Vortex based atom traps and atomic electrometry on a superconducting atom chip

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Vortex Based Atom Traps and Atomic Electrometry on a Superconducting Atom Chip

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BY

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*To my beloved parents, brothers and sister who are always
there for me,*

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Abstract

This thesis focuses on the realization of the magnetic micro-trap using the vortex current in a superconducting structure and the measurement of the electric field from the adsorbates. Persistent currents are induced in the type-II superconductor - yttrium barium copper oxide (YBCO) to generate a magnetic potential for the ultracold atoms. Various programmable magnetic potentials are accomplished through the manipulation of the multiple magnetic field pulses sequence used in imprinting the vortices. A special example is the achievement of self-sustaining trap which its trapping potential is formed without any requirement for external magnetic field. A demonstration of a single trap to a four traps is also shown, indicating a possible interferometry type of experiment using superconductors. Afterwards, a focus is shifted from the magnetic field towards the electric field of the superconducting atom chip. By using the Rydberg state of the rubidium atom, we perform Rydberg spectroscopy to measure the electric field above the atom chip at room temperature and cryogenic temperature. In addition, to study the temperature dependent adsorption, temperature is varied from room to cryogenic temperature. Model based on Langmuir equation is simulated to explain the experimental result. This thesis concludes with a summary and the future outlook.

List of Publications

1. T. Müller, B. Zhang, R. Fermani, **K. S. Chan**, Z. W. Wang, C. B. Zhang, M. J. Lim, and R. Dumke,
Trapping of Ultra-Cold Atoms with the Magnetic Field of Vortices in a Thin-Film Superconducting Micro-Structure,
New Journal of Physics, 12, 043016 (2010).
2. T. Müller, B. Zhang, R. Fermani, **K. S. Chan**, M. J. Lim, and R. Dumke,
Programmable Trap Geometries with Superconducting Atom Chips,
Physical Review A, 81, 053624 (2010).
3. M. Siercke, **K. S. Chan**, B. Zhang, M. J. Lim, and R. Dumke,
Superconducting Atom Chips,
Quantum Electronics Conference & Lasers and Electro-Optics (CLEO/IQEC/PACIFIC RIM), pp. 907-09 (2011).
4. B. Zhang, M. Siercke, **K. S. Chan**, M. Beian, M. J. Lim, and R. Dumke,
Magnetic Confinement of Neutral Atoms Based on Patterned Vortex Distributions in Superconducting Disks and Rings,
Physical Review A, 85, 013404 (2012).
5. M. Siercke, **K. S. Chan**, B. Zhang, M. Beian, M. J. Lim, and R. Dumke,
Reconfigurable Self-Sufficient Traps for Ultracold Atoms Based on a Superconducting Square,
Physical Review A, 85, 041403 (2012).
6. **K. S. Chan**, M. Siercke, C. Hufnagel, and R. Dumke,
Adsorbate Electric Fields on a Cryogenic Atom Chip,
Physical Review Letters, 112, 026101 (2014).

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Chapter 1

Introduction

Atom chips are engineered with surface fabricated micro-structures. These devices have shown great capability, functionality and versatility in cooling, trapping and manipulating ultracold atoms [1]. Benefiting from the existing mature and advanced technology in semiconductor industry, atom chips can be designed and fabricated with various degrees of complexity using different types of materials (metals, insulators, semiconductors, permanent magnets, etc) to achieve the desired functionality [2]. Atom chips have been used to achieve Bose-Einstein condensates (BEC) (using normal conducting wires [3, 4], permanent magnets [5, 6] and superconductors [7]), to realize an atomic clock [8, 9], to study a one-dimensional Bose gases system [10], to study atoms-surface interactions [11], to realize an interferometer [12], to build a surface ion trap [13, 14] and implementing various atom optical elements (mirrors [15], diffraction gratings [16] and lenses [17]). Thus, the use and application of atom chips are wide ranging and their future prospects in the research are promising.

Atom chips based on normal conducting structures have severe drawbacks like spin-flip loss, heating, decoherence and fragmentation [18, 19]. Spin-flip loss is caused in a magnetic trap due to spin-flip transition from a trapped to an untrapped magnetic sublevel [20]. Two main contributing factors to this loss rate are the thermal noise (or Johnson-Nyquist noise) and the technical noise. Thermal noise originates from the random motion of the charge carriers inside an electrical conductor at a certain temperature which induce electromagnetic field fluctuations [21]. The technical noise is attributed to the current fluctuations of the external power supply [22]. In addition, if the thermal noise and technical noise have frequency components at the harmonics of the trapping frequency, they will be able to excite the trapped atoms to higher vibrational state and give rise to heating [23, 24]. Subsequently, if the cloud temperature rises above the trap depth, it will also be lost from the trap [24]. Irrespective of thermal or technical noise, the magnetic field fluctuation leads to a fluctuation of the energy of the Zeeman sub-level [25]. This can decrease the coherence time for superposition state between Zeeman sub-levels which could be a serious drawback for applications in the field of quantum information processing and quantum simulation [25]. In some circumstances, due to surface and edge roughness on the current-carrying wires, there is a distortion to the current flow which leads to corrugated trapping potential [24]. It has been shown by experiment that the atomic cloud (both thermal cloud [19, 26, 27, 28] and BEC [19, 24, 29, 30]) fragments in the vicinity of the current-carrying normal conducting wires [19, 24, 28, 29, 30] and

permanent magnets [26, 27].

