspecies scattering length between $^{87}\text{Rb}$ and $^{84}\text{Sr}$.

### 3.6 Conclusion and outlook

We have presented the production of $^{88}\text{Sr}-^{87}\text{Rb}$ and $^{84}\text{Sr}-^{87}\text{Rb}$ double BECs. Crucial to our success are the favorable interaction properties of the two mixtures. For both
mixtures we observe efficient thermalization. At the same time the mixtures do not suffer from large inelastic three-body losses. These interaction properties cannot be predicted by \textit{ab-initio} calculation and were completely unknown prior to our work.

A central stage in our scheme is sympathetic narrow-line laser cooling of Rb by Sr. This powerful technique will also be useful to cool other species besides $^{87}$Rb, including fermions, and should work if one of the four Sr isotopes has good interspecies scattering properties with the target species. It might even be possible to sympathetically laser cool the target species to quantum degeneracy without using evaporation [89]. This goal will be facilitated by selectively increasing the density of the target species with a species-specific dipole potential [99] or by using a target species of low mass, which leads to a high critical temperature or Fermi temperature for a given density.

Our next goal is the creation of RbSr molecules. We plan to associate atoms to weakly-bound molecules by either magneto-association [74] or stimulated Raman adiabatic passage (STIRAP) [156, 80]. These molecules will then be transferred into the ro-vibrational ground-state by STIRAP [157, 65, 38]. We are currently performing photoassociation spectroscopy of the $^{84,88}$Sr-$^{87}$Rb mixtures, in order to precisely determine the interspecies scattering lengths of all Sr-Rb isotopic combinations, the magnetic field values of Sr-Rb magnetic Feshbach resonances, and STIRAP paths for molecule association and ground-state transfer. The optimal Sr-Rb isotopic mixture for our task will depend on these properties, especially the interspecies scattering length, which determines the miscibility of the Rb and Sr quantum gases.
Chapter 4

Population of an optical lattice with heteronuclear doublons

The efficient creation of a double BEC of Rb and Sr with long lifetimes is a great success on the way towards high phase-space density samples of polar ground-state molecules. In order to use the ground-state molecules in a quantum simulation experiment, the molecules must have long lifetimes as well. Unfortunately, RbSr molecules are not stable. The chemical reaction \( 2\text{RbSr} \rightarrow \text{Rb}_2 + \text{Sr}_2 \) occurs on a fast timescale. Even if the molecules were chemically stable, the loss mechanism described in [47] would make quantum simulation experiments in 3D bulk samples of molecules difficult. The molecules have to be protected against the losses, which could be achieved by suppressing collisions between the molecules. In fact staking protective measures against lossy collisions already during the molecule association steps is desirable for the production of low entropy and high molecule number samples. Vibrational relaxation, that quenches the weakly-bound molecules produced in the first association step because of atom-molecule or molecule-molecule collisions, deteriorate the transfer efficiency of the second step producing ground-state molecules. Experimental efforts in producing bi-alkali ground-state molecules [38, 41, 43, 44] from ultracold atom mixtures in 3D-traps show relatively small production efficiencies.

The 3D optical lattice is an ideal tool to create molecules from pairs of ultracold atoms. In a deep lattice potential particles are trapped in individual potential sites like eggs in an egg-carton box. Tunnelling between the sites is suppressed and therefore molecules are protected against collisions. The lattice potential is generated from a
dipole trap laser beam that is retro-reflected onto itself. The trapping beam and the reflection will interfere and create a standing wave pattern trapping particles in equidistant anti-nodes\(^1\) of the standing wave. The dipole trap potential is cut into a sequence of “pancake”-shaped potentials. The particles can move freely in the plane of one flat lattice site. A second and third optical lattice beam in orthogonal configuration is necessary to immobilize the particles in all three directions. The tunnelling of particles between the lattice sites can be tuned continuously by changing the power of the lattice beams.

Ultracold bosons in an optical lattice are described by the Bose-Hubbard model. In the simplest case two parameters define the Bose-Hubbard Hamiltonian: on-site interaction and tunnel coupling. Both parameters can be controlled by the depth of the lattice. The tunnel coupling depends on the potential barrier between wells. The on-site interaction depends on the lattice oscillation frequency and the scattering length. The system exhibits two ground-states which occur in dependence of the strength of the on-site interaction compared to tunnelling. For large tunnel coupling the atoms are delocalized over the entire optical lattice potential. The ground-state is a superfluid described by a macroscopic wavefunction. When the tunnel-coupling is decreased such that on-site interaction dominates, then the atoms localize on the lattice sites with a fixed number of particles. The state cannot be described by the macroscopic wavefunction since coherence of the matter-wave is destroyed. The system undergoes a phase-transition to the Mott-insulator state.

Ultracold atoms in optical lattices is a platform to study many-body physics [6]. In this thesis we use the optical lattice as a tool towards the efficient creation of a low-entropy sample of weakly-bound molecules. The weakly-bound molecules are protected against lossy collisions when immobilized on the lattices sites. Moreover the optical lattice increases the local density of the atom pairs. The local density increase assists our optical association method [80].

### 4.1 Overlap of the Rb and Sr BECs

Our goal is to load the Rb-Sr double BEC into an optical lattice crossing the superfluid to Mott-insulator phase transition to immobilize the atoms. By using BECs instead of

\(^1\)A blue-detuned, retro-reflected beam will trap the particles at the nodes of the standing wave.
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**Figure 4.1:** The upper panels show the potentials of $^{84}$Sr (dashed) and $^{87}$Rb (solid) in gravity direction. The lower panels show the Thomas-Fermi density profiles of the two-component BEC of $^{84}$Sr (dashed) and $^{87}$Rb (solid). a) The two BECs are in the 1064-nm dipole trap and sag under gravity, b) the gravitational force is set to zero, c) the BECs are immiscible and the trapping potentials are similar, and d) the miscible BECs are held in similar trapping potentials.

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thermal clouds, we guarantee the population of the harmonic oscillator ground-state of each lattice well. Careful choice of the overall potential landscape, geometry and atom numbers of Rb and Sr BECs during the loading of the optical lattice creates a Mott-insulator state with as many lattice wells as possible, where the population is characterized by one Rb and one Sr atom in the ground-state of many lattice wells. The choice of this population is optimal for the association of molecules. If thermal clouds are loaded into the lattice, the control of the occupation number would be difficult.

The first consideration for this engineering of the Mott-insulator is the overlap of the Rb-Sr BECs. If the two species are spatially separated during the loading, then the Mott-insulator will contain zero doubly occupied sites. An ultracold mixture can be
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separated because of interspecies repulsion and different sag in the direction of gravity.

**Gravitational sag.** The atomic cloud trapped in a harmonic trap is offset by
\[ z_{\text{sag}} = -g/\omega_z^2 \]
in the direction of gravity. The offset depends on the trap frequency \( f_z = \omega_z/2\pi \) in z-direction, which is influenced by the depth of the potential and the mass of the atom. The depth of the optical potential can be vastly different for different atomic species, because the detuning in frequency between the main transition of the species and the wavelength of the trapping laser leads to different polarizabilities. Our two atomic species experience different dipole trap potentials because Rb has a 2.7 times larger polarizability than Sr at the wavelength of the dipole trap laser (1064 nm). The Rb potential is 3 \( \mu \)K deep and has trap oscillation frequencies of \((f_x, f_y, f_z) = (55 \text{ Hz}, 50 \text{ Hz}, 400 \text{ Hz})\). Sr is weaker confined with a trap depth of 1 \( \mu \)K and trap oscillation frequencies of \((50 \text{ Hz}, 45 \text{ Hz}, 240 \text{ Hz})\). Both gas clouds have a pancake shape with a Thomas-Fermi radius of \( R_{x,y} = 1.38 \mu \text{m} \) in the radial plane and \( R_z = 0.53 \mu \text{m} \) in the vertical direction for Rb, and \( R_{x,y} = 1.8 \mu \text{m}, R_z = 0.78 \mu \text{m} \) for Sr. Since the gravitational force is similar for both species, but the vertical trap oscillation frequency is significantly lower for Sr, the Sr BEC sags 3 \( \mu \)m more by gravity than the Rb cloud, leading to a complete separation of the two species, see Fig. 4.1a. This separation needs to be overcome in order to prepare a Mott insulator with sites that are occupied by both species.

The difference in gravitational sag of the two species can be tamed by employing a magic wavelength ODT trapping the two species with equal trapping frequencies. This approach was successfully demonstrated in the K-Rb mixture, where the differential gravitational sag because of the different masses was balanced by a 807 nm dipole trap \[158\]. Another approach to minimizing the differential gravitational sag is based on a combination of optical and magnetic trapping \[159, 160\]. This scheme works especially well for mixtures of non-magnetic (Sr, Yb) and magnetic atoms (e.g. Rb). The magnetic and non-magnetic species are trapped in an ODT. The position of the cloud of the magnetic atoms is adjusted by a magnetic field gradient, while the non-magnetic atoms stay at the same place.

**Interspecies repulsion.** The interspecies repulsion is characterized by the immiscibility parameter \( \Delta = g_1g_2/g_{12} \), which compares the intraspecies \( g_i = 4\pi\hbar^2a_i/m_i \) with the interspecies interaction \( g_{12} = 4\pi\hbar^2a_{12}/m_{12} \), where \( a_i \) and \( a_{12} \) are the corresponding scattering lengths and \( m_i \) are atomic masses and \( m_{12} = 2m_1m_2/(m_1 + m_2) \). If \( \Delta < 1 \),
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then the two BECs are immiscible. The ground-state configuration of an immiscible mixture exhibits phase separation. In bi-alkali systems the immiscibility parameter can be tuned with the help of a Feshbach resonance. Feshbach resonances between Rb and Sr are predicted to be narrow and possibly not usable for tuning the immiscibility parameter. Luckily the measurement of the interspecies scattering length, which will be presented in Chap. 5, revealed that the isotopic combination $^{87}$Rb-$^{84}$Sr is miscible. Using the $^{87}$Rb-$^{84}$Sr interspecies scattering length determined by us (see Chap. 5) and literature values for all other quantities [161, 148, 149], we find $\Delta = 1.3(1) > 1$. The phase-separation condition is not fulfilled and the BECs should therefore be miscible. In order to demonstrate the difference between our system and a phase-separated system, we show in Fig. 4.1c the density distributions for a hypothetical system with all parameters as ours, with the exception of the interspecies scattering length, which was set to $11.1 \times a_0$ instead of $91 \times a_0$ and the trapping potentials for Rb and Sr are set to be very similar. The larger interspecies scattering length corresponds to a miscibility parameter $\Delta = 0.986$. The two BECs mutually expel each other, overlapping only in a shell with the thickness of the healing length, about $0.2 \mu m$. The contrast between the hypothetical, immiscible system and the actual $^{87}$Rb-$^{84}$Sr BECs becomes evident by comparing Fig. 4.1c and d. In Fig. 4.1d the $^{87}$Rb-$^{84}$Sr BECs with interspecies scattering length of $91 \times a_0$ are stored in an optical potential, for which the trapping frequencies for both species are very similar. The density distributions now overlap over the extent of the BECs.

In the following I will report our attempts to overlap the Rb and Sr BECs. For the first attempt described in Sec 4.1.1 we tried to overcome the gravitational sag by employing a magnetic field gradient. With the help of the magnetic gradient the BECs were levitated on top of each other. Despite the fact that the BECs were brought closer spatially by the magnetic gradient force we observed only small overlap between the BECs. In the 1064-nm ODT the tighter trapped Rb BEC caused a ground-state configuration of the mixture with a small overlap by interspecies repulsion. For the second attempt described in Sec. 4.1.2 we used a dipole trap consisting of two beams that differ in wavelength. With the help of this bichromatic trap the trap frequencies in z-direction were balanced for the Sr and Rb BECs. The gravitational sag and interspecies repulsion were overcome at same time. In the last section of the chapter 4.1.4 we demonstrate the loading of an optical lattice from the bichromatic trap and obtain $3.1 \times 10^4$ doubly occupied sites. These Rb-Sr doublons serve as a starting point for the association of
weakly-bound molecules that can be transferred to the absolute ground state in the future.

4.1.1 Differential gravitational sag compensation by magnetic field gradient

The first method exploits the difference in magnetic moment of the non-magnetic Sr ground-state atoms and the Rb atoms in a magnetic Zeeman sub-states. A vertical magnetic field gradient does not influence the Sr, but can pull the Rb $m_F = -1$ BEC component downwards. A well adjusted field gradient will overlap the trap minima for those two species [159, 160]. Overlap can only be achieved for one of our three Rb $m_F$ component, since the non-magnetic Rb $m_F = 0$ component is not influenced by the magnetic field gradient and the $m_F = +1$ component, which has the opposite magnetic moment of the $m_F = -1$ component, is pushed upwards. The magnetic field gradient is created by the coils we use for the magneto-optical trap. The centre of the quadrupole field is positioned 2.5 mm below the BECs using an offset magnetic field. The low-field seeking Rb $m_F = -1$ BEC component is pulled in direction of gravity, towards the quadrupole centre, with increasing magnetic field gradient.
4.1. Overlap of the Rb and Sr BECs

To measure the interspecies overlap, we induce photoassociation (PA) loss on a RbSr molecular transition that is red detuned by 173.5(2) MHz from the Sr $^1S_0-^3P_1$ atomic transition. During the 200 ms PA pulse with an intensity of 5 W/cm$^2$ all Sr atoms are lost, either by PA with Rb or by heating through off-resonant scattering of PA beam photons and subsequent evaporation from the trap. To measure the population of the Rb $|F=1, m_F\rangle$ states individually, the magnetic sub-states are separated by a magnetic field gradient during free expansion. A detailed description of the Stern-Gerlach separation and PA spectroscopy techniques will be given in Chap. 5. The Rb sub-state atom numbers are measured by absorption imaging. We record the number of Rb $m_F=-1$ atoms that are left after the photoassociation pulse in dependence of the magnetic field gradient, see Fig. 4.2. Without a magnetic field gradient, no Rb atoms are lost by photoassociation, confirming that the Rb and Sr clouds do not overlap. Around a magnetic field gradient of 31.5 G/cm a weak PA loss feature appears. We were not able to increase the PA loss of $m_F=-1$ Rb atoms beyond 13%, which indicates that even when compensating the differential gravitational sag, the two species do not overlap well.

The reason for the limited overlap is found by self-consistently solving the Gross-Pitaevskii equations describing the Sr and Rb $m_F=-1$ BECs. This calculation includes the interspecies mean-field interaction, where we use an interspecies scattering length of $a_{\text{RbSr}} = 91 a_0^2$, which we deduce from two-colour PA spectroscopy combined with ab-initio molecular potential calculations [162]. A cut through the calculated density distributions along the vertical direction is shown in Fig. 4.1b. The stiffer trap for Rb compared to Sr leads to a larger Rb peak density in spite of the lower Rb $m_F=-1$ atom number. The interspecies mean-field potential that Rb exerts on Sr exceeds the chemical potential of the Sr BEC. The Sr BEC cannot penetrate into the centre of the Rb BEC and forms a hollow shell around it. Only 25% of the Rb atoms and 10% of the Sr atoms are in the region of spatial overlap of the two species. This small overlap renders PA inefficient, explaining the weak PA loss.

The simulations show that compensating the gravitational sag by means of a magnetic gradient force creates small overlaps. Fortunately the scattering properties of $^{87}$Rb and $^{84}$Sr renders the doubly degenerate gas miscible. For this miscible mixture

\[ a_0 = 52.917721067(12) \text{ pm} \]
a larger overlap is achieved by adapting the optical potentials for equal trapping frequencies. Our method of changing the trapping potentials independently will be presented in the following.

4.1.2 Trap frequency matching by bi-chromatic dipole trap

To better overlap the Rb and Sr BECs, we now engineer an optical dipole trap that has the same trap frequencies for the two species. Calculated density profiles of the two
4.1. Overlap of the Rb and Sr BECs

BECs in such a trap are shown in Fig. 4.1d. The equal vertical trap frequencies together with the similar mass of $^{87}\text{Rb}$ and $^{84}\text{Sr}$ lead to negligible differential gravitational sag. It is therefore unnecessary to apply a magnetic field gradient to overcome the sag, making it possible to overlap all three $m_F$ components of the Rb BEC with the Sr BEC. Reducing the Rb trap frequencies down to the value of the Sr trap frequencies lowers the Rb density, reducing the mean-field repulsion on the Sr BEC and making it possible for the Sr BEC to cover the whole volume of the Rb BEC. The Rb BEC has a higher peak density than the Sr BEC even for identical trap frequencies and atom numbers, because of the lower intraspecies mean-field interaction of Rb ($g_{\text{Rb}}/g_{\text{Sr}} = 0.77$). In order to equalize the trap frequencies of the two species, we create a bi-chromatic dipole trap by adding dipole beams with a wavelength of 532 nm. These beams create a repulsive potential $U_{\text{Rb}}$ for Rb and an attractive potential $U_{\text{Sr}}$ for Sr with a ratio $U_{\text{Rb}}/U_{\text{Sr}} = -2.9$. A 532-nm dipole trap beam is superimposed with each of the two 1064-nm dipole trap beams. The waists of the 532-nm beams are matched to within 10% to the waists of the co-propagating 1064-nm beams and the foci of all beams are aligned on a common spot. By increasing the power of the 532-nm beams, the Rb potential is reduced while the Sr potential is increased. If the 532-nm beam has 0.4 times the power of the 1064-nm beam, the same trap potential is obtained for Rb and Sr. At this power ratio we expect a good overlap between the Rb and Sr BECs. For illustration, calculated vertical Rb and Sr trap frequencies are shown in Fig. 4.3a in dependence of the horizontal 532-nm beam power. Here the power of the horizontal 1064-nm beam is kept constant at 190 mW, as will be the case for the PA loss measurement described below. With increasing 532-nm beam power, the trap frequency of Sr increases, whereas the Rb trap frequency decreases. At a power of 75 mW the two trap frequencies are the same. Above 400 mW, Rb will spill out of the trap under the effect of gravity. We characterize the improvement in Rb-Sr BEC overlap by measuring PA loss in dependence of the horizontal 532-nm beam power. The power of the vertical dipole trap beams are adjusted such that both species have the same trap oscillation frequency $f_y = 55$ Hz in the propagation direction of the horizontal beams. In any case, the matching of the trapping frequencies in the horizontal plane ($f_x, f_y$) is less important than in the gravity direction ($f_z$) for the increase of the overlap of the BECs.

The starting point of each PA loss measurement is a Rb BEC of $N_{\text{Rb}} = 4.0 \times 10^4$ together with a thermal fraction of $3.5 \times 10^4$ Rb atoms and a Sr BEC of $N_{\text{Sr}} = 1.5 \times 10^5$ atoms. In absence of a magnetic field, all three Rb $m_F$ components are coupled to RbSr molecular
states by the same, single-frequency PA beam. Therefore we now measure the total Rb atom number after pulsing the PA beam on for 300 ms. To measure the Rb atom loss by PA, we subtract two measurements for each setting of the horizontal 532-nm beam power. We first take a reference measurement during which the PA beam is detuned by five linewidths from the PA line (linewidth $\gamma = 470(30)$ kHz). During this measurement Rb is mainly lost by mechanisms other than PA, for example spilling out of the bi-chromatic trap because of reduced trap depth or evaporation from the trap after being heated by collisions with Sr, which in turn is heated by off-resonant scattering of PA beam photons on the Sr $^1S_0-^3P_1$ atomic transition. During the second measurement the PA beam is resonant with the molecular transition, leading to additional loss of Rb by PA if the Rb and Sr clouds are overlapped. Figure 4.3b shows the difference of Rb atom number between these measurements in dependence of the horizontal 532-nm beam power. At zero power, no PA loss is observed because the two species do not overlap, see Fig. 4.1a. The PA loss reaches a maximum of $4 \times 10^4$ atoms at a power of 75 mW before decreasing again to zero above a power of 200 mW. To understand this behavior we show calculated vertical trap frequencies of Rb and Sr in dependence of the horizontal 532-nm beam power in Fig. 4.3a. With increasing 532-nm beam power, the trap frequency of Sr increases, whereas the Rb trap frequency decreases. The power for which the two trap frequencies are the same coincides with maximum PA loss, confirming our expectation that similar traps for Rb and Sr lead to good overlap of the two clouds. For higher powers, the Sr cloud size shrinks whereas the Rb cloud size increases, leading to less overlap and reduced PA loss. Above 400 mW, Rb will spill out of the trap under the effect of gravity.

To obtain a better understanding of the PA loss feature, we numerically calculate the cloud overlap

$$c = \frac{1}{N_{Rb}N_{Sr}} \int n_{Rb}(\vec{r})n_{Sr}(\vec{r})d\vec{r},$$

(4.1)

where $n_{Rb}$ and $n_{Sr}$ are the density distributions of the two clouds. This cloud overlap is proportional to the PA rate. In a first attempt of this calculation, we use BEC distributions determined from coupled Gross-Pitaevskii equations for $n_{Rb}$ and $n_{Sr}$, see appendix. The PA rate calculated from these distributions is shown in Fig. 4.3b (red solid) and is highest if the trap is equal for both species, as expected. However the width of the PA rate peak is much smaller than the experimentally determined loss.
4.1. Overlap of the Rb and Sr BECs

The discrepancy is firstly due to the presence of the thermal Rb cloud accompanying the Rb BEC. The thermal cloud has a larger extent than the BEC leading to PA loss for a larger range of bi-chromatic trap parameters. Secondly, during the PA pulse the thermal fraction increases and the Sr BEC heats up contributing to the broader overlap range in Fig. 4.3b. The Sr BEC heats up because of off-resonant scattering of PA beam photons on the Sr $^1S_0-^3P_1$ transition. If the Rb cloud overlaps with the Sr cloud, Rb is heated by elastic collisions with Sr. Indeed, TOF absorption images show a large thermal component with $T = 75 \pm 20 \text{nK}$ for both BECs after the photoassociation pulse. In a second attempt to describe the PA loss feature, we therefore use thermal distributions of 50 nK temperature for Rb and Sr. This temperature is an estimate for the average temperature during the PA pulse. The resulting PA rate, scaled to match the data points best, is shown in Fig. 4.3b as dashed line. The match between the experimental data and the calculated thermal cloud overlap alludes that shorter photoassociation pulses could be used to reduce heating such that the overlap of the BECs is not overestimated. Nevertheless for similar photoassociation pulse durations and intensities the optically balanced mixture has a five fold increase in atom loss during photoassociation than the magnetically overlapping BECs in the 1064 nm-trap. If the atom loss was due to thermal atoms solely, we would not observe improvement.

Since the Rb BEC is accompanied by a thermal component and the Rb and Sr BECs are heated by the PA light, the observed PA loss is not a strong indication for miscibility of our BECs. The overlap measured by PA loss is overestimated because the thermal components have a larger spatial extent in the trap penetrating each other. We will show a much stronger indications in the following by strong damping of out-of phase oscillations between the two BECs.

4.1.3 Collective dipole oscillations of the two-component BEC

The excitation of collective modes and measurement of their eigenfrequencies is an important tool for investigating the interaction properties of many-body systems. The collective excitation measurements were applied to the investigation of spin-orbit coupling [163] and dipole-dipole interaction [164] in BECs for example. Collective dipole oscillations are simple centre-of-mass oscillations of the atomic cloud. In the simplest case, the dipole oscillations occur at the trapping frequencies. In a mixture the dipole oscillations change because of the interspecies interaction - the collective modes and
Chapter 4. Population of an optical lattice with heteronuclear doublons

Figure 4.4: Time series of dipole oscillations for two different potential landscapes. 

a) The dipole oscillations are excited in the 1064 nm ODT.

b) Dipole oscillations in the bi-chromatic trap with comparable trapping frequencies in gravity direction.

frequencies vary across the miscible-immiscible transition [165]. When the ground state consists of two phase-separated BECs the oscillation is underdamped. The BECs flow frictionless through each other without breaking up the interspecies boundaries because of the high surface tension (compare Fig.4.1b). In contrast the interspecies interaction in a miscible two-component BEC acts in the bulk. The oscillation frequency changes because of the interspecies mean field. Also the oscillation is damped because the center-of-mass motion dissipates into sound-waves. High enough relative velocities facilitate a counterflow instability [166, 167].

We excite dipole oscillation for two conditions. First the trapping potential of Rb and Sr are different, because only the 1064-nm trap is on. The oscillations are excited by modulating the intensity of the trap laser for five periods. After the excitation we let the BECs evolve in the trap and take absorption images with a time-of-flight of 22 ms. Figure 4.4a shows the time series of the oscillations of Rb and Sr BECs in the imbalanced trap. In the imbalanced potential the dipole oscillation is excited by intensity modulation of the horizontal 1064-nm beam at the Sr oscillation frequency (244 Hz). Mainly the Sr cloud oscillates here since the Rb trap oscillation frequency (400 Hz) is far off resonance. The sinusoidal centre-of-mass motion in gravity direction is seen clearly. The Sr BEC features a low-density region in the middle for almost all of the absorption images. The depletion of Sr from the middle can be attributed to the repulsion of the dense Rb BEC assembled in the stiff IR trap. The surface tension between the two components is not overcome on the timescale of the measurement and the two BECs
4.1. Overlap of the Rb and Sr BECs

flow through each other during an oscillation period. Although the mixture is miscible, the mean-field potential of Rb BEC prevents the Sr from deeply penetrating the high-density Rb BEC. For the next series of experiments the trapping potentials are adjusted to be roughly equal using the Rb-anti-trapping 532 nm-beam. The Rb trapping frequency is 370 Hz and the Sr frequency is 355 Hz. The oscillations in this balanced trap are excited by abruptly (in 0.2 ms) switching on the 532-nm beam. The fast change of the trapping potentials leads to an acceleration of the clouds in gravity direction. The BECs start to oscillate. Figure 4.4b shows the time series of the oscillations of the fully overlapping BECs in the balanced trap. The Sr BEC does not show the density depletion in the middle of the cloud as in the case before. Instead both BECs have a similar profile. The sizes of the BECs increases over the course of the measurement and the elongated shape of the initial BECs turns into a potato. The size increase and deformation of the BECs could be explained by the excitation of sound-waves from the counterflowing BECs. The turbulent, modulated density of the BECs appears as a size increase and deformation in the TOF density profiles.

If the interspecies scattering was larger than 110 a₀ as was simulated for the density profile shown in Fig. 4.1c, the dipole oscillations would show a similar behaviour as shown in Fig. 4.4a. The interspecies repulsion would create density-deplete regions in the Sr BEC even when the trapping potentials were balanced. The qualitatively different dynamics of the double BECs shows however that the Rb-Sr BECs are miscible. As will be shown in the next section, the bi-chromatic trap enables us to load the double BEC into an optical lattice with the result of lattice sites occupied by one Rb and one Sr atom. Such configuration is an optimal starting point to create ultracold molecules.

In conclusion, we realize overlapping Rb-Sr BECs by means of two overlayed far-detuned dipole traps. The first dipole trap beam creates an attractive potential for both species, because its wavelength is 1064 nm hence red-detuned from the main transitions in Rb and Sr. The potential for Rb is almost three-fold deeper than for Sr leading to a large differential gravitational sag. The ground-state configuration of the double BEC yields minimal spatial overlap even when the gravitational sag is compensated by magnetic forces. The imbalance of the trapping potential results in a high-density Rb BEC, which expels the Sr BEC from the centre of the trap. The minimal overlap between Rb and Sr caused by the gravitational sag as well as the repulsive mean-field potential of Rb are remedied by the second ODT beam. Its wavelength is 532 nm, which is red-detuned from the main transition in Sr but blue-detuned for Rb. This
beam can be used to tune the trapping frequencies for the two species independently. The overlapping BECs are trapped in far-detuned beams leading to long lifetimes of the condensates. A nice property of our technique is that it does not rely on magnetic trapping and is therefore applicable to all magnetic sub-states of the Rb BEC. In addition both dipole trap wavelengths are far detuned from all transitions in Rb and Sr, leading to negligible off-resonant scattering. In some other relevant mixtures (e.g. Rb-K) a dipole trap with opposite polarization for each species and a trap depth of several µK would lead to significant off-resonant scattering. Finally high power lasers at the wavelengths of both beams of the bi-chromatic trap are standard equipment in the lab.

4.1.4 Two-component lattice gas

Now the overlapping double BEC can be transferred into the optical lattice. We have optimized the loading sequence to populate the lattice with roughly $3.1 \times 10^4$ doubly occupied sites. The number of doubly-occupied sites is measured with the help of one-colour PA.

The lattice setup was described in Sec. 2.2.8. The lattice laser wavelength is 1064 nm and has a maximum output power of 48 W. In the vertical plane the lattice is formed by two focused beams each about 45° with respect to gravity, which we call diagonal
4.1. Overlap of the Rb and Sr BECs

A third focused beam is in the horizontal plane nearly orthogonal to the diagonal beams. The horizontal lattice beam and the horizontal ODT beam are at an angle of $15^\circ$. The Gaussian beam waists at the focus of the diagonal beams are $208(7) \mu m$ and the horizontal beam waist is $78(5) \mu m$. The power of the lattice laser is distributed across the lattice beams in such a way that the lattice depth is the same for the three beams at maximum RF power of the AOMs (see Sec. 2.2.8). The lattice trap depths we reach with the available power of the laser are $U_{3Dlattice} = 460 E_R$ for Rb and $150 E_R$ for Sr. The lattice trap depth is calibrated using the Raman-Nath diffraction method [98].

The sequence of loading atoms into the lattice starts with a double BEC of $10^5$ Rb atoms and $2 \times 10^5$ Sr atoms in the 1064-nm trap. In order to have a controlled initial state for molecule creation the Rb BEC is in the magnetic sub-state $m_F = 0$. The pure $m_F = 0$ Rb BEC is prepared by applying a magnetic field gradient during forced evaporation, which expels the $m_F = \pm 1$ components from the trap. After evaporation the 532-nm beam is switched on in 200 ms creating a trapping potential with trapping frequencies of 535 Hz in gravity direction and 50 Hz in the horizontal plane for both species. The three lattice beams then are ramped up with equal powers in three stages. The first stage occurs in 300 ms ramping on the lattice to a depth of $13 E_{R,Rb}$ for Rb and $4.3 E_{R,Sr}$ for Sr. The lattice depth for the Rb atoms is close to the literature value of the super-fluid to Mott-insulator transition of $12 E_{R,Rb}$ [6]. After the first ramp the Rb is in the Mott-insulator phase. Although the presence of a second species shifts the critical depth [168] of the super-fluid to Mott-insulator transition, the absence of phase coherence in the Rb cloud confirms that the Rb is in the Mott-insulator phase. The phase coherence of the quantum gases can be probed by absorption imaging after TOF. While the Sr cloud exhibits a typical diffraction pattern when it is released suddenly from the lattice, the Rb diffraction pattern is absent.

During the second ramp stage the lattice depth is increased to $46.7 E_{R,Rb}$ and $15 E_{R,Sr}$ in 100 ms. After the second ramp the Sr is in the Mott-insulator phase, which is proven by absorption imaging again. Both atomic clouds do not exhibit a diffraction pattern. Both atomic species are localized on the sites of the optical lattice. In the final ramp the lattice depth is increased to $460 E_{R,Rb}$ and $150 E_{R,Sr}$ in 100 ms and the atoms are deep in the Mott-insulator regime with a negligible tunnelling rate.

The initial atom numbers of the BECs prior to lattice loading are optimized on the PA signal amplitude shown in Fig. 4.5. The Rb-Sr mixture is loaded into the lattice of $460 E_{R,Rb}$ (150 $E_{R,Sr}$) depth and illuminated with a PA pulse of 10 ms duration on a
molecular resonance. The molecular state that we address during the PA pulse has a binding energy of 174 MHz with respect to the atomic threshold of the $^3P_1$ state of Sr. The frequency of the PA laser is varied across 300 kHz around the molecular resonance. The Rb-Sr atom pairs are associated into molecules resulting in loss. The Sr atom number decreases from $5 \times 10^4$ to $2 \times 10^4$ atoms due to PA. The Rb number also decreases by $3 \times 10^4$.

The sample in the deep optical lattice decays on three timescales. First an initial decay in less than 1 ms occurs. The Rb number drops by $2.5 \times 10^4$ while the Sr atom number remains the same. We attribute the initial loss to decay of high-number occupation lattice sites. Before lattice loading the Rb BEC density in the balanced ODT is higher as shown in Fig. 4.1d. The higher density leads to the population of high-number lattice sites in the centre region of the Mott-insulator. The sites with an occupation number larger than two decay because of three-body recombination. The Sr BEC is not as dense, which leads to fewer lattice sites with high-number occupation in the Mott-insulator. The fast initial loss can be decreased by loading a smaller number of Rb atoms into the lattice. After this initial decay of Rb, we observe a second decay process with a $1/e^2$ of $130(30)$ ms. Rb and Sr decay by $3 \times 10^4$ atoms. The number of atoms decaying on that timescale is consistent with the number of lost atoms after a PA pulse was applied (see Fig. 4.5). If we apply the PA pulse after loading the lattice and waiting for 150 ms no PA loss is observable. This is why we assume that this decay process involves sites occupied by Rb and Sr atoms. The decay of sites containing both atomic species could be due to the absorption of lattice laser photons. The lattice laser with an energy of 9398.5 cm$^{-1}$ could very well excite Rb-Sr atom pairs to a vibrational level in the $1(\Omega = 3/2)$, $2(\Omega = 1/2)$ and $3(\Omega = 1/2)$ molecular potentials shown in Fig. 5.3.

A third decay process is on the order of seconds corresponding to single-species lifetimes in the optical lattice limited by background-gas collisions and off-resonant scattering of photons.

### 4.2 Conclusion and outlook

In summary we created a overlapping two-component BEC of $^{87}$Rb and $^{84}$Sr by means of a bi-chromatic dipole trap. The trap features selective anti-trapping beams balancing the trap frequencies for the two species. The BECs have significant overlap only
when the potential landscape is similar for both species. The presence of overlap is confirmed by PA loss measurements and the observation of qualitatively different dipole oscillation dynamics. The miscible $^{87}\text{Rb}-^{84}\text{Sr}$ mixture exhibited complex dynamics mediated by interspecies interactions. With higher imaging resolution turbulence and vortex formation could be investigated. The miscible two-component BEC of $^{87}\text{Rb}$ and $^{84}\text{Sr}$ is transferred into an optical lattice potential resulting in up to $3.1 \times 10^4$ doublons. Such a sample is an ideal starting point to proceed with optical [80] or magnetic [162] association steps creating paramagnetic polar ground-state molecules.
Chapter 5

Photoassociation spectroscopy

The main goal of the experiments described in the following is identifying a pathway for the creation of weakly-bound molecules as an intermediate step towards RbSr molecules in the rovibrational ground-state. In the case of bi-alkali ground-state molecule production this intermediate step usually involves magneto-association. A magnetic sweep across a Feshbach resonance of two alkali atoms creates molecules close to the dissociation threshold. The weakly-bound molecules then are transferred to the absolute ground-state with the help of further association steps.

Because of the absence of electronic spin in ground-state Sr atoms, Feshbach resonances between Rb and Sr atoms are predicted to be impractical for the magneto-association of weakly-bound molecules [74]. The alternative strategy involves coupling the free-atom pair to a molecular state with resonant laser light in an incoherent [169] or in a coherent manner. The latter approach is based on the use of STIRAP [170] and was successfully applied in our experiment to produce Sr$_2$ molecules in the electronic ground state [80]. Before weakly-bound RbSr molecules can be created via STIRAP the binding energies of the molecular states involved in the STIRAP sequence must be known. Ab-initio calculations of the binding energies are not precise enough for that purpose. Therefore the energies of molecular states relevant to the molecule association technique have been determined by us using photoassociation spectroscopy (PA). The PA was performed of the mixtures $^{87}$Rb$^{84}$Sr and $^{87}$Rb$^{88}$Sr. The favourable scattering properties of $^{84}$Sr allow us to create double BECs of $^{87}$Rb$^{84}$Sr with high atom numbers and a Mott-insulator with many doubly occupied sites. The isotopic combination $^{87}$Rb$^{84}$Sr therefore was chosen to be used to create ultracold molecules. Because of the negative scattering length of $^{88}$Sr only small atom number BECs of $^{87}$Rb$^{88}$Sr were created (see Chap. 3). This isotopic combination is not suitable for molecule creation. Nevertheless PA spectroscopy on $^{87}$Rb$^{88}$Sr gives us valuable information about
the properties of RbSr molecules.

In the following I will summarise briefly the most important theoretical considerations concerning diatomic molecules and the state-of-the art knowledge about RbSr molecules. Then the most important aspects of PA spectroscopy and their implementation in the experiment will be explained. Finally I will present the PA results. The results of the PA measurements will be presented in the following order. First the one-colour spectra of the mixtures $^{87}\text{Rb}^{88}\text{Sr}$ and $^{87}\text{Rb}^{84}\text{Sr}$ are shown. Secondly, the spectra of the electronic ground-state of $^{87}\text{Rb}^{88}\text{Sr}$ then $^{87}\text{Rb}^{84}\text{Sr}$ accessed through two-colour PA will be presented. Two conclusions drawn from the results will be presented at the end of this chapter. Firstly I will explain why the attempts creating weakly-bound molecules by STIRAP were unsuccessful with the chosen isotopic combination and the molecular states found. Secondly, the spectroscopy data was used to determine the interspecies scattering properties of Rb and Sr and prediction are made towards a more favourable isotopic combination ($^{87}\text{Sr} - ^{87}\text{Rb}$) for the successful creation of weakly-bound molecules by STIRAP.

5.1 Diatomic molecules

The literature on diatomic molecules is vast [171, 76]. Here I will touch on the most essential theoretical concepts of diatomic molecules.

5.1.1 Molecular structure

The first, essential step in solving the Schrödinger equation of a molecule, consisting of nuclei and electrons, is to separate the electronic from the nuclear motion. This so-called Born-Oppenheimer approximation is based on the difference in mass between electrons and nuclei. The electron cloud is assumed to follow quasi-instantaneously the motion of the much heavier nuclei. First, the electronic part of the Hamiltonian is solved for fixed internuclear distance $R$. The solutions, the electronic wavefunction and energy, is parametrized by $R$. Second the nuclear Hamiltonian can be solved after the potential energy of the nuclei is completed with the $R$-dependent electronic energy. The potential which the nuclei are subject to consists of the nuclear coulombic repulsion and the electronic energy. A typical potential energy of the nuclei is shown in Fig. 5.1. For small internuclear distances the coulombic repulsion dominates. For
5.1. Diatomic molecules

large distances the potential energy vanishes. At intermediate distances the potential supports a series of bound states, corresponding to vibrational states of the nuclei.

To describe the shape of the potential a set of parameters, which are called spectroscopic parameters, is introduced. The distance \( R \) between the nuclei at minimum energy is called equilibrium distance \( r_e \). The depth of the potential \( D_e \) is defined as the difference in energy between the potential at infinite \( R \) (dissociation threshold) and \( r_e \). Around its minimum the potential can be approximated by a harmonic potential with vibrational frequency \( \omega_e \). The similarity of the potential around \( r_e \) with a harmonic potential gives rise to equally spaced vibrational states of the nuclei with energy \( E(\nu) = \omega_e \left( \nu + \frac{1}{2} \right) \), where \( \nu \) denotes the vibrational quantum number. The majority of spectroscopic methods as well as ab-initio calculations access the region close to \( r_e \) [171]. For large nuclear distance the harmonic approximation breaks down and the molecular potential can be approximated by

\[
V(R) = D_e - \sum_{n=1}^{\infty} \frac{C_n}{R^n}
\]

and additional dispersion coefficients \( C_n \) need to be determined to predict energy levels across the full range of internuclear distances correctly. The terms of different \( n \) in the sum originate from different interaction effects [171]. Terms of order \( n = 1,2,4 \) arise when one or both atoms in the molecule are charged. The \( n = 3 \) term originates from dipole-dipole interactions in homonuclear molecules. If one atom is in the ground-state and the other atom is electronically excited, then the exchange of the excitation gives rise to the dipole-dipole long-range term in the potential [172]. The quadrupole-quadrupole interaction between atoms [173] leads to the \( n = 5 \) term. The molecular potentials considered in our studies dissociate to thresholds of atom pairs with either Rb or Sr in the ground state. The S states of the Sr and Rb atoms are spherically symmetric rendering quadrupole-quadrupole interactions negligible. The main contribution to the long-range branch of the potentials in our spectroscopy studies is the term with \( n = 6 \). The spectroscopic region near the dissociation threshold is very well accessible with PA spectroscopy. The \( C_6 \)-coefficient can be retrieved from the PA spectroscopic data.

In addition to vibrations the molecule can rotate around an axis perpendicular to
Chapter 5. Photoassociation spectroscopy

the internuclear axis. For a simple rigid rotor model the energy levels of rotation follow the series \( E_{\text{rot}} = BR(R+1) \) with the rotational constant \( B \) and rotational quantum number \( R \).

Hund’s cases

Over the course of the last hundred years ever finer spectroscopic resolution necessitated taking more intricate effects inside the molecules into account to model experimental data. The simple mechanical picture of diatomic molecules described above has to be extended by the spin of the electrons and nuclei as well as their interactions with the orbital angular momentum, rotational motion and each other. Depending on the interaction strength one chooses different basis sets to simplify solving the coupled Schrödinger equation. The different basis sets belong to limiting coupling cases, called Hund’s cases a to e. The eigenstates of short range molecules, where the nuclear distance is close to \( r_e \), are mostly expressed in Hund’s case a or b. At short range the electronic wavefunctions of the atoms overlap. The electrostatic interaction is stronger than the spin-orbit coupling of the individual atoms. Where in atoms the angular momentum \( L \) is a conserved quantity, in the cylindrically symmetric molecule \( L \) precesses around the internuclear axis. The component of \( L \) along the internuclear axis is conserved. Therefore the orbital angular momentum of the molecule is expressed in the new quantum number \( \Lambda \), which is the projection of \( L \) onto the molecular axis. If the rotational energy is smaller than the spin-orbit coupling\(^1\) (Hund’s case a), then the electron spin is expressed as the quantum number \( \Sigma \), which is the component of the atomic electronic spin \( S \) along the internuclear axis. The projection \( \Sigma \) of the spin together with \( \Lambda \) give rise to a total electronic angular momentum \( \Omega = |\Sigma + \Lambda| \). The rotational spectrum is described by the total rotational number \( J \) which is the sum of \( \Omega \) and the nuclear rotation quantum number \( R \). The molecular states described by the limiting coupling case a are denoted by the good quantum numbers \( \Lambda, S, \Sigma, J, \Omega \). The molecular potentials’ nomenclature consists of the following symbol \( P^{2S+1}\Lambda|\Omega|_{u,g} \). \( P \) designates the degree of electronic excitation. For the electronic ground state \( P \) takes the label “X” and higher excited potentials are designated by arabic numerals with increasing energy. \( \Lambda \) is represented by capital greek letters \( \Sigma, \Pi, \Delta, \) etc. equivalent to atomic orbital

\(^1\)The opposite case (Hund’s case b) is found by coupling nuclear rotation \( R \) to \( \Lambda \). The angular momentum quantum number \( N \) comprised of \( \Lambda \) and \( R \) is then coupled to the total electronic spin to form the total angular momentum \( J \).
nomenclature. The $\pm$-superscript is relevant to the $\Sigma$ states ($\Lambda = 0$) and marks the parity-symmetry of the electronic wavefunction upon reflection at a plane containing the intermolecular axis. The $u,g$-subscript$^2$ is relevant to homonuclear molecules only.

The molecules we create with PA are weakly bound. The nuclear distance of weakly-bound molecules is very large. To find good quantum numbers describing the molecular states of weakly-bound molecules one needs the limiting coupling case c. Hund’s case c stems from the fact that the electron clouds are more confined to the individual atoms due to the large distance, such that the atomic spin-orbit coupling dominates over the molecular electrostatic interaction. The total electronic angular momentum is expressed in terms of the atomic momenta $J_a = S + L$. The electronic angular momentum couples to the intermolecular axis to form the projection $\Omega$. Finally the total angular momentum $J$ is obtained by adding $\Omega$ to the nuclear rotation $R$. The quantum numbers describing the molecular states are $J_a, \Omega, J$. The nomenclature of the molecular potentials consists of $P|\Omega|$. Again $P$ takes the values X, 1, 2, 3, etc. for electronic ground-state and higher excited states.

### 5.1.2 Molecular structure of RbSr

Compared to a great amount of experimental and theoretical results on the molecular structure of bi-alkali molecules [174], the characterization of RbSr molecular potentials is limited. Traditional spectroscopy measurements, such as FTIR applied in a heat-pipe containing metal vapors, have not been published$^3$.

Recently spectroscopic studies of RbSr molecules on He-nanodroplets were published [175]. The authors produce a beam of He droplets by supersonic expansion. The He beam picks up Rb and Sr atoms while flying through vapour cells of the elements. Once one Rb and one Sr atom are caught on the surface of the He-droplet, the atoms undergo a chemical reaction relaxing into the ro-vibrational ground-state. The He droplet absorbs the binding energy and dissipates it through evaporation. The RbSr doped He droplet flies further into the laser spectroscopy region of the experiment. Two spectroscopic techniques are used to probe the molecules. Firstly, resonance enhanced two-photon ionization is used to probe the excited molecular potentials. Here a spectroscopy laser scans across the exited molecular potentials. Once the laser is

---

$^2$Marks the parity of the electronic wavefunction upon reflection around the molecule origin.

$^3$to the best knowledge of the author.
resonant with a molecular transition a second laser excites the molecule further to an ionized state that is visible on an ion detector. The second method is a fluorescence measurement. An excitation laser produces molecules that decay back into the ground state thereby emit photons. The spectrum of the fluorescent light provides information about bound-bound transitions. If a molecule is excited to a higher electronic state by the excitation laser, it detaches from the He-droplet. The molecular structure probed by fluorescence measurements therefore is not disturbed by the He mean-field.

Estimation of the dispersion coefficients by ab-initio and semi-empirical calculations are given in several articles. A summary of the literature survey is given in Tab. 5.1. Early ab-initio calculations of the ground-state potential of RbSr molecules

<table>
<thead>
<tr>
<th>C_6</th>
<th>10^{-4} C_8</th>
<th>10^{-6} C_{10}</th>
<th>Reference</th>
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<td>46.18</td>
<td>58.44</td>
<td>[178]</td>
<td>2015</td>
</tr>
</tbody>
</table>

**Table 5.1:** Literature dispersion coefficients of the RbSr ground-state potential in atomic units.

![Figure 5.1](image)

**Figure 5.1:** The ground-state potential energy curve of RbSr adapted from [162]. Results of two different theoretical methods are shown.
5.1. Diatomic molecules

\[ 1^1\Pi \quad \begin{cases} (\Sigma = \pm \frac{3}{2}, \Lambda = \pm 1) \\ (\Sigma = \mp \frac{3}{2}, \Lambda = \pm 1) \end{cases} \rightarrow (\Omega = 5/2) \rightarrow 1(\Omega = \frac{7}{2}) \]

\[ 3^3\Sigma^+ \quad \begin{cases} (\Sigma = \pm \frac{1}{2}, \Lambda = 0) \end{cases} \rightarrow (\Omega = 3/2) \rightarrow 4(\Omega = \frac{1}{2}) \]

\[ 2^3\Pi \quad \begin{cases} (\Sigma = \pm \frac{1}{2}, \Lambda = \pm 1) \end{cases} \rightarrow (\Omega = 1/2) \rightarrow 2(\Omega = \frac{1}{2}) \]

\[ 1^3\Pi \quad \begin{cases} (\Sigma = \pm \frac{1}{2}, \Lambda = \mp 1) \end{cases} \rightarrow (\Omega = 3/2) \rightarrow 1(\Omega = \frac{7}{2}) \]

\[ 2^3\Sigma^+ \quad \begin{cases} (\Sigma = \pm \frac{3}{2}, \Lambda = 0) \end{cases} \rightarrow (\Omega = 1/2) \rightarrow 3(\Omega = \frac{1}{2}) \]

\[ \chi^3\Sigma^+ \quad \begin{cases} (\Sigma = \pm \frac{3}{2}, \Lambda = 0) \end{cases} \rightarrow (\Omega = 1/2) \rightarrow 2(\Omega = \frac{1}{2}) \]

**Figure 5.2:** Correlation diagram for the RbSr electronic states, which dissociate into the most relevant atomic asymptotes accessible in our experiment. The electronic states in Hund’s case a) basis are computed in [162]. Their components in the \(|S\Lambda\Sigma\rangle\) basis are coupled by the spin-orbit interaction. The resulting electronic states are expressed in Hund’s case c). \(\Omega = |\Lambda + \Sigma|\) is a good quantum number and is used for the labeling of the potential energy curves. The spin-orbit coupled electronic states dissociate to a combination of free atom states of Rb and Sr.

were published in 2010 [179, 74]. In 2014 Žuchowski et al. provided ab-initio calculations of the ground-state as well as electronically excited molecular potentials using different methods and gave a comparison to the previously published results [162]. The details of the methods can be found in the publication. Here I will summarize the main results of the calculations and show the relevant potential energy curves for our spectroscopy investigations. The ground-state potential energy curve is shown in Fig. 5.1. The depth of the potential is 1034 cm\(^{-1}\) with an estimated uncertainty of 5%. The equilibrium distance and harmonic constant are \(r_e = 8.8(1) \, a_0\) and \(\omega_e = 38.5(5) \, cm^{-1}\). The slight discrepancy between the potential energy curves calculated by two different methods shown in Fig. 5.1 leads to a different number of supported bound states. The number differs by one bound state and one of the aims of the following spectroscopy
studies is the determination of the exact number. Furthermore the long-range part of the potential is to be determined with the help of the PA measurements. The long-range behaviour is modelled with $-C_6/R^6 - C_8/R^8$. The $C_6$ coefficient as well as the position of the last bound state is used to calculate the interspecies scattering lengths of Rb-Sr as described in [180].

In [162] first the excited RbSr potential energy curves in Hund’s case a are calculated. Relevant potentials to this work ($3^2\Sigma^+, 2^2\Pi, 1^4\Sigma^+, 1^4\Pi$) dissociate asymptotically to Rb($5s^2S$)+Sr($5s5p^3P$) at long interparticle distances. While the lowest quartet states $1^4\Sigma^+, 1^4\Pi$ and the states dissociating to the Rb($5p^2P$)+Sr($5s^2\ ^1S$) manifold show a low discrepancy between the calculation methods, the depth of the states $3^2\Sigma^+, 2^2\Pi$ exhibit a difference of 25% between the two methods. Experimental data is needed to obtain more accurate predictions.

Hund’s case a potentials are used to calculate the spin-orbit coupled potentials. The author of [162] assumes the spin-orbit coupling strength to be constant and equal
to the atomic spin-orbit coupling strengths but mentions that an internuclear distance
dependence of the SO coupling has to be taken in consideration in the future. Figure 5.2
shows the correlation diagram of the electronic states illustrating the method of obtaining
spin-orbit coupled Hund’s case c potential energy curves. Firstly the Hund’s case
a states are expressed in the asymptotic free-atom basis $|S_L \Sigma \Lambda \rangle$ using Clebsch-Gordan
coefficients. The matrix elements of the spin-orbit coupling Hamiltonian with good
quantum number $\Omega$ can then be constructed. The solutions of the Hamiltonian are the
Hund’s case c potentials shown in Fig. 5.3. The degeneracy of the asymptotes to different
atomic $J$ states is lifted. The diagram and potential plots show that spectroscopy on the
intercombination line $^1S_0 - ^3P_1$, as performed by us, accesses three electronic states
$5(\Omega = 1/2)$, $6(\Omega = 1/2)$, $2(\Omega = 3/2)$.

Hyperfine interaction is not taken into account in the calculations [162]. The coupling
strength of the Rb nuclear magnetic moment to all other momenta in the molecule
can be safely assumed to be weak. Therefore the nuclear angular momentum is added
to the good quantum number $\Omega$ resulting in the grand total angular momentum $T = \Omega + I_{\text{Rb}}$. The values of $T$ of the relevant electronic states $5(\Omega = 1/2)$, $6(\Omega = 1/2)$, $2(\Omega = 3/2)$
can be $T = 0, 1, 2, 3$ and the ground-state $X(\Omega = 1/2)$ has a hyperfine structure of $T = 1, 2$.
In general the hyperfine interaction Hamiltonian consists of several relativistic terms
attributed to for example electron-nuclear Fermi contact interaction and interactions of
the nuclear magnetic moment with the electron spin and orbital magnetic moments, vibration, rotation and higher order contributions [171]. To gain full knowledge of the
molecular structure the hyperfine interactions should be modelled precisely [181], for
which more spectroscopic data must be acquired than this thesis provides. The correct
model for the hyperfine interactions will be especially important for the fermionic
molecule $^{87}\text{Rb}^{87}\text{Sr}$ with a contribution of $I = 9/2$ from the $^{87}\text{Sr}$ atom to the molecular
hyperfine structure.

5.1.3 One-colour photoassociation

One-colour PA is a measurement technique resolving the vibrational states in the electrically
excited molecular potentials. Figure 5.4 shows the states and laser involved in the one-colour PA process. The starting point are two colliding Rb and Sr atoms (state $|a\rangle$) slightly above zero energy, because of finite kinetic energy. The atom pair
absorbs a photon from the laser $L_{\text{FB}}$ (free-bound laser) and undergoes a transition to a
Chapter 5. Photoassociation spectroscopy

Figure 5.4: Ground and excited-state molecular potentials from [162] relevant to PA spectroscopy on the intercombination line. Laser $L_{FB}$ with frequency $\nu_{FB}$ drives free-atom pairs of Rb and Sr to an excited molecular state $|m\rangle$ with Rabi-frequency $\Omega_{FB}$. Laser $L_{BB}$ with frequency $\nu_{BB}$ couples $|m\rangle$ to a molecular state $|g\rangle$ in the electronic ground-state with Rabi-frequency $\Omega_{BB}$. The energies of $|m\rangle$ and $|g\rangle$ are not to scale.
bound state $|m\rangle$ in the electronically excited molecular potential. The essence of the spectroscopy method is scanning the frequency $\nu_{FB}$ of $L_{FB}$ to detect a signature of molecules. Several methods can be employed to detect the associated molecules. They could be detected directly by further excitation to ions and subsequent ion counting. Although detecting the product molecules has a higher sensitivity [182] we employ a technically simpler approach. The molecule association rate is inferred by measuring the number of remaining atoms in the trap after irradiating an ultracold Rb-Sr mixture for about 500 ms with PA light. If $L_{FB}$ is resonant with a molecular state, the excited state molecules decay by spontaneous emission. The decay products, atoms or stable molecules, either leave the trap or are not detected by absorption imaging. From the atom loss features as shown in Fig. 5.6c one can infer the binding energy and lifetime of the molecular state as well as the strength of the free-atom to molecule transition [182]. Ultracold PA measurements can be performed in MOTs. The sample preparation time for MOTs is shorter allowing for a faster scan over a wide frequency range. The temperatures of MOTs is tens of $\mu$K [182]. The Doppler width of the spectroscopy features then is on the order of 1 MHz. The rotational structure of the molecule can be observed, because the atoms scatter in higher partial waves. The collision energies at such temperatures are high enough to overcome the centrifugal barrier of higher partial wave collisions. The association of molecules in rotationally excited states is possible. Our Rb-Sr samples used for PA spectroscopy are prepared in an ODT. The cycle time is longer compared to spectroscopy on MOTs. The advantage of using the ODT is the low temperature of the sample, which is on the order of 1 $\mu$K. The Doppler width is 22 kHz and the associated molecules are rotation-less. At temperatures of 1 $\mu$K $s$-wave collisions dominate the scattering of the atoms. The associated molecules therefore do not carry rotational angular momentum. Rotational spectra are easier to assign.

### 5.1.4 Two-colour photoassociation

Two-colour PA spectroscopy is used to measure the binding energies of the vibrational states in the electronic ground state. In addition to laser $L_{FB}$ the colliding atoms are subject to the bound-bound laser $L_{BB}$, which couples state $|m\rangle$ of the electronically excited potential to a vibrational state in the ground-state, $|g\rangle$. In this method, we keep the frequency of $L_{FB}$ fixed at resonance with the $|a\rangle \leftrightarrow |m\rangle$ transition, which we found previously by one-colour PA. Atom loss will be present because of formation of excited
molecules. As we scan the frequency of L_BB across a |m⟩ ↔ |g⟩ transition, the energy of |m⟩ is changed by the light-shift due to presence of resonant L_BB laser light. The rate of molecule creation |a⟩ ↔ |m⟩ decreases and the atom number measured by absorption imaging increases.

With L_BB on resonance and at high intensity, |m⟩ and |g⟩ can be expressed in the dressed state picture as a doublet |I⟩ = 1/√2 (|m⟩ + |g⟩) and |II⟩ = 1/√2 (|m⟩ − |g⟩). The energy splitting of the doublet is proportional to the Rabi-frequency Ω_BB induced by L_BB. This Autler-Townes doublet arranges symmetrically around the energy of |m⟩, therefore the probe laser L_FB no longer is on resonance and the PA atom loss decreases [183].

In addition to Autler-Townes splitting, also electromagnetically induced transparency (EIT), a process that requires coherence between the two PA lasers, can lead to atom number increase in the two-colour PA spectrum. The EIT phenomenon occurs when the coupling Ω_BB is set to a value below a certain threshold [184]. To explain EIT the three-level system with two couplings is described by the two states |bright⟩ = (Ω_FB |a⟩ + Ω_BB |g⟩) / Ω' and |dark⟩ = (Ω_BB |a⟩ − Ω_FB |g⟩) / Ω' in the dressed state picture. The states are normalized by Ω' = √Ω_FB² + Ω_BB². The two excitation pathways from |dark⟩ to |m⟩ by laser L_BB and L_FB interfere destructively leading to a sharp transparency window in the spectrum. In order to observe the EIT phenomenon L_FB and L_FB must be phase-coherent, otherwise projection of the |dark⟩ onto |bright⟩ occurs. Furthermore, the dark state can be quenched by collisions and radiative decay of |g⟩, which reduces the sharp signature of EIT.

The Autler-Townes and EIT spectra can be acquired once the bound-bound transition frequency is found by two-colour PA. The free-bound laser is scanned across the Autler-Townes doublet or EIT feature while the bound-bound laser is kept on resonance with the |m⟩ → |g⟩ transition.

5.2 Experimental procedure

Now the necessary steps to obtain the long-range molecular spectra of RbSr with one- and two-colour PA will be described starting from the preparation of an ultracold trapped mixture of Rb and Sr. Then I will sketch the PA light source and means of data acquisition and the interpretation of the spectra.
5.2. Experimental procedure

5.2.1 Sample preparation

The sample preparation sequence for PA spectroscopy measurements is similar to the one described in Chapter 3. For the PA studies the samples are not brought to quantum degeneracy by evaporative cooling. Instead the spectroscopy samples are sympathetically laser cooled to a temperature of 1 µK in the ODT. Although the low temperature and high density of BECs lead to increased PA rates, performing the PA measurements in thermal clouds has practical advantages. Firstly avoiding the evaporation sequence results in a shorter cycle-time of the experiment. The spectroscopy scans are acquired faster. Secondly the overlap between thermal clouds of Rb and Sr is less affected by gravity and interspecies interactions (see Chapter 4). The atoms can be trapped in a simple 1064-nm ODT instead of employing the alignment-sensitive bichromatic trap. The typical atom numbers for spectroscopy are roughly $10^6$ Rb and $5 \times 10^6$ Sr atoms. The one-colour PA measurements showed that the best signal strength could be achieved when the Sr atom number is as high as possible. This saturation in Sr atom number is necessary to compensate the off-resonant scattering losses of Sr from the PA laser. The Rb atoms are in the $F = 1$ hyperfine state occupying the three magnetic sub-levels almost equally with a slight preference for $m_F = -1$.

5.2.2 Spectroscopy laser

The frequency diagram of the one-colour PA laser is depicted in Fig. 5.5. The PA laser is derived from the red MOT master oscillator close to the $^1S_0 - ^3P_1$ transition in Sr. The frequency of the light is adjusted by several AOMs depending on the Sr isotope used and the targeted frequency scanning range. Typically two AOMs are employed to set the PA laser frequency: one AOM adapts the frequency to the isotope and the rough frequency range to be scanned (AOM I). A second AOM (AOM II) is used to scan the frequency by steps of 100 kHz or finer. The frequency-shifted beam is amplified by injection locking a slave laser diode. Further amplification can be provided by a tapered amplifier if higher intensity is required. The amplified laser intensity is controlled with another AOM at the output of the spectroscopy laser setup (AOM out). A mechanical

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4 For the ongoing spectroscopy studies on the $^{87}$Rb-$^{87}$Sr mixture A. Ciamei installed a beat-lock setup for scanning the frequency of the spectroscopy laser on a wider range, but with a larger linewidth compared to the injection locked PA laser.

5 A cascade of up to 6 AOMs was used to reach a frequency shift of 4.2 GHz.
Figure 5.5: Laser frequency diagram of the free-bound PA laser for $^{87}\text{Rb}^{88}\text{Sr}$ spectroscopy. The master oscillator is referenced to the $^1S_0 - ^3P_1$ transition in $^{88}\text{Sr}$ with an 80 MHz offset (see [185, 84]). AOM I compensates the isotope shift and AOM II is used to scan the laser in frequency below the dissociation threshold. The final AOM “out” is used to control the intensity of the PA light electronically. For spectroscopy measurements on $^{87}\text{Rb}^{88}\text{Sr}$ AOM I is set to diffract in +1-order shifting the frequency above the master laser. AOM II and AOM “out” shift the frequency back below the $^{88}\text{Sr}$ line, because they diffract in -1-order.

shutter avoids the exposure of atoms to stray light during the sample preparation. The PA beam is brought close to the glass cell via an optical fibre. The waist of the beam at the location of the atom cloud is adjusted in a range from 380 $\mu$m to 30 $\mu$m for different intensity requirements by combinations of a fibre collimator lens and a final focusing lens before the main experimental chamber.

The bound-bound spectroscopy laser is set up in a similar way as the free-bound laser. The red MOT master oscillator laser is split off to a second pair of AOMs, injection locked slave laser and an output AOM. Free-bound and bound-bound lasers are combined by a beam splitter and coupled into the same optical fibre, which transports the light to the glass cell. The frequencies of the bound-bound AOMs are set such that the following condition is fulfilled: $\Delta = |\Delta_{BB} - \Delta_{FB}|$, where $\Delta_{FB}$ is the free-bound laser detuning, $\Delta_{BB}$ is the bound-bound laser detuning and $\Delta$ is the binding energy of the vibrational state of the ground-state molecule.
5.2. Experimental procedure

5.2.3 Data acquisition and interpretation

In the following a detailed overview of the measurement procedure will be given. I will explain how we gain information about the magnetic structure and the lifetime of the excited molecular states. In the measurements presented here only the Rb atom number loss is considered because most of the Sr atoms are expelled from the ODT due to off-resonant scattering during the PA pulse. This is why the atomic cloud sample used for spectroscopy contains a five times higher number of Sr than Rb atoms. This large number difference gives each Rb five chances to associate, creating pronounced Rb loss features. At the same time the Rb atom number is not influenced as strongly by the PA laser close to resonance with a Sr transition. Spurious frequency components in the PA laser easily produce unwanted Sr atom loss features while the Rb atom number stays steady.

Figure 5.6a illustrates the states involved in the PA process. The initial state consists of colliding Sr and Rb atoms. The $F = 1$ Rb atoms can be in either one of the three
magnetic sub-states. The spectroscopy measurements are performed in presence of a homogeneous magnetic field, which lifts the degeneracy of the initial states.

The excited state is in either of the three molecular potentials shown in Fig. 5.4 with a high vibrational quantum number and a pair of hyperfine quantum numbers \( T, m_T \).

The magnetic sub-states of the excited levels acquires Zeeman energy from the magnetic field, which contributes to the observed number of features in the PA spectrum. The observed features must be assigned to a vibrational level as well as the total and azimuthal quantum number \( T, m_T \).

To identify the magnetic quantum number of the initial state we measure the magnetic sub-state distribution of Rb with the help of Stern-Gerlach separation. Prior to releasing the atoms from the dipole trap for time-of-flight (TOF) followed by absorption imaging a magnetic field gradient is applied in the direction of gravity. After the dipole trap is switched off the Rb \( |F = 1, m_F = \pm 1 \rangle \) sub-states are accelerated in opposite directions while the \( |1, 0 \rangle \) Rb falls freely under gravity. After 5 ms the magnetic gradient is switched off and the accelerated clouds fly apart for the remainder of the expansion time. Then an absorption image is taken. In Fig. 5.6b, the three components of Rb are distinctly visible. For each experimental run the PA laser frequency is increased step-wise. If the PA laser is scanned across a molecular resonance, the Rb clouds experience losses. Because the Zeeman splitting of the initial states is larger than the linewidth of the molecular transition we see individual Rb clouds disappear as the laser frequency matches the transition frequency plus Zeeman energy (see Fig. 5.6b,[i, ii, iii]). A exemplary PA spectrum at 50 MHz binding energy is shown in Fig. 5.6c. The colours of the spectral plots denote the magnetic quantum number of the initial state as measured with Stern-Gerlach TOF absorption imaging. The magnetic field strength is set to 3 G during this PA pulse.

With the help of Stern-Gerlach separation the initial magnetic state can be identified clearly. The magnetic structure of the molecular state addressed needs to be identified as well. We are interested in finding the quantum numbers \( T, m_T \) and the magnetic moment of the molecular state. The magnetic moment is the change of energy of a state in dependence of the magnetic field strength.

The assignment of the loss features of a one-colour PA spectrum to the quantum numbers \( T, m_T \) is realized by taking spectroscopy measurements of the loss features at varying magnetic field strengths. We observe the evolution of the centre frequencies of the loss features in dependence of the magnetic field. The three prominent features
5.2. Experimental procedure

Figure 5.7: Magnetic structure of the “i” line at 260.54(5) MHz. a) Loss spectra of the line at increasing magnetic field values. The three \( m_F \) clouds of Rb exhibit loss features separated by the magnetic field in frequency. b) The Rb Zeeman energy (Breit-Rabi) and the energy at zero B-field is subtracted from the center frequencies of the three loss features and plotted against magnetic field strength. The solid line is a second order polynomial fit to the data with quadratic Zeeman constant: -24(7) kHz/G^2.

On the right-hand side of Fig. 5.6c for example converge towards the \( m_F = 0 \) feature at 55 MHz with decreasing magnetic field strength indicating that they belong to the same molecular state. A feature with similar magnetic field dependent behaviour to the one at 55 MHz binding energy is shown in Fig. 5.7a. At zero B-field all magnetic sub-state populations of Rb show one loss feature centred around the same binding energy value. With increasing B-field the features move apart in frequency. The magnetic field dependent frequency shift of the loss features is the sum of the Zeeman energy of the molecular state and the initial state. The Zeeman energy of the initial state is well known by the Breit-Rabi relation of Rb \( F = 1 \) Zeeman structure. If we subtract this Rb Zeeman structure from the centre frequencies of each loss feature, we obtain the Zeeman structure of the molecular state (see Figure 5.7b). The centres of the loss features in each initial state collapses on one line. We conclude that the total angular momentum quantum number of this molecular state is \( T = 0 \). The magnetic field strength evolution of the state is fitted with \( E(B) = aB + bB^2 \), where \( a \) is the linear and \( b \) is the quadratic Zeeman constant. As expected for a \( T = 0 \) state, only the quadratic Zeeman constant is different from zero: \( b = -24(7) \) kHz/G^2. The magnetic structure of the other molecular features are analyzed in the same way and will be presented in the next section.
Typical lineshapes of one- and two-colour PA spectra are derived in [186, 187, 188]. The authors give expressions for the scattering matrix elements describing the collisions of ultracold atoms in presence of the free-bound and bound-bound lasers. To get the observed lineshapes the inelastic scattering matrix elements have to be averaged over the thermal distribution of the atoms. If the temperature of the spectroscopy sample is larger than $\hbar \Gamma / k_B$, where $\Gamma$ is the natural linewidth of the transition, then the lineshape will be subject to inhomogeneous Doppler-broadening. Our experimental data shows no linewidth of the one-colour loss features narrower than 120(20) kHz corresponding to $h \Gamma / k_B = 0.9 \mu K$. The spectroscopy samples have a temperature of 1 $\mu$K. Therefore the resulting one-colour lineshapes resemble Lorentzian functions closely. The linewidth of the master laser is 2 kHz and will not influence the spectroscopic width. The one-colour loss features and Autler-Townes spectra are fitted with Lorentzian functions to retrieve the centre frequencies, heights and widths of the resonances. The binding energies of the molecular states are given by the centre frequencies of the spectroscopy features relative to the frequency of the $^1S_0-^3P_1$ transition, which is determined by loss spectroscopy of a Sr sample. The relative frequency can be retrieved from the total AOM frequency shift of the PA laser (see Fig. 5.5). The binding energy is reported with the statistical errors from the fits. The systematic error caused by AC-Stark shifts from trapping and PA lasers as well as the spectroscopy lock offset of the master laser are estimated to be 100 kHz. We confirm this estimate by comparing the $^{88}\text{Sr}_2 \nu = -1$ spectroscopic feature with literature values. Our measurement of this molecular line deviates by 100 kHz from the literature value of 23.932(33) reported in [189].

5.3 Experimental results

In the following a complete report of our findings of the vibrational states in the electronically excited and ground-state molecular potentials of the isotopologues $^{87}\text{Rb}^{88}\text{Sr}$ and $^{87}\text{Rb}^{84}\text{Sr}$ will be given.

5.3.1 Electronically excited $^{87}\text{Rb}^{88}\text{Sr}$

Figure 5.8 shows a one-colour PA spectroscopy scan of the $^{87}\text{Rb}^{88}\text{Sr}$ mixture. There are two binding energy regions separated by 200 MHz where spectroscopy features were
found. The top panel shows many molecular lines between 18 MHz and 60 MHz, the bottom panel shows three lines around 265 MHz binding energy. The spectra are assembled from different experimental runs. The empty intervals between experimental runs are confirmed to be flat by PA measurements that are not shown. The intensity of the PA beam used in the spectroscopy scans is chosen to produce a good signal-to-noise ratio without saturating the transitions. For the upper panel the intensity is 50(25) mW/cm\(^2\) and hundred-fold higher (I\(_{\text{PA}}\) = 5(2) W/cm\(^2\)) for the lower panel. The atoms are illuminated by the PA light for 1 s. The RbSr resonances are indicated by orange vertical lines and labelled in alphabetical order with increasing binding energy. The feature marked with a blue vertical line is a \(^{88}\text{Sr}_2\) molecular resonance at 23.932 MHz binding energy [189]. The appearance of this feature in the Rb atom number is probably caused by inelastic collisions of Rb with photo-associated, excited \(^{88}\text{Sr}_2\) molecules as well as their products from spontaneous emission, \(^{88}\text{Sr}_2\) molecules in the ground-state.

The magnetic structure of the three features i, j and k is exemplary. The magnetic

![Graph](image-url)

**Figure 5.8:** One-colour PA loss spectrum of \(^{87}\text{Rb}^{88}\text{Sr}\).
structure of the “i” line was presented in the previous section to show how the magnetic structure is retrieved from the loss spectrum. Based on the described analysis procedure the magnetic structure of the “j” and “k” lines will be presented in the following.

After subtraction of the Rb Zeeman energy and the $|B| = 0$ binding energy from the acquired data the bare magnetic structure of the “j” and “k” states show a larger number of magnetic sub-states than the “i” line. In Fig. 5.9a five magnetic sub-states appear from the $f = 265.58(5)$MHz state. The grand principal quantum number is $T = 2$. The transitions to the stretched states $m_T = \pm 2$ at lower magnetic field values appeared too weak to be resolved.

The next lower bound state “k” at 272.91(7) MHz binding energy has seven magnetic sub-states arising from $T = 3$ as seen in Fig. 5.9b. Transitions up to $m_T = \pm 3$ can be accessed from the Rb $F = 1$ manifold.

The molecular states closer to dissociation threshold with binding energies from 18.8 MHz to 53.5 MHz are observed with much less PA laser intensity suggesting that the transition probabilities from the free atom state to these states are larger than to the states belonging to the “i”, “k” and “j” spectroscopic lines.

The interpretation of the magnetic structure of the shallower bound states is more difficult. The high density of spectroscopic features made their assignment hard. Also, several expected transitions were not observed even though the spectroscopy scans
were performed with a finer magnetic field search. The magnetic field was applied in perpendicular and parallel orientation with respect to the PA-beam, so that pi and sigma transitions are mapped out respectively. However, some transitions remain unobserved.

Instead of showing the Zeeman structures of all lines a to h, the results of our investigation is summarized in Table 5.2. The mentioned ambiguity in the number of the observed magnetic sub-states is indicated by a star in Tab 5.2. Only two or four magnetic sub-states are observed when the “b, “e” lines or “h” line are mapped out at different magnetic fields. Even numbers of magnetic sub-states are only possible when the molecule has half-integer grand principle quantum number. The $^{87}\text{Rb}^{88}\text{Sr}$ is bosonic leading to the conclusion that not all magnetic sub-states are detected. The result for the linear Zeeman shift of lines “b”, “e” and “h” appears close to the values of the neighbouring lines, but needs further refinement.

The natural linewidth of “f”, “h” and “i” is measured with several spectroscopy scans with decreasing intensity of the PA beam. The last column of Tab. 5.2 gives the value of the probe beam intensity at which the smallest linewidth in the scan series is obtained. The linewidth of all other lines around 30 MHz binding energy as well as the “j” and “k” line is estimated with a few scans and does not deviate beyond 20% from the observed values presented in the table.

The one-colour PA line “f” is the strongest and narrowest observed in the study. The feature is measurable at an intensity of 2(1) mW/cm$^2$, a factor $10^3$ smaller than the necessary intensity to observe the “i”, “j” and “k” lines or most lines in the $^{87}\text{Rb}^{84}\text{Sr}$ mixture. The low intensity required to initiate PA loss indicates a higher transition probability between the free atom state and the molecular state corresponding to the “f” line.

### 5.3.2 Electronically excited $^{87}\text{Rb}^{84}\text{Sr}$

Table 5.3 shows the binding energies and observed properties of the four spectroscopic features found in $^{87}\text{Rb}^{84}\text{Sr}$. Molecular bound states at 173.5(2) MHz, 387.4(5) MHz and 427.8(2) MHz binding energy are measured using samples at similar temperatures and densities as described above. The 11.74(10) MHz resonance could be observed only when atom pairs of Rb and Sr are confined in a 3D optical lattice.

The molecular spectrum of excited $^{87}\text{Rb}^{84}\text{Sr}$ is qualitatively different from the
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<table>
<thead>
<tr>
<th>Line</th>
<th>Binding energy (MHz)</th>
<th>Observed magnetic states</th>
<th>Linear Zeeman shift (MHz/G)</th>
<th>Observed linewidth (kHz)</th>
<th>Relative height</th>
<th>Minimal linewidth (kHz)</th>
<th>Probe intensity (mW/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>a</td>
<td>18.80(10)</td>
<td>3</td>
<td>0.98(10)</td>
<td>245(20)</td>
<td>0.45</td>
<td></td>
<td></td>
</tr>
<tr>
<td>b</td>
<td>21.00(15)</td>
<td>2*</td>
<td>0.9*</td>
<td>310(10)</td>
<td>0.625</td>
<td></td>
<td></td>
</tr>
<tr>
<td>c</td>
<td>32.23(60)</td>
<td>1</td>
<td>0.42(5)</td>
<td>360(10)</td>
<td>0.95</td>
<td></td>
<td></td>
</tr>
<tr>
<td>d</td>
<td>36.26(50)</td>
<td>5</td>
<td>0.67(10)</td>
<td>240(30)</td>
<td>0.63</td>
<td></td>
<td></td>
</tr>
<tr>
<td>e</td>
<td>36.94(10)</td>
<td>2*</td>
<td>0.8*</td>
<td>240(30)</td>
<td>0.22</td>
<td></td>
<td></td>
</tr>
<tr>
<td>f</td>
<td>41.39(60)</td>
<td>3</td>
<td>0.90(20)</td>
<td>570(15)</td>
<td>1.0</td>
<td>120(20)</td>
<td>2(1)</td>
</tr>
<tr>
<td>g</td>
<td>52.54(30)</td>
<td>5</td>
<td>0.27(30)</td>
<td>700(40)</td>
<td>0.55</td>
<td></td>
<td></td>
</tr>
<tr>
<td>h</td>
<td>53.50(40)</td>
<td>4*</td>
<td>0.4*</td>
<td>1930(60)</td>
<td>0.95</td>
<td>390(20)</td>
<td>21(5)</td>
</tr>
<tr>
<td>i</td>
<td>260.54(5)</td>
<td>1</td>
<td>0.05(5)</td>
<td>280(20)</td>
<td>0.75</td>
<td>265(25)</td>
<td>1500(300)</td>
</tr>
<tr>
<td>j</td>
<td>265.58(5)</td>
<td>5</td>
<td>0.24(7)</td>
<td>370(20)</td>
<td>1.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>k</td>
<td>272.91(7)</td>
<td>7</td>
<td>0.28(8)</td>
<td>490(30)</td>
<td>0.5</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 5.2: Summary of the observed one-colour PA lines of $^{87,88}$Rb$^{88}$Sr with important properties. The stars above the values indicate that not all magnetic sub-states were resolved. The lack of data makes the predicted values ambiguous.

$^{87}$Rb$^{88}$Sr case. Resonances “c” and “d” appear broader than any resonance in the $^{87}$Rb$^{88}$Sr molecule. The shallow-bound line “a” is three to four times broader than any $^{87}$Rb$^{88}$Sr counterpart.

The line “b” shows the largest PA loss at lowest intensity. Still the intensity required to photo-associate molecules on the “b” transition is $10^3$ times higher than for the strong “f”-line in $^{87}$Rb$^{88}$Sr. The transition probability of the free atom state to the vibrational states in the electronically excited potential of $^{87}$Rb$^{84}$Sr isotopologue seem to be much weaker than in the $^{87}$Rb$^{88}$Sr molecule.

5.3.3 Ground state of $^{87}$Rb$^{88}$Sr

We were able to find two near-threshold vibrational states with binding energies of 356.99(3) MHz and 2153.83(15) MHz in the electronic ground state of the $^{87}$Rb$^{88}$Sr molecule. The results are summarized in Tab. 5.4. Figure 5.10 shows a two-colour PA scan of the vibrational level $\nu = -2$. The one-colour PA laser was resonant with the “k” line at 273 MHz binding energy during the spectroscopy scan. The magnetic field
5.3. Experimental results

<table>
<thead>
<tr>
<th>Line</th>
<th>Binding energy (MHz)</th>
<th>Observed magnetic states</th>
<th>Linear Zeeman shift (MHz/G)</th>
<th>Observed linewidth (MHz)</th>
<th>Probe intensity (W/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>a</td>
<td>11.74(10)</td>
<td>0</td>
<td>0.8(2)</td>
<td>0.2</td>
<td></td>
</tr>
<tr>
<td>b</td>
<td>173.5(2)</td>
<td>1</td>
<td>-0.05(5)</td>
<td>0.47(3)</td>
<td>3.3</td>
</tr>
<tr>
<td>c</td>
<td>387.4(5)</td>
<td>3</td>
<td>0.47(17)</td>
<td>5.0(1.0)</td>
<td>3.0</td>
</tr>
<tr>
<td>d</td>
<td>427.8(2)</td>
<td>5</td>
<td>0.5(2)</td>
<td>5.0(1.0)</td>
<td>3.0</td>
</tr>
</tbody>
</table>

Table 5.3: Summary of the observed one-colour PA lines of $^{87}$Rb$^{84}$Sr

of 5G allowed to address the $m_F = 0$ Rb sub-population selectively. The frequency of $L_{BB}$ was scanned across the relevant region and the Rb $m_F = 0$ atom number was measured by absorption imaging after TOF expansion with Stern-Gerlach separation. Three peaks in the Rb atom number appear. We measured the evolution of the energy of the three peaks with magnetic field. The magnetic structure of the state is close to the magnetic moment of the Rb atom in the $F = 1$ hyperfine state. We conclude that the magnetic structure of the molecular state originates from the nuclear magnetic moment. The total angular momentum of the rotational state is $T = 1$. The coupling strength between the excited state and the ground state of the molecule can be measured with the help of Autler-Townes spectra (see Fig. 5.16). After finding the resonance frequency for $L_{BB}$ by two-colour PA, $L_{BB}$ is set to that frequency and a fixed intensity and the frequency of $L_{FB}$ is scanned across the one-colour resonance. The Autler-Townes doublet appears in the atom loss measurement. The splitting of the doublet is proportional to the Rabi-frequency induced by $L_{BB}$. Several Autler-Townes

<table>
<thead>
<tr>
<th>Vibrational level</th>
<th>Binding energy (MHz)</th>
<th>Total quantum number</th>
<th>Linear Zeeman shift (MHz/G)</th>
<th>Coupled excited state</th>
<th>Coupling strength $(2\pi , \text{kHz}/\sqrt{\text{mW/cm}^2})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>-1</td>
<td>356.99(3)</td>
<td>T = 1</td>
<td>0.67(5)</td>
<td>$f$ (41.39 MHz)</td>
<td>3.39(25)</td>
</tr>
<tr>
<td>-2</td>
<td>2153.83(15)</td>
<td>T = 1</td>
<td>0.68(3)</td>
<td>$i$ (260.54 MHz)</td>
<td>0.6(2)</td>
</tr>
</tbody>
</table>

Table 5.4: Properties of the observed ground-state vibrational and hyperfine states of $^{87}$Rb$^{88}$Sr.
features at different $L_{BB}$ intensities are taken to obtain the intensity-dependent Rabi-frequency, which is $2\pi \times 0.6(2) \text{ kHz}/\sqrt{\text{mW/cm}^2}$ between the $\nu = -1$ in the ground state and the “i”-state.

The $\nu = -1$ was addressed through a shallower bound vibrational state in the excited molecular potential, the “f” state with a binding energy of 41.39 MHz. The $\nu = -1$ state exhibits a similar magnetic structure as the $\nu = -2$ state (see Tab. 5.4). The coupling strength between the “f” and the $\nu = -1$ molecular states is five-fold higher than in the case described above.

**Nuclear distance dependent hyperfine coupling**

The existence of narrow, magnetically tunable Feshbach resonances between Rb and the non-magnetic Sr ground-state atoms are predicted [74] to rely on an interaction effect, which is based on the modification of the hyperfine coupling of Rb because of the presence of the nearby Sr atom during a collision. PA spectroscopy of ultracold Rb-Sr mixtures on the $^1S_0-^3P_1$ intercombination line of Sr is ideally suited for the investigation of this interaction effect. The Rb hyperfine coupling is predicted to change by up to 20% when the Rb and Sr atom are at proximity in a deeply bound vibrational state [74]. The accuracy of our PA spectroscopy method compared to the Rb hyperfine coupling constant is on the order of $10^{-5}$, which is enough to reveal the effect even when the atoms are far apart in a shallow bound vibrational state.
5.3. Experimental results

With the help of PA we can bring two atoms close to each other in a controlled manner. We can choose selectively, which vibrational state we populate by the choice of the resonance frequencies of the PA lasers and therefore choose which distance the atoms have. The near-threshold vibrational states we populate with two-colour PA have distances of tens of $a_0$.

The measurement of the hyperfine coupling strength is possible with PA spectroscopy as well. We measure the difference in binding energies of the same vibrational state in the $T = 1$ and $T = 2$ hyperfine manifolds. The principle of the hyperfine coupling measurement for one vibrational state is shown in Fig. 5.11. The binding energy of the $\nu = -1, T = 1$ molecular state is measured by two-colour PA as before. As discussed in the previous section the measurement of the magnetic structure shown in Fig. 5.12a confirms that the total angular momentum is $T = 1$. The binding energy of the same vibrational state in the $T = 2$ manifold is measured by changing the frequency of the bound-bound laser by $\zeta_{\text{Rb}} \simeq 6834$ MHz, which is close to the hyperfine coupling of $^{87}\text{Rb}$. Figure 5.12b shows upper half of the magnetic structure of this feature. Because of the symmetry of the magnetic structure this is enough to say that the total angular momentum of the state is $T = 2$. If the hyperfine coupling was not influenced by the presence of Sr, the position of the $\nu = -1, T = 2$ resonance frequency would be offset exactly by the Rb hyperfine splitting. The two-colour PA measurement reveals this state at a slightly different frequency, which shows that the hyperfine coupling strengths has changed. The binding energy of the $T = 2$ state is calculated by deducting the Rb hyperfine splitting from the found resonance frequency. Table 5.5 shows the measured binding energies of the hyperfine doublets of the $\nu = -1$ and $\nu = -2$ states. The mean distance was calculated using the wavefunctions of the vibrational states found by our fitting procedure described in Sec. 5.5.

Our measurement shows, that the hyperfine coupling constant varies with internuclear distance. For a distance of 41.74 $a_0$ the change is 3.07(15) MHz or $4.5 \times 10^{-4}$.

---

6Expectation value of the position operator of the radial wavefunction of that vibrational state.
Chapter 5. Photoassociation spectroscopy

<table>
<thead>
<tr>
<th>Vibrational level</th>
<th>Mean distance ($a_0$)</th>
<th>Binding energy $T = 1$</th>
<th>Binding energy $T = 2$</th>
<th>$\Delta E$</th>
</tr>
</thead>
<tbody>
<tr>
<td>-1</td>
<td>56.46</td>
<td>356.99(3)</td>
<td>357.87(4)</td>
<td>0.88(4)</td>
</tr>
<tr>
<td>-2</td>
<td>41.74</td>
<td>2153.83(15)</td>
<td>2156.90(15)</td>
<td>3.07(15)</td>
</tr>
</tbody>
</table>

Table 5.5: Change of binding energy with internuclear distance.

### 5.3.4 Ground state of $^{87}$Rb$^{84}$Sr

The two-colour PA measurements reveal two bound states in the ground state potential of the $^{87}$Rb$^{84}$Sr molecule. The results are summarized in Tab. 5.6. The binding energy of the bound states is 29.01(3) MHz and 744.525(22) MHz. The measurement of the magnetic structure of the states did not bring as much clarity as in the case of $^{87}$Rb$^{88}$Sr. Only one and two magnetic sub-states were measured by two-colour PA spectroscopy with varying magnetic field. The two-colour measurement of the state at 29 MHz was performed using the intermediate “d” and the 744.5 MHz bound state was

![Figure 5.12](image-url)  

**Figure 5.12:** Magnetic structure of the $\nu = -2$ state for different hyperfine states. 
- a) Magnetic structure of the feature at a binding energy of 357 MHz. 
- b) Magnetic structure taken 6.8 GHz (hyperfine splitting Rb) blue detuned.
5.4. STIRAP pathway for weakly-bound $^{87}\text{Rb}^{84}\text{Sr}$

Here I will quantify the chosen STIRAP pathway consisting of two transitions. The free-bound transition couples the free atoms $|a\rangle$ to an excited intermediate state $|m\rangle$ and the bound-bound transition couples $|m\rangle$ to the weakly-bound target state $|g\rangle$. The efficiency of molecule creation via STIRAP depends on the coupling strength between the states and the lifetime of the states. A high $\frac{\Omega_{FB}^2}{\gamma_m}$ and $\frac{\Omega_{BB}^2}{\gamma_g}$ is required [42]. The choice of the intermediate states in the $^{87}\text{Rb}^{84}\text{Sr}$ mixture is restrictive. In contrast to weakly-bound Sr$_2$ molecules, which we have associated by STIRAP [80], the excited state decay rate of the strongest lines is fifty-fold larger. Also in order to observe $^{87}\text{Rb}^{84}\text{Sr}$ free-bound resonances a higher intensity of the PA laser is required indicating that the Rabi coupling is small for the technically achievable laser intensities in our experiment.

<table>
<thead>
<tr>
<th>Vibrational level</th>
<th>Binding energy (MHz)</th>
<th>Observed magnetic substates</th>
<th>Linear Zeeman shift (MHz/G)</th>
<th>coupled excited state</th>
<th>Coupling strength (2$\pi$ kHz/$\sqrt{\text{mW/cm}^2}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>-1</td>
<td>29.01(3)</td>
<td>1</td>
<td>0.030(7)</td>
<td>d (427.8 MHz)</td>
<td>16(10)</td>
</tr>
<tr>
<td>-2</td>
<td>744.525(22)</td>
<td>2</td>
<td>1.15(4)</td>
<td>d (427.8 MHz)</td>
<td>290(100)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>b (173.5 MHz)</td>
<td>6.7(3)</td>
</tr>
</tbody>
</table>

Table 5.6: Properties of the observed ground-state vibrational states of $^{87}\text{Rb}^{84}\text{Sr}$.

addressed through the “d” as well as “b” states in the electronically excited potential. The bound-bound transitions show a two- to hundred-fold higher Rabi-frequency than we observed in the $^{87}\text{Rb}^{88}\text{Sr}$ two-colour PA measurements. The Franck-Condon overlap between the vibrational levels in the excited state and the ground state of $^{87}\text{Rb}^{84}\text{Sr}$ must be higher than for the other isotopologue.

We can now chose one free-bound transition and one bound-bound transition to use them in a STIRAP sequence for weakly-bound molecule creation. The arguments for our choice will be presented in the following section.
The strongest free-bound transition we found is the “b”-line addressing a vibrational state with 174 MHz binding energy. In order to estimate the STIRAP efficiency based on the ratio between coupling strength and decay rate we first measure the excited state lifetime. Then the coupling strength of the transition is quantified in terms of the Rabi-frequency achieved at a certain free-bound intensity.

The choice of the target level in the electronic ground-state is not as crucial. The bound-bound Rabi-frequency is obtained by measuring Autler-Townes splittings and the result shows that the coupling strength is large for both transitions found by spectroscopy. Nevertheless the target state is chosen to be the ν = −2 vibrational state at 744 MHz binding energy because it is deeper bound. The wavefunction of the deeper bound state has a smaller spatial extend potentially easing further shrinking of the weakly-bound molecules into the ro-vibronic ground-state by a second STIRAP sequence.

The natural linewidth of the “b”-transition is determined with the method described in Sec. 5.2.3. The spectroscopic feature “b” is acquired with step-by-step decreasing intensities of the PA laser. The power broadening decreases with smaller intensities as can be seen in Fig. 5.13a. The spectroscopic features are fitted with Lorentzian profiles and the widths of the profiles are plotted against the intensity. Figure 5.13b shows the evolution of the linewidth. Fitting the model \( \gamma = \gamma_m \sqrt{1 + I/I_0} \) gives the natural linewidth \( \gamma_m = 2\pi \times 480(50) \text{ kHz} \).

To determine whether STIRAP will successfully produce weakly-bound molecules, we estimated the free-bound Rabi-frequency \( \Omega_{FB} \). In a bulk mixture of Rb-Sr \( \Omega_{FB} \) is dependent on the density of the two species. In order to reproduce the initial condition of the STIRAP experiments, the estimate of \( \Omega_{FB} \) is performed on the Mott-insulator state of many Rb-Sr doubly occupied sites. Because of the large natural linewidth of the free-bound transition, the observation of Rabi-oscillations, and therefore a precise measurement of \( \Omega_{FB} \) is possible only if \( \Omega_{FB} \) exceeds the natural linewidth. In addition the temporal resolution of the experiment has to be smaller than fractions of microseconds. The measurement of coherent Rabi flopping is involved. Therefore \( \Omega_{FB} \) is estimated with the help of PA loss measurements in time.

An estimation of \( \Omega_{FB} \) is obtained by the following procedure. The mixture is brought to quantum degeneracy as described in Chap. 3. The double BEC is loaded into a deep optical lattice (200 E\(_{R,Rb}\)). Roughly \( 3 \times 10^4 \) lattice sites are occupied by one
5.4. **STIRAP pathway for weakly-bound $^{87}$Rb$^{84}$Sr**

Figure 5.13: Measurement of the natural linewidth of the free-bound transition to “b”. a) shows the spectroscopic features taken at different free-bound PA intensities with Lorentzian fits. b) Lorentzian width of the features is plotted against the PA intensity and fitted with $\gamma = \gamma_m \sqrt{1 + I/I_0}$. The natural linewidth is 480(50) kHz.

Rb and one Sr atom (Chap. 4). Then the free-bound laser resonant with with the transition “b” is switched on for a variable time. The atom number remaining after the pulse is measured via time-of-light absorption imaging. Fig. 5.14a shows the relative Rb atom number decay in the lattice with PA pulses of different peak intensities. For long pulses the Rb atom number approaches the number of Rb atoms in sites without Sr, which corresponds to zero in Fig. 5.14a. The atom-number decay is fitted with $N(t)/N_0 = \exp(\Omega_{FB}^2 t/\gamma)$ with Rabi-frequency $\Omega_{FB}$ and excited-state decay rate $\gamma$ [190]. The excited state decay rate is given by the natural linewidth measured as described before. The intensity dependent Rabi-frequency is plotted against the square root of the PA laser intensity. The linear fit gives $\Omega/\sqrt{I} = 2\pi \times 20(8) \text{ kHz}/\sqrt{W/cm}^2$.

The free-bound coupling can be increased in the experiment by focusing the STIRAP laser to a smaller spot size or by using a tapered amplifier to maximize the available laser power. The maximum free-bound laser power achieved is 40 mW, which, focused to 30 $\mu$m yields, a peak intensity of 2.8 kW/cm$^2$. The estimated Rabi-frequency amounts to $2\pi \times 1$ MHz, which is high enough compared to the excited state life time to perform STIRAP molecule association. At such intensities AC-Stark shifts become
a valid concern. During the STIRAP pulse the resonance frequencies of the intermediate and target states shift dynamically posing a limitation on the STIRAP transfer efficiency.

The AC-Stark shifts of $|m\rangle$ and $|g\rangle$ are measured by determining the centre of the spectroscopic features with varying intensity of the free-bound probe laser. Figure 5.15a shows the centre positions of the free-bound resonance “b” in dependence of the free-bound laser intensity with a linear fit. With higher intensity the binding energy becomes smaller. The intensity dependent shift is $-3.0\text{kHz/W/cm}^2$.

The AC-stark shift of the target state $\nu = -2$ is measured by two-colour PA. The free-bound laser intensity is varied and the bound-bound laser frequency is scanned to record the dark-resonance feature. Figure 5.15b shows the centre of the two-colour PA peak in dependence of the free-bound intensity. The target state shifts to lower frequencies with increasing free-bound intensity at $-4.4(3)\text{kHz/W/cm}^2$. 

**Figure 5.14:** Free-bound Rabi-frequency estimation in the optical lattice measured by atom loss in dependence of free-bound PA pulse time. a) shows the Rb atom number decay for different PA intensities with exponential fits. The y-axis is normalized by the number of doubly occupied Rb-Sr sites. b) shows a plot of the free-bound ($2\pi \times \text{Rabi-frequencies}$ calculated from the decay rates in dependence of the square root of PA intensity and a linear fit with $20(8)\text{kHz/}\sqrt{\text{W/cm}^2}$ slope.
The coupling strength between the intermediate state and the target state is obtained with the help of Autler-Townes spectra. Figure 5.16a shows Autler-Townes doublets with increasing splittings because of increasing intensities of $L_{BB}$. The splitting is plotted against the square-root of the $L_{BB}$ intensity in Fig. 5.16b. The linear fit leads to the intensity dependent Rabi-frequency of $7.45(50) \text{ kHz/} \sqrt{\text{mW/cm}^2}$.

In the next section the STIRAP sequence is simulated taking the acquired quantities into consideration.

### 5.4.1 Weakly-bound molecules and STIRAP

Our attempts of producing weakly-bound molecules of $^{87}\text{Rb}^{84}\text{Sr}$ by STIRAP have not been successful. The following section will explain the negative outcome. Numerical simulations of the STIRAP sequence help to pinpoint the requirements for successful creation and detection of weakly-bound molecules, which have not been met with the chosen isotopologue. The transfer efficiency of STIRAP must be higher than a threshold, if one is to detect the associated molecules. The successful detection of
produced molecules by STIRAP depends on the detection method. The direct detection of the created molecules by absorption imaging for example is not straightforward \[191\]. Therefore the transfer efficiency of the STIRAP is measured indirectly. The measurement method relies on two STIRAP sequences. During the first STIRAP sequence the weakly-bound molecules are created. The free atoms, which were not associated during the first STIRAP sequence, are blasted away with a near-resonant pulse. Only molecules remain, which are dissociated into free atoms by a second, reverse STIRAP sequence. The atom number can be measured by absorption imaging and related to the molecule number.

In the following the model will be introduced briefly. Then, three cases of STIRAP will be presented. Firstly, the STIRAP sequence was simulated with the actual experimental conditions with the outcome of no measurable molecule creation. Secondly, we assume that the AC-Stark shifts can be compensated to a level of 1%. In this hypothetical scenario, 20% of the atom-pairs in the lattice could be associated into weakly-bound molecules. Thirdly, we assume that the Rabi-coupling of the free-bound transition is

### Figure 5.16: Measurement of the bound-bound Rabi frequency from intermediate state “b” to target state $\nu = -2$.

a) shows the spectroscopic features when scanning the frequency of the free-bound laser taken at different bound-bound intensities: red circles $I_{BB} = 46 \text{ W/cm}^2$, blue triangles $I_{BB} = 16.6 \text{ W/cm}^2$, black squares $I_{BB} = 8.67 \text{ W/cm}^2$. b) Difference frequency of the two peaks from the spectrum against square root of $I_{BB}$. The intensity dependent Rabi-frequency is $7.45(50) \text{ kHz}/\sqrt{\text{mW/cm}^2}$
ten-fold higher and the AC-Stark shift is compensated in an experimentally reasonable way. The transfer efficiency in this case could reach 50%.

The interaction of quantum states with electromagnetic fields is modelled with the master equation [192],
\[ \frac{d\rho}{dt} = \frac{i}{\hbar} [\rho, H] - L\rho. \] (5.2)
The system is described by the density matrix $\rho$. Diagonal elements of the matrix are the populations of the quantum states. The free-atom state, excited molecule and ground-state molecule populations are labelled by $\rho_a$, $\rho_m$, and $\rho_g$, respectively. After applying the rotating wave approximation the three-level Hamiltonian $H$ is
\[ H = \begin{pmatrix}
0 & -\frac{\Omega_{FB}(t)}{2} & 0 \\
-\frac{\Omega_{FB}(t)}{2} & -\Delta a(t) & -\frac{\Omega_{BB}(t)}{2} \\
0 & -\frac{\Omega_{BB}(t)}{2} & -\Delta a(t) - \Delta b(t)
\end{pmatrix}. \] (5.3)
The detuning of the free-bound laser from the $|a\rangle \rightarrow |m\rangle$ transition is $\Delta a$ and the bound-bound laser detuning is $\Delta b$. Both detunings and the Rabi-frequencies $\Omega_{FB}$, $\Omega_{BB}$ are time-dependent, because the laser intensity is changed during the STIRAP sequence. The STIRAP pulses have Gaussian shapes
\[ \Omega_{FB}(t) = \alpha_{fb} \sqrt{I_{FB}} \exp\left(-\frac{(t - \Delta t)^2}{4\sigma_{fb}^2}\right), \] (5.4a)
\[ \Omega_{BB}(t) = \alpha_{bb} \sqrt{I_{BB}} \exp\left(-\frac{t^2}{4\sigma_{bb}^2}\right). \] (5.4b)
The parameters $\alpha_{fb}$, $\alpha_{bb}$ link the laser intensities to the Rabi-frequencies. The energies of the intermediate state $|m\rangle$ and target state $|m\rangle$ move due to the AC-Stark shift induced by the free-bound laser
\[ \Delta a(t) = \beta_m I_{FB} \exp\left(-\frac{(t - \Delta t)^2}{2\sigma_{fb}^2}\right), \] (5.5a)
\[ \Delta b(t) = \beta_g I_{BB} \exp\left(-\frac{(t - \Delta t)^2}{2\sigma_{bb}^2}\right). \] (5.5b)
The AC-Stark shift is proportional to the free-bound laser intensity by the parameters $\beta_m$ and $\beta_g$. The pulse lengths $\sigma_{fb}$, $\sigma_{bb}$ and the separation of the pulses $\Delta t$ are around
The dissipation term $L\rho$ in the master equation is given by

$$L = \begin{pmatrix} \gamma_{\text{scat}} & 0 & 0 \\ 0 & \gamma_m & 0 \\ 0 & 0 & 0 \end{pmatrix}. \quad (5.6)$$

The free-atom decay rate $\gamma_{\text{scat}}$ is dominated by the off-resonant scattering PA-light. At the peak intensity of the free-bound laser the Sr atoms scatter at a rate of $\gamma_{\text{scat}} = 80$ Hz. The electronically excited molecule decays on a much faster time scale $\gamma_m = 480(50)$ kHz. The third diagonal entry is zero, because we assume that the lifetime of the weakly-bound molecules is large compared to the STIRAP pulse time. The optical lattice protects the molecules from inelastic losses [97].

The master equation is constructed with the help of the AtomicDensityMatrix package for Mathematica [193]. The resulting nine coupled differential equations of the entries of the density matrix are not shown. The equations are solved numerically for three different parameter choices. At the beginning of the STIRAP sequence the system is in the free-atom state $|a\rangle$, therefore the initial condition is $\rho_{a} = 1$. The diagonal entry of the numerical result $\rho_{g}$ gives the population of the weakly-bound ground state molecules during the STIRAP pulses. Its value at the end of the STIRAP sequence is the transfer efficiency.

The numerical results are shown in Fig. 5.17. The populations of the three states during the STIRAP sequence are presented in the first row of Fig. 5.17. The second row shows the AC-Stark shifts of the molecular states $|m\rangle$ and $|g\rangle$ and the third row shows the Rabi-frequencies in time.

The STIRAP sequence in Fig. 5.17a was simulated with parameters equal to the measured values presented in the previous section. This simulation represents our attempts of creating weakly-bound molecules of $^{87}\text{Rb}^{84}\text{Sr}$ by STIRAP. The peak intensities of the STIRAP lasers were chosen to reach peak Rabi-frequencies of 350 kHz satisfying the condition $\frac{\Omega_{FB}^2}{\gamma_m} \gg 1$. The power requirements for that condition are well within our experimental capabilities. The AC-Stark shifts of the molecular levels are on the order of MHz. Because of the large AC-Stark shifts during the STIRAP pulses the population of weakly-bound molecules remains nearly zero as shown in the first graph of Fig. 5.17a.

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7Free-bound power $I_{FB} = 10$ mW with a waist of 30 $\mu$m at the position of the sample.
5.4. STIRAP pathway for weakly-bound $^{87}\text{Rb}^{84}\text{Sr}$

![Numerical results of the STIRAP sequence.](image)

**Figure 5.17:** Numerical results of the STIRAP sequence.  

- **a)** Simulation using the measured experimental parameters. 
- **b)** The AC-Stark shift parameters $\beta_m$ and $\beta_g$ are a hundred-fold smaller. 
- **c)** The Rabi-coupling strength $\alpha_{fb}$ is ten-fold higher and $\beta_m = 1.4\text{kHz/W/cm}^2$ and $\beta_g = -0.044\text{kHz/W/cm}^2$. The first row shows the diagonal elements of the density matrix representing the populations of $|a\rangle$, $|m\rangle$, and $|g\rangle$. The second row shows the AC-Stark shifts of the $|m\rangle$ and $|g\rangle$ energies during the STIRAP pulses. The third row shows the STIRAP pulses.

Figure 5.17b shows the numerical result of a STIRAP sequence, where both AC-Stark shift parameters $\beta_m$ and $\beta_g$ are reduced by a factor of hundred. The peak intensities and pulse durations were optimized on the STIRAP transfer efficiency, which reached nearly 20%. This efficiency would enable further STIRAP steps towards ground-state molecules. The dynamic compensation of the AC-Stark shift is possible by employing an additional optical field as will be explained in the next section. Compensating the AC-Stark shifts of both molecular levels at the same time seems difficult.

Figure 5.17c shows the numerical result of a STIRAP sequence with 50% transfer efficiency, which would be excellent for the creation of ground-state molecules. The chosen parameters for the simulation are realistic. Firstly, the Rabi-coupling $\alpha_{fb}$ is ten-fold higher than before. Secondly, the AC-Stark shift parameter $\beta_g$ is compensated
by a factor of hundred and $\beta_m$ is partially reduced to 1.4 kHz/W/cm$^2$. The choice of the larger Rabi-coupling is motivated by the observation that the free-bound transition strengths in $^{87}$Rb$^{88}$Sr is higher than in $^{87}$Rb$^{84}$Sr. The right choice of the isotopologue could provide the ten-fold increase in free-bound Rabi-frequency. The strategy of AC-Stark compensation will be explained briefly in the following.

**AC-Stark shift compensation**

It is evident from the simulations that the AC-Stark shift of the molecular states during the STIRAP sequence has to be reduced for a successful creation of molecules. The AC-Stark shift compensation is achieved by introducing a third laser beam into the STIRAP sequence. The frequency of the third laser beam is blue detuned from the $^1S_0-^3P_1$ transition. The AC-Stark shift arising from this laser has the opposite effect compared to the free-bound laser. The molecular states shift in the opposite direction. Both shifts can cancel out, if the ratio between the compensation and free-bound intensities is 87%. With this choice the compensation beam intensity has to follows the pulse shape of the free-bound laser precisely. Experimentally we realize the compensation beam by superimposing it with the free-bound laser, equalizing the modes of both beams by sending them through one optical fibre, and pulse shaping their intensities using one AOM that produces the pulse for the STIRAP. The free-bound and compensation pulse are formed by sending a pulsed RF signal to the AOM. The diffracted beam from this AOM is coupled into a polarization maintaining fibre and sent to the main chamber. The source of the compensation beam is the tapered amplifier (TA) that produces the free-bound beam. The output of the TA is split into two parts, the free-bound beam and the compensation beam. An AOM serves as the beam splitter. The 0th diffraction order is not frequency-shifted and serves as the free-bound beam. The +1st-order diffraction of the AOM is shifted by 270 MHz and is then recombined with the free-bound beam by a 50:50-beam splitter before both beams are send to the pulse-shaping AOM. The detuning of the compensation beam from the $^1S_0-^3P_1$ transition is about 100 MHz, far off resonant compared to the linewidth of the transition and therefore not causing much off-resonant scattering. The ratio between the compensation and free-bound intensities for which AC-Stark compensation is achieved is controlled by the RF-intensity of the beam-splitting AOM.

Measurements of the AC-Stark shift of the target state showed that the shift can
be compensated with the help of our strategy. Nevertheless, the creation of RbSr molecules could not be confirmed even when performing STIRAP with the compensation pulse. Because the binding energy of the target state shifts at a different rate than the intermediate state, the compensation pulse remedied the target state AC-Stark shift but a residual shift of the intermediate state remains. The simulations show that for successful RbSr molecule creation by STIRAP both shifts have to be compensated. Compensating the shift of the intermediate state could in principle be realized by a second compensation beam at a different frequency in the future, putting a molecular association efficiency of 20% within reach.

However our strategy compensating the AC-Stark shift was successfully applied to the production of $^{84}\text{Sr}_2$. When using the compensation beam the production efficiency was enhanced to 80% compared to 50% without compensation. The results will be published in the near future [194].

### 5.5 Long-range ground-state potential and scattering properties

The near threshold vibrational state energies we obtained by two-colour PA provide us with valuable information about the long-range branch of the ground-state molecular potential. Knowing the long-range form of the potential energy curve, we can predict the binding energies of deeper lying states and hence speed up spectroscopy measurements. More spectroscopic data would refine the theoretical potential shape at shorter distances and bench-mark the ab-initio calculations. Measurements of the internuclear-distance dependent hyperfine splitting and magnetic structure of the molecular hyperfine states could lead to the first observation of Feshbach resonances between ground-state alkaline-earth and alkali atoms. Furthermore the $C_6$ coefficient and the binding energy of the last bound state determine the interspecies scattering length. As discussed in previous chapters the interspecies scattering length determines how accessible the combination of different isotopes of Rb and Sr will be in the experiment. Unfavourable interspecies interaction could lead to prohibitive three-body recombination losses, immiscibility of the mixtures. The creation of a low-entropy Mott-insulator in optical lattices that contains many doubly occupied sites could also be impeded.
First we use the well-known Leroy-Bernstein formula to fit the two vibrational energies per isotopic combination that we found. The fit result shows a large discrepancy between the two isotopic combinations. We conclude that more spectroscopic data is needed for the approach of Leroy and Bernstein to be successful. Second we use the ab-initio parameters of the ground-state potential given in [162] and solve the stationary Schrödinger equation of the molecule numerically. Then the depth and the dispersion coefficients are varied to match all four spectroscopically found binding energies with the numerical solutions. The resulting $C_6$ coefficient and the binding energy of the last bound state give us the interspecies scattering length of all isotopic combinations of Rb and Sr.

5.5.1 Leroy-Bernstein formula

It is common in the literature to fit the near-threshold vibrational energies obtained by PA spectroscopy [195, 196, 58] with an analytical formula derived in [197]. The vibrational energies $\Delta$ are fitted with

$$\Delta = -\frac{1}{\hbar} \left( \frac{2\pi}{\mu} \right)^{\frac{1}{2}} \frac{\Gamma(1 + \frac{1}{n})}{\Gamma(\frac{1}{2} + \frac{1}{n})} \frac{n}{\sqrt{C_n}} (\nu_D - \Delta \nu) \frac{n-2}{2n} \frac{n}{n-2} \right)^{\frac{2n}{n-2}}. \tag{5.7}$$

The constant $\mu$ in this expression is the reduced mass of the molecule, $\nu_D$ is the non-integer index of the free-atom threshold, $C_n$ are the dispersion coefficients of order $n$ and $\Gamma(x)$ is the gamma function. The derivation of Eq. 5.7 is based on the first-order WKB condition of quantized action [192]. In order to fit Eq. 5.7 to experimental data successfully, knowledge of four or more rotationless vibrational energies is required. We found four vibrational states in total, two for each isotopologue. Fitting the two least bound states with the Leroy-Bernstein formula of order $n = 6$ independently for the two isotopologues $^{87}$Rb$^{84}$Sr and $^{87}$Rb$^{88}$Sr works well within the measurement uncertainty, but the $C_6$ coefficients obtained from the fits differ substantially. While the fit result of the two vibrational levels in the ground-state potential of $^{87}$Rb$^{88}$Sr is $C_6 = 3527(100) \, \text{E}_h \, \text{a}_0^6$, the two vibrational levels of the isotopologue $^{87}$Rb$^{84}$Sr are fit with $C_6 = 3181(100) \, \text{E}_h \, \text{a}_0^6$.

None of the fitted $C_6$ coefficients match the ab-initio calculations presented in Sec. 5.1.2. The electronic potential shapes are expected to be independent of the mass,
except for small correction due to a possible isotope-dependent nuclear-quadrupole-electronic interaction [171].

For a successful fit of the Leroy-Bernstein formula or its extensions [198] to the binding energies of each isotopologue independently more spectroscopic data is needed.

5.5.2 Numerical solution

Instead of using the semi-classical Leroy-Bernstein formula to retrieve the long-range potential shape, we take an approach based on numerically solving the Schrödinger equation for the RbSr molecule with the method described in [199]. The solution to the stationary Schrödinger equation \((\hat{K} + U(R))\psi = E\nu \psi\), where \(\hat{K}\) is the kinetic energy operator of the nuclear motion, gives the vibrational energies \(E\nu\) for both isotopologues. The mass of the different isotopologues is contained in \(\hat{K}\). The potential energy \(U(R)\) is approximated by \(U(R) = D_e e^{-\alpha(R-r_e)} - C_6/r^6 - C_8/r^8\). The shape of the potential is first fitted to the ab-initio result of [162]. After solving the Schrödinger equation numerically the resulting vibrational energies in the immediate proximity of the dissociation threshold are compared to the four experimental results. The spectroscopic parameters and the dispersion coefficients of \(U(R)\) are varied to obtain the best match of experiment and numerical results.

For the best fit of the potential to our experimental results we find that the molecular potential supports 58 bound states for both isotopologues. The theoretical binding energies up to \(\nu = -5\) are shown in Tab. 5.7. The fitted results deviate a few MHz from the measured values. With the help of more experimental data, the \(- C_{10}/R^{10}\) term in the long-range branch of \(U(R)\) could provide a better match in the future.

The long-range branch of the fitted potential is formed by the dispersion coefficients \(C_6 = 3855.5(5.0) E_0 a_0^0\) and \(C_8 = 299788(50)\). The depth of the potential is found to be \(D_e = 1012(10) \text{ cm}^{-1}\) and is close to the ab-initio value.

5.5.3 Interspecies scattering lengths

The spectroscopic determination of the long-range branch of the RbSr ground-state potential and the binding energy of the last bound state is useful to obtain the interspecies scattering length of Rb-Sr. The analytical expressions, which give the scattering length, are given in [180]. The results of the calculation are tabulated in Tab. 5.8. The measured interspecies scattering lengths of the combination of \(^{87}\text{Rb}\) with \(^{88}\text{Sr}\) and
Chapter 5. Photoassociation spectroscopy

<table>
<thead>
<tr>
<th>Vibrational level</th>
<th>Isotopologue</th>
<th>Binding energy (MHz)</th>
<th>Experiment</th>
<th>Fit</th>
</tr>
</thead>
<tbody>
<tr>
<td>-1</td>
<td>$^{87}\text{Rb}^{84}\text{Sr}$</td>
<td>29.01(3)</td>
<td>30.28</td>
<td></td>
</tr>
<tr>
<td>-2</td>
<td></td>
<td>744.525(22)</td>
<td>742.14</td>
<td></td>
</tr>
<tr>
<td>-3</td>
<td></td>
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<td>3334.15</td>
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<tr>
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<tr>
<td>-5</td>
<td></td>
<td></td>
<td>19083</td>
<td></td>
</tr>
<tr>
<td>-1</td>
<td>$^{87}\text{Rb}^{88}\text{Sr}$</td>
<td>356.99(3)</td>
<td>357.67</td>
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</tr>
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<td></td>
<td>2153.83(15)</td>
<td>2155.9</td>
<td></td>
</tr>
<tr>
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<td>-5</td>
<td></td>
<td></td>
<td>28071</td>
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</tr>
</tbody>
</table>

Table 5.7: Measured and predicted binding energies for the near-threshold states up to $\nu = -5$ for the isotopologues $^{87}\text{Rb}^{84}$ and $^{87}\text{Rb}^{88}\text{Sr}$.

$^{84}\text{Sr}$ are favourable for cross-thermalization, explaining the success of our strategy of sympathetic laser cooling and the creation of double BEC without loosing many atoms to three-body recombination as shown in Chap. 3. A more careful approach could be necessary when mixing the fermionic Sr with $^{87}\text{Rb}$ atoms. The interspecies scattering length is at a larger and negative value, so that three-body losses could hinder a straightforward experimental realization of a doubly-degenerate Bose-Fermi mixture. If intolerable atom losses were encountered in future experiments with fermionic Sr and $^{87}\text{Rb}$, one could employ an experimental strategy similar to experiments with Rb-Cs mixtures [114, 200]. Firstly, because of interspecies recombination losses in Rb-Cs, spatially separated dimple traps made the production of BECs of Rb and Cs in the same experimental run possible [114]. The dimple beam storing the Cs BEC was movable by means of a piezo-actuated mirror, therefore the two BECs could be overlapped after production. Populating an optical lattice with immiscible Rb-Cs BECs from the separated dimple traps was achieved by transporting the Cs superfluid on top of a Rb Mott insulator [200]. This experimental strategy enabled the production of more than 5000 Rb-Cs molecules in the ro-vibronic ground state in an optical lattice and could be employed in our experiment easily.

\[8\text{In addition the interspecies scattering length was set to zero during the transport with the help of a Feshbach resonance in order to create a low-entropy Mott-insulator with many doubly occupied sites.}\]
5.6 Free-bound transition strengths

The simulations of the STIRAP sequence in Sec. 5.4.1 showed that the association of weakly-bound RbSr molecules will be successful, if we use free-bound transitions with larger transition dipole moments than we observed in the $^{87}$Rb-$^{84}$Sr spectra. The one-colour PA spectra of the isotopic combination $^{87}$Rb-$^{88}$Sr showed strong lines, which could lead to a successful STIRAP. The scattering properties of $^{88}$Sr prohibit us to create BECs with large atom-numbers and a Mott-insulator with many doubly occupied sites of $^{87}$Rb-$^{88}$Sr. Therefore we can not use the $^{87}$Rb-$^{88}$Sr mixture for our goal to create ultracold molecules. Rb has two and Sr has four stable isotopes and there are eight possible isotopic combinations in total. We could find a combination that has large enough transition dipole moments for a successful STIRAP and at the same time has favourable scattering properties for the creation of quantum gas mixtures. One-colour PA spectroscopy helps us to predict, which of the six remaining combinations we should explore experimentally. Here I will summarize the main result of the analysis of the one-colour PA spectra and show that the combination of the fermionic isotope $^{87}$Sr with $^{87}$Rb has large free-bound transition dipole moments. This isotopic combination will enable us to create fermionic weakly-bound molecules. A detailed analysis of the spectra will be published in the near future.

The transition strength between the free-atom state and an excited molecular state is proportional to the Franck-Condon factor (FCF). The FCF is calculated by integrating the product of the free-atom and the bound wavefunctions. The wavefunctions of the bound, excited molecule were obtained numerically with the help of the excited molecular potentials shown in Fig. 5.3. The long-range branch of the potentials

<table>
<thead>
<tr>
<th></th>
<th>$^{85}$Rb</th>
<th>$^{87}$Rb</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{84}$Sr</td>
<td>303.27</td>
<td>90.81</td>
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<td>88.94</td>
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<td>$^{87}$Sr</td>
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<td>-387.67</td>
</tr>
<tr>
<td>$^{88}$Sr</td>
<td>-10.70</td>
<td>273.43</td>
</tr>
</tbody>
</table>

Table 5.8: Interspecies scattering lengths in units of $a_0$ calculated by the semi-empirical result of [180]. The binding energy of the last bound state for the determination of the scattering lengths in bold were measured with our experiment. The other values are predictions based on the fit of the molecular potential to the experimental values.
was modified such that the vibrational energies in the potentials fit the one-colour PA results. The free-atom wavefunction was found with the help of the ground-state potential discussed in 5.5 using the semiclassical approximation [180].

The FCFs between the free-atom state and the excited vibrational states of $^{87}\text{Rb}^{84}\text{Sr}$ are a factor of ten smaller than in the case of $^{87}\text{Rb}^{88}\text{Sr}$. We could see this difference qualitatively in our one-colour PA spectra (see Sec. 5.3). The estimated FCFs between free atoms and vibrational states in the excited molecule $^{87}\text{Rb}^{87}\text{Sr}$ are on the same order as the ones in the case of $^{87}\text{Rb}^{88}\text{Sr}$. A factor of ten in the FCF of one free-bound transition in $^{87}\text{Rb}^{87}\text{Sr}$ would give us high enough free-bound Rabi-frequency for the STIRAP. Together with the AC-Stark shift compensation technique we could obtain a molecule association efficiency of 50% as simulated in Sec. 5.4.1.

5.7 Conclusion and outlook

With the help of PA spectroscopy on a narrow intercombination line we determined a possible pathway for STIRAP to transfer free atoms of $^{87}\text{Rb}$ and $^{84}\text{Sr}$ to weakly-bound molecules. The association of molecules was not successful, because the free-bound transition strength is too small. The small transition strength lead to small free-bound Rabi-frequencies for molecule association via STIRAP. Increasing the Rabi-frequency by a larger coupling laser intensity leads to an AC-Stark shift of the molecular states during the STIRAP pulse, which inhibits the population transfer. We attempted to overcome the AC-Stark shift by means of a compensation beam. The compensation beam has the same spatial and temporal pulse shape as the free-bound beam and shifts one of the molecular states in the opposite direction as the free-bound beam. We demonstrated the feasibility of the AC-Stark shift compensation by enhancing the $^{84}\text{Sr}_2$ molecule association from 50% to 80%. The AC-Stark shifts in the RbSr case are much stronger. The molecule association via STIRAP was not successful even when the AC-Stark shift of the target state was compensated. A second compensation beam could cancel the shift of the intermediate state during the STIRAP sequence and bring a molecule association efficiency of 20% within reach. A higher association efficiency could be achieved by simply using a different isotopic combination that has stronger free-bound transitions. With the help of PA spectroscopy we identified that the combination of $^{87}\text{Rb}^{87}\text{Sr}$ is promising to reach our goal.

Furthermore we measured the interspecies scattering lengths between Rb and Sr
atoms precisely. The interspecies scattering length is a crucial parameter which determines whether the ultracold mixture is miscible or immiscible and whether the mixture can be transferred into an optical lattice producing a Mott insulator easily.

We measured the change of the hyperfine splitting of Rb by the presence of another atom. The distance dependent hyperfine splitting is proposed to be the main effect that leads to Feshbach resonances between Rb and the non-magnetic alkaline-earth metal atom Sr.

Currently the spectroscopy studies are extended to the isotopic combination $^{87}$Rb-$^{87}$Sr. Other combinations could be explored as well. In order to gain precise knowledge of the molecular potentials more spectroscopic data is needed. The PA spectroscopy could be performed in a MOT to resolve the rotational structure and speed up the data acquisition. Using the clock transitions of Sr to access molecular states could allow us to perform optical spectroscopy of molecules at a precision below 1 kHz.
Chapter 6

Tapered amplifier laser with frequency-shifted feedback

We present a frequency-shifted feedback (FSF) laser based on a tapered amplifier. The laser operates as a coherent broadband source with up to 370 GHz spectral width and 2.3 $\mu$s coherence time. If the FSF laser is seeded by a continuous-wave laser a frequency comb spanning the output spectrum appears in addition to the broadband emission. The laser has an output power of 280 mW and a center wavelength of 780 nm. The ease and flexibility of use of tapered amplifiers makes our FSF laser attractive for a wide range of applications, especially in metrology.

6.1 Introduction

An FSF laser consists of a pumped gain medium inside a cavity that also contains a frequency shifting element such as an acousto-optic modulator (AOM). After each round trip the photons experience an increment in frequency through the AOM. Consequently exponential amplification of single frequency modes will not occur, since the fields of different round trips through the cavity cannot interfere constructively. Instead, spontaneously emitted photons from the gain medium are frequency shifted and amplified, resulting in a coherent modeless broadband emission spectrum. Seeding the FSF laser with a continuous-wave (cw) laser adds a second spectral component to the laser output, a comb of equidistantly spaced frequency components [201]. If parameters are chosen correctly, the FSF laser can generate Fourier-limited laser pulses with tunable repetition rate [202, 203, 204].

FSF lasers have many applications. Their coherent broadband emission is used
for optical domain ranging, as in [205], where a distance of 18.5 km was measured with parts-per-million accuracy. The broad spectrum of FSF lasers allows one to cover a larger velocity class in Doppler broadened atomic transitions, improving optical pumping of room temperature He\textsuperscript{*} gases [206] or increasing the fluorescence in Na vapors, which could improve astronomy guide star techniques [207]. The frequency comb like feature of an externally seeded FSF laser can be applied to spectroscopy, wavelength division multiplexed coherent communications [208] and optical frequency referencing [209].

Several implementations of FSF lasers with different gain media were reported. Early versions were based on HeNe [210], dye [211, 212], Ti:sapphire [203], and Nd:YLF lasers [204]. Modern implementations make use of a Yb\textsuperscript{3+} doped fiber amplifiers [213] as well as semiconductor lasers [214, 215, 216, 217].

In this article we demonstrate an FSF laser that uses a tapered amplifier (TA) as gain medium. TAs are electrically pumped semiconductor laser amplifiers, which are capable of emitting single-mode laser radiation at Watt levels. TA chips are commonplace in many laboratories and can be built into miniaturized setups [218]. The relatively cheap devices are available at a wide range of emission wavelengths. Our work leverages these advantages, making FSF lasers easily accessible to more researchers, thereby widening the range of applications of these light sources. In mutually independent work, a similar system was developed for broadband laser cooling, as indicated by conference abstract [219]. In the following, we present first our experimental setup and then the spectral properties of the FSF laser.

### 6.2 Experimental setup

The experimental setup consists of four electro-optical sub-systems shown in Fig. 6.1a-d, namely the FSF laser (a), the seed source (b), and the characterization tools (c,d). Each sub-system will now be described in detail. Our FSF laser consists of a TA and an AOM inside a four-mirror ring-cavity of 1440 mm optical length. The total loss through the mirrors is 1.4\%. The gain medium of the laser is realized by a tapered amplifier (Toptica TA-0780-2000-4). Its gain profile is centered at 780 nm with a full width at half maximum (FWHM) of 10 nm. The TA chip is temperature stabilized to 20 °C and operated at a current of 1 A yielding an output power of 300 mW. The divergent output beam of the TA is collimated by the lens \( L_{TA2} \). The Faraday isolator (FI) prevents laser
6.2. Experimental setup

light from traveling in the backwards direction, which would damage the input facet of the TA. The emission of the TA is split into two orthogonally polarized parts by a half-wave plate and a polarizing beam splitter (PBS). The vertically polarized reflection off the PBS serves as the output port of the FSF laser which is sent to the diagnostic tools via optical fiber. The horizontally polarized transmitted transmission of the PBS continues into the feedback loop. The power in the feedback loop is deduced from the power of the leakage light through mirror $M_1$. The half-wave plate is adjusted such that the power at the TA input facet remains below 30 mW in order to avoid damaging the TA chip.

The feedback light propagating through the acousto-optic modulator (AOM, Gooch & Housogo 3080-120 or Crystal Technology 3350-190) is shifted in frequency by $f_{\text{AOM}} = 60$ to 380 MHz with efficiencies of up to 87%. Lenses $L_1$ and $L_2$ adapt the beam waist for optimal injection of the TA through focusing lens $L_{TA1}$. The focal distance of $L_{TA1}$ and the orientation of mirrors $M_2, M_3$ is adjusted to achieve maximal output power of the TA.

The seed laser light (Fig. 6.1b) is provided by an external cavity diode laser (ECDL, Toptica DL Pro). The ECDL single-mode emission is tunable around 384.25 THz (780.2 nm) with a linewidth below 1 MHz on a timescale of 5 µs. The ECDL optical frequency is measured by a Burleigh 1500 wavemeter (WM) with 0.5 GHz precision. The seed laser beam is introduced into the FSF ring cavity through the back of the AOM co-aligned with the first order diffraction of the feedback.

The spectral properties of the output of the FSF laser are analysed in the optical and

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig6_1.png}
\caption{Schematic of the FSF laser and characterization tools. (a) cavity of the FSF laser, (b) seed laser, (c) radio frequency characterization setup, (d) grating spectrometer}
\end{figure}
the RF domain. For phase noise measurements in the radio frequency (RF) domain the output of the FSF laser is sent through a Michelson interferometer shown in the upper half of Fig. 6.1c. The light from the output port of the interferometer is detected by a 2 GHz bandwidth photodiode (PD) connected to a spectrum analyzer (SA, Rohde-Schwarz FSH8). A second interferometer setup, used to resolve the mode structure of the FSF laser, is shown in the lower part of Fig. 6.1c. The output is coaligned with the equally polarized beam of a second ECDL laser on a PD, producing a beat signal between the two lasers. The beat signal is measured by the SA and the optical frequency of the reference ECDL is measured by the Burleigh 1500 wavemeter (WM).

The optical spectrum of the FSF laser output is recorded by a spectrometer (Fig. 6.1d). The dispersive element of the spectrometer is a holographic grating with 1800 grooves per mm and a size of 50 mm × 50 mm. In order to illuminate a large number of grooves and increase the resolution of the spectrometer, the input beam is widened by a beam expander (BE) to roughly 20 mm. The first order diffraction is guided back onto the same grating at a different angle by a pair of mirrors. The same arrangement is repeated so that the resolution is enhanced threefold. The third diffraction off the grating is sent through a telescope (L₁s, L₂s) to decrease the beam waist to 1 mm. Subsequently the diffracted beam is projected onto a CMOS camera (Cam) by a cylindrical lens of 150 mm focal length. The spectrometer is calibrated using the seed laser set to different optical frequencies. The measured resolution of the spectrometer is 4 GHz.

### 6.3 Properties of the laser

Two modes of operation of the frequency-shifted feedback laser are realized. Firstly, the laser is internally seeded by spontaneous emission from the tapered amplifier, resulting in the emission of modeless broadband light. Secondly, the cw-laser seed light is introduced into the cavity resulting in a comb of narrow, equidistant frequency components in addition to the modeless emission.
6.3. Properties of the laser

Figure 6.2: Optical spectrum of the FSF laser measured with the grating spectrometer without external seed (solid black) and an external seed power of 460 µW (dashed blue) and 720 µW (dotted red).

6.3.1 Broadband modeless laser

We first discuss the operation of the laser without external seed. We use \( f_{\text{AOM}} = 80 \) MHz and chose the +1-order diffracted beam of the AOM for feedback. The diffraction efficiency is 75%. After the feedback is injected into the TA the output power of the FSF laser reaches 280 mW, which matches the specified output power of the TA.

The optical spectrum of the FSF laser is shown in Fig. 6.2. The center of the profile is tunable over 1 THz (2 nm) by changing the angle of the injection mirror \( M_2 \) in the horizontal plane and here is set to 383.0 THz (782.75 nm). The full width half maximum of the spectrum is 120 GHz, independent of the center frequency. The spectrum can be broadened by increasing the AOM frequency, as shown in Sec. 6.3.2.

To prove that the broadband laser emission is modeless we use the beat setup (Fig. 6.1c). The reference ECDL frequency is set close to the center of the FSF output spectrum and scanned over 200 GHz. The measurement reveals an RF spectrum without beat signals as expected for a modeless spectrum.

The RF spectrum obtained through the Michelson interferometer (see Fig. 6.3a) also shows the characteristic features of a modeless broadband laser. The spectrum exhibits a comb structure with lines separated by integer multiples of the cavity free spectral range \((n \times f_{\text{FSR}})\), where each comb line is accompanied symmetrically by a pair of lines at \(n \times f_{\text{FSR}} \pm f_b\). The comb structure stems from the fact that the initial spontaneous emission of the TA reoccurs after each round trip [201] and has a spacing of
Chapter 6. Tapered amplifier laser with frequency-shifted feedback

\( f_{FSR} = 208.54(6) \) MHz. The frequency difference between the comb lines and the side peaks, \( f_b \), is related to the arm length difference \( \Delta L \) of the Michelson interferometer and \( f_b = \gamma 2 \Delta L / c \), where \( \gamma = f_{AOM} / f_{FSR} = 1.6683(3) \times 10^{16} \) Hz/s is called the chirp rate of the FSF laser [220]. The measured value \( f_b = 28.95(6) \) MHz corresponds to an arm length difference of 260.12(5) mm. This value is consistent with a direct length measurement using a ruler, demonstrating the use of our FSF laser as range finder [220]. It was shown in [201] that the peaks in the RF noise spectrum are of Gaussian shape with a width related to the effective photon lifetime inside the cavity. Figure 6.3a2 shows the peak at \( f_{FSR} \) with higher resolution. The FWHM of the peak is 430(7) kHz and hence the lifetime of a photon in the cavity is \( t_{coh} = 2.33(4) \) \( \mu \)s.

**Figure 6.3**: RF noise spectrum of the Michelson interferometer output. (a) Output of the modeless broadband laser. (a2) Zoom on the peak at \( f_{FSR} \) and Gaussian fit. The width of the Gaussian is 430(7) kHz. (b) Output of the FSF laser seeded with 450 \( \mu \)W cw narrow-band light. (b2) Zoom on the peak at \( 3f_{AOM} \). The half-width at half-maximum is (30±2) Hz.
6.3.2 Externally seeded laser

In the experiments described so far spontaneous emission is the only source of seed for the FSF laser. Now the laser is seeded with a small amount of narrow-band cw radiation. The cw seed introduces a mode with fixed frequency into the ring cavity from which the amplification and frequency shifting process starts. A sequence of comb lines alongside the modeless broadband emission of the FSF laser is expected to appear [201], which we demonstrate experimentally in the following. Since the AOM shifts light to higher frequencies, the seed laser frequency is set to a value close to the low-frequency edge of the output spectrum of the FSF laser without external seed. The spectrum changes compared to the case without external seed, as shown in Fig. 6.2 for two seed powers. For 460 µW seed power a sharp increase in intensity appears at the frequency of the seed laser and extending 50 GHz to higher frequencies. For even higher frequencies the spectrum is similar to the spectrum of the modeless FSF laser. If the laser is seeded with higher power (720 µW, red curve), the feature near the seed frequency decays faster, and the lobe at higher frequencies shrinks substantially and the overall width decreases.

Compared to the situation without external seed, the RF spectrum at the output port of the Michelson interferometer (Fig. 6.3b) contains additional sharp lines at integer multiples of $f_{AOM}$. Figure 6.3b2 shows a high-bandwidth measurement of such a line at $3f_{AOM}$. The FWHM width is less than 100 Hz and therefore narrower than the broadband emission peaks shown in Fig. 6.3a2 by four orders of magnitude. Figure 6.4 shows the spectrum of a frequency comb spanning 370 GHz. To obtain a frequency comb over this larger spectral width compared to before we find it necessary to increase the AOM frequency and diffraction efficiency. In the following we worked with $f_{AOM} = 370$ MHz and optimized the diffraction efficiency to 87%. The power of the seed laser at the TA input facet is set to 80 µW. The mode structure of the cw seeded FSF laser is resolved by means of the beat measurement with the reference ECDL. The inset shows an example RF spectrum of the beat photodiode signal. Every comb line at $f_n = f_{seed} + n \times f_{AOM}$ of the FSF laser beats with the reference laser at frequency $f_{ref}$, leading to peaks (dashed red) at $|f_n - f_{ref}|$. The other features in the inset are beat signals between comb lines at $m \times f_{AOM}$ and more noisy peaks at multiples of $f_{FSR}$. The reference laser frequency $f_{ref}$ is now incremented in 5 GHz steps. The height of the strongest beat signal between the reference laser and the FSF output in a range from
DC to 2 GHz is plotted against the optical frequency of the reference laser in Fig. 6.4. We observe that the spectral weight of the frequency comb decreases roughly exponentially with frequency. For comparison we also show the spectrum obtained by the grating spectrometer in Fig. 6.4 (solid blue). Since this spectrum shows the sum of the modeless broadband spectrum and the frequency comb, we observe that the ratio of broadband intensity to comb intensity increases with higher frequency. This behavior is a consequence of the accumulation of amplified spontaneous emission for higher frequencies.

Although the presence of a narrow-band cw seed adds a sequence of equidistant modes to the spectrum, the signature of broadband emission is present in the spectrum for all seed intensities explored here. In contrast to a conventional laser where a single mode can dominate the spectrum due to exponential amplification [221], in an FSF laser single modes cannot be favoured. The frequency shift in each round trip does not allow for constructive interference. Instead spontaneous emission as well as a cw seed is amplified in the FSF laser. Therefore the discussed spectral features, the frequency comb and the broadband emission, coexist in the laser when seeded with a cw source.
To characterize the relative importance of the two spectral features, we measure the intensity of the corresponding RF peaks (see Fig. 6.3a2 and 6.3b2), in dependence of the seed power. For each seed power the power of the light circulating in the cavity is adjusted to a safe, fixed value using the intracavity half-wave plate. We record the height of the RF peaks relative to the background at \( n \times f_{FSR} \) and \( m \times f_{AOM} \). The averaged values of each type of frequency component for \( m, n = 1 \) to 5 are shown in Fig. 6.5. Within the range of parameters examined the comb intensity increases exponentially with seed power, whereas the broadband emission is exponentially suppressed.

The comb can be stabilized in frequency by locking the seed laser to a desired frequency, e.g. a Rb spectroscopy line. In previous work such stabilization required an additional lock of a comb line to a reference laser, see [213, 222] and references therein.
6.4 Conclusion and outlook

We have demonstrated an FSF ring laser based on a tapered amplifier. The FSF laser emits coherent modeless broadband radiation with 120 GHz bandwidth and a coherence time of $2.3\,\mu$s. The center frequency of the broadband source is tunable over 1 THz. A frequency comb spanning up to 370 GHz was realized by seeding the FSF laser with a narrow-band cw source. Both spectral components existed simultaneously in the FSF laser and we measured their relative strength depending on seed power.

Since TAs are relatively cheap devices, available for many wavelengths, we expect that our work makes FSF lasers easily accessible to more researchers, leading to more applications of these devices. In contrast to other frequency combs[223, 208] our comb has a narrower spectrum. The comb covers 0.7 nm, which is a fraction of the 3dB gain bandwidth of our TA (11 nm). We observe that the spectrum can be broadened by increasing the AOM frequency, although less than proportional to that frequency. Broadening the comb width would be valuable to increase the range of applications of our scheme and could be a topic for further research. Still, a comb spanning hundreds of GHz is relevant to many applications and we plan to use the frequency comb of our FSF laser as frequency reference for photoassociation spectroscopy of RbSr molecules[224].
Conclusion and outlook

This thesis lays the foundation for the creation of ultracold polar RbSr molecules, which will be used for quantum simulation experiments in the future. The large electric dipole moment of the ground-state molecules will allow us to observe new quantum phases emerging from the long-range dipole-dipole interactions. The additional magnetic moment of the open-shell molecules will provide us an additional control parameter. Magnetic and electric fields could be used to engineer spin-dependent, long-range interactions and repulsive van-der-Waals interactions.

The pathway towards the ultracold polar molecules started with an experimental apparatus that creates quantum degenerate gases of Sr atoms. During my thesis the experimental apparatus was equipped with the ability to produce quantum gas mixtures of Sr and Rb atoms. Two isotopic combinations of Rb and Sr were Bose-condensed. Because of the favourable interspecies scattering properties the $^{87}$Rb-$^{84}$Sr mixture was chosen for further steps towards ultracold polar molecules. We proceeded to transfer the double BEC of $^{87}$Rb-$^{84}$Sr into an optical lattice with the goal of creating atom pairs trapped in the periodic potential and then associating them into molecules. The use of an optical lattice increases the association efficiency and lifetime of the molecules, because they are immobilized on the lattice sites suppressing lossy collisions. Before the lattice could be populated with a high number of doubly occupied sites, we had to adjust the trapping potential from which the Rb-Sr BEC was loaded into the lattice. The trapping potential was shaped to create an overlapping Rb-Sr BEC with the help of a bichromatic dipole trap. Loading the optical lattice from the bichromatic trap we obtain up to $3.5 \times 10^4$ lattice sites occupied by a single Sr together with a single Rb atom. The optical lattice filled with atom pairs is an ideal starting point for the association steps that transform the pairs into molecules in the ro-vibronic ground-state.

The association procedure will take place in two stages, because squeezing the wavefunction of free-atoms to the size of ground-state molecules in one step is unachievable. First weakly-bound molecules will be created and then the weakly-bound molecules will be transferred to the absolute ground-state. The non-magnetic nature of
the ground-state Sr atoms puts obstacles in the way of using the conventional Feshbach association technique to create weakly-bound molecules. The Feshbach resonances are predicted to be very narrow and would require a great experimental effort to be used for the magneto-association. Instead of magneto-association we chose to use STIRAP for the first step of molecule creation. In STIRAP we use two lasers coupling the atom pairs to the weakly-bound molecule via an intermediate molecular state. In order to be able to use this technique, precise knowledge of the binding energies of the intermediate state and the weakly-bound molecule has to be retrieved. We used the well-known method of photoassociation spectroscopy to obtain the necessary knowledge. In addition to finding a pathway for STIRAP, our spectroscopy studies allowed the precise determination of the interspecies scattering length between all isotopic combinations of Rb and Sr. We measured the change of the hyperfine splitting of $^{87}$Rb caused by the presence of a $^{88}$Sr atom. We also understood, why our attempts to create weakly-bound molecules of $^{87}$Rb$^{84}$Sr were not successful but should be successful in the isotopic combination $^{87}$Rb$^{87}$Sr.

Finally we built and characterized a frequency-shifted feedback laser based on a tapered amplifier. The output of such a laser exhibits equidistant, discrete modes spanning hundreds of GHz. This frequency comb-like spectrum can be used to reference the frequency of a spectroscopy or STIRAP lasers to existing lasers in the lab with a bandwidth relevant for photoassociation of near-dissociation threshold molecular states. The device could speed up future photoassociation studies and could be used to lock STIRAP lasers to obtain molecules with binding energies of hundreds of GHz.

The PhD students continuing with my work modified the experimental apparatus to produce a $^{87}$Rb-$^{87}$Sr mixture while I wrote the thesis. Already they found suitable molecular resonances with the help of photoassociation spectroscopy. The one-colour resonances and two-colour resonances are strong. The next step is the creation of a quantum gas mixture and the loading of an optical lattice. Then the weakly-bound molecules can be created with the help of STIRAP. In order to transfer the molecules to the absolute ground state, spectroscopy of deeply bound states has to be performed. The STIRAP pathway suggested in [225] together with spectroscopic data of RbSr molecules on helium droplets [175, 226] points at the range of binding energies my fellow PhD students should consider. If we can identify a suitable STIRAP path, we will be able to obtain polar, open-shell molecules at ultracold temperatures.
Bibliography


[53] private communications with the authors of [51].


Summary

Fascinating quantum physics phenomena such as super-conductivity and quantum magnetism often arise from strong interactions between the particles involved in the phenomena. Because of the strong interactions many particles influence each other at the same time. The phenomena cannot be understood by simply studying the behaviour of a single particle. One must consider the whole ensemble of particles that make up such a many-body system. Studying many-body phenomena emerging from the collective behaviour of quantum particles is not straight forward. Although one can formulate the equations that describe a many-body system easily, solving these equations becomes impossible above a certain number of particles involved. Solving the equation of motion for one particle requires the information about all particles that interact with this one particle. Even the most powerful super computer runs out of computational power when solving the equation for about thirty interacting spin-1/2 particles. One possible ansatz to study many-body systems is quantum simulation. Simulation is used frequently to find out information about a hard to access problem. Astronauts routinely simulate space-walks in a swimming pool and practice in this almost gravity-free environment for a later mission outside a space-station. In quantum simulation one uses a quantum system in the laboratory that can be controlled very precisely to mimick a quantum system that needs to be understood. The quantum simulator is set up for a simulation by forcing the quantum particles, that make up simulator, to interact with each other in a similar way as the system one would like to study. Then, by measuring the properties of the simulator, one obtains information about the properties of the actual system.

Our experiment is designed to perform quantum simulation experiments in the future. The quantum simulator will use molecules comprised of one strontium and one rubidium atom to mimick quantum particles that interact strongly and show emergent many-body phenomena. The molecules have a dipole moment that can be used to make them interact over long distances. In order to bring out the quantum nature of the molecules one has to make them very cold. The temperature should be below
1 \mu K otherwise the thermal motion will drown out the delicate quantum properties. Instead of cooling the molecules directly we use atoms that we can cool with the help of lasers well below 1 \mu K and then associate the atoms into molecules without heating them up. Before I started my thesis the experimental apparatus was capable of cooling strontium atoms to ultracold temperatures and create Bose-Einstein condensates (BEC), which are quantum gases of perfectly synchronised atoms. During the thesis I managed to add rubidium into the experimental apparatus and cool it with lasers. I obtained a double BEC consisting of the two atomic species. The next step was to create the molecules from the atoms in the BECs. First I loaded the BECs into an optical lattice, which is a trap for atoms looking like an egg-carton box. In each lattice site one rubidium and one strontium atom was stored. These atom pairs can be associated to molecules with the help of laser radiation. To find the right frequency for the association lasers I performed spectroscopy on rubidium-strontium mixtures. The spectroscopy results showed on the one hand, that the association of the atoms will be very hard. On the other they showed, that choosing the right isotope of strontium and making a new quantum gas mixture with rubidium and that isotope could lead to a successful production of RbSr molecules. Then these molecules could be used for quantum simulation of many-body systems in the future.
Samenvatting

Fascinerende kwantummechanische fenomenen zoals supergeleiding en kwantum-magnetisme ontstaan vaak uit sterke wisselwerkingen tussen de deeltjes, die in de fenomenen betrokken zijn. Door de sterke wisselwerkingen beïnvloeden de deeltjes elkaar tegelijkertijd. Deze fenomenen kunnen niet begrepen worden door het bestuderen van de interactie van slechts twee deeltjes. Men moet de hele verzameling van deeltjes, die deel uitmaken van een dergelijk veeldeeltjessysteem, meenemen. Het bestuderen van dit soort veeldeeltjessystemen, die ontstaan door het collectieve gedrag van een verzameling kwantum-deeltjes, is niet eenvoudig. Alhoewel men de vergelijkingen die dit soort deeltjessystem beschrijven makkelijk kan formuleren, is het oplossen van deze vergelijkingen onmogelijk als het aantal van deeltjes te groot wordt. Het oplossen van de vergelijking voor één deeltje vereist informatie over alle andere deeltjes die met het eerste deeltje “in contact staan”. Zelfs de krachtigste supercomputer heeft te weinig rekenkracht om deze vergelijkingen op te lossen voor ongeveer dertig wisselwerkende spin-1/2 deeltjes. Eén mogelijkheid om veeldeeltjessystemen te bestuderen is kwantumsimulatie. Simulatie wordt vaak gebruikt om informatie te vinden over een moeilijk toegankelijk probleem. Astronauten simuleren routine-matig “space-walks” in een zwembad en oefenen in dit omgeving voor een latere missie buiten een ruimte-station. In kwantumsimulatie gebruikt men een gecontroleerd kwantumsysteem in een laboratorium, om het gedrag van andere systemen te imiteren in de hoop ze beter te begrijpen. De kwantumsimulator forceert de kwantumdeeltjes waaruit zij bestaat om interacties met elkaar aan te gaan op een soortgelijke wijze als in het systeem dat men eigenlijk zou willen bestuderen. Door het meten van de eigenschappen van de simulator verkrijgt men informatie over de eigenschappen van het ingewikkeldere systeem. Ons experiment is ontworpen om in de toekomst kwantumsimulatie-experimenten te doen. De kwantumsimulator zal moleculen gebruiken die uit één strontium en één rubidium atoom bestaan, om kwantumdeeltjes na te bootsen. Deze moleculen moeten sterke wisselwerkingen
en veledeeltjes-kwantumfenomenen laten zien. De moleculen hebben een dipoolmo-
ment, dat het wisselwerken over lange afstanden mogelijk maakt.

Om het kwantum karakter van de moleculen relevant te laten zijn moet men ze erg
koud maken. De temperatuur moet onder 1 µK zijn, ander zal de thermische beweg-
ing de delicate kwantumeigenschappen overstemmen. In plaats van het direct koelen
van de moleculen gebruiken we atomen die we heel goed met behulp van lasers onder
1 µK koelen kunnen en laten de atomen tot moleculen samensmelten zonder ze op te
warming

Voordat ik mijn werk met de experimentele opbouw begon, was men in staat om
strontium atomen tot ultralage temperaturen te koelen en om Bose-Einstein conden-
saten (BEC) te produceren - kwantumgassen, die uit perfect gesynchroniseerde atomen
bestaan. Tijdens de promotieperiode voegde ik rubidium atomen aan de experimentele
opstelling toe en koelde ze met lasers. Ik kreeg kwantumgassen, bestaande uit de
twee atomaire soorten, rubidium en strontium. De volgende stap om moleculen uit de
atomen in het BEC te creëren, is het laden van de BEC’s in een optisch rooster. In iedere
roosterplaats is één rubidium en één strontium atoom gevangen. Deze atoomparen
kunnen in een molecule omgezet worden met behulp van laserstraling. Om de juiste
frequentie voor de lasers te vinden, voerde ik spectroscopie op de rubidium-strontium
mengsels uit. De spectroscopie resultaten toonden enerzijds, dat de associatie van de
atomen zeer moeilijk wordt. Anderzijds toonden zij, dat met de keuze van een ander
isotoop van strontium een kwantum-gasmengsel van rubidium en dit isotoop mogelijk
is dat tot een succesvolle productie van RbSr moleculen kan leiden. In de toekomst
kunnen deze moleculen voor de simulatie van kwantum veledeeltjesystemen gebruikt
worden.
List of publications

First author publication

Co-author author publication

In preparation
A. Bayerle, A. Ciamei, C.-C. Chen, B. Pasquiou and F. Schreck, “Double species Mott-insulator of Rb and Sr” (2017), based on chapter 4 of this thesis.
A. Bayerle, A. Ciamei, S. M. Tzanova, B. Pasquiou and F. Schreck, “Photoassociation spectroscopy of RbSr molecules” (2017), based on chapter 5 of this thesis.
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