There were some attempts to solve these different issues. A reduction of technical noise can be achieved by imposing a stringent requirement for the current stability (fluctuation less than $1 \mu\text{A}$) [22] of the power supply used. Nevertheless, to avoid the cables used from picking up external RF signals, their length are to be minimized [24]. For avoidance of fragmentation, Pritchard, D. E.'s group used a dipole trap and showed its lack of fragmentation compared to magnetic microtraps [24]. The reason was attributed to the absence of current flow in the wires [24]. The group by I. Bouchoule eliminated the fragmentation by modulating the current at tens of kHz to average the potential to zero [31]. It is also possible to reduce the surface and edge corrugation through the use of advanced fabrication techniques [32]. In terms of decoherence, Treutlein, P. *et al.* increased the coherence time by applying a homogeneous “magic” magnetic field of 3.23 G and chose two insensitive magnetic sublevels of ^{87}Rb as the qubit states [8]. Thermal noise can be reduced by reducing the wire thickness so it becomes smaller than the material's skin depth [21]. Alternatively, it can also be cooled to lower temperature in order to suppress the thermal fluctuations [21] even though it was found not being able to be eliminated completely [33]. To circumvent this problem, there were suggestions to optimize the choices of materials used [34]. It can be done through the trapping of atoms near insulators [35] or replace the trapping wires with alloys [34], permanent magnets [36] or superconductors [37].

Using superconductors in atom chip will prevent spin-flip loss, heating, decoherence and fragmentation all at once. The spin-flip loss rate due to thermal noise is suppressed for atoms trapped near the superconductors (niobium [37, 38, 39] and YBCO [37]). Numerical simulation shows that the lifetime of an atomic cloud near superconductors is several orders of magnitude longer compared to normal conductors [37, 39, 40]. Also, the heating rate for atoms trapped using superconductors (both niobium and YBCO) are computed to be lower compared to metals [37]. In addition, decoherence is evaluated to be negligible even at distance as close as $1 \mu\text{m}$ [39]. To overcome the spin-flip loss of atoms due to technical noise, superconductor offers a possible solution by eliminating any use of an external current source. It can be done by inducing a persistent current in a superconducting closed loop through field cooling (FC) [41, 42]. If a magnetic field is applied perpendicular to a superconducting closed loop during the cool down, the magnetic flux will be freezed inside this closed loop [41, 42]. In type-II superconductors (strip [43], disk [44] and ring structures [44]), persistent currents can be induced by applying a magnetic field pulse above first critical field B_{c1} but below second critical field B_{c2} . These currents are mediated by vortices. Using advanced fabrication techniques, superconductors can be made to have negligible roughness and thus be able to overcome fragmentation [45]. In addition, there is an added benefit owing to the cryogenic environment required for superconducting atom chips. The vacuum pressure is lower compared to room temperature setup due to cryogenic pumping. This reduces the background gas collision and with this increases the lifetime of the trapped atoms up to minutes [46].

The first superconducting atom chip was realized in 2006 by a group led by Haroche, S. at ENS France using niobium (Nb, critical temperature $T_c = 9.2 \text{ K}$) at liquid helium temperature [47]. A year later, a Japanese group from NTT Basic Research Laboratories demonstrated a successful trapping of ultracold atoms with superconductor magnesium diboride (MgB_2) which has a T_c of 39 K [42]. In the year 2008, a group from Tübingen

also achieved a superconducting atom chip using niobium [48]. In the same year, our group also fabricated a superconducting atom chip. Our atom chip is based on yttrium barium copper oxide ($\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$) which has a T_c of 87 K [45]. The T_c is high enough to be reached using liquid nitrogen. Thus, this condition makes our cryogenic system less stringent and cheaper compared to a liquid helium cryogenic system [49, 50, 51].

Roux, C., *et al.* [7] and Imai, H., *et al.* [52] had both achieved Bose-Einstein condensation (BEC) using superconducting atom chips. The lifetime of atoms near superconductor was measured to be longer compared to normal conductors (copper) due to the suppression of thermal noise [53]. However, in the case of the flux penetrated superconductors, the atoms loss rate was dominated by vortex dynamics but still the lifetime was longer compared to normal conductors [54, 55, 56]. Heating rate was also measured to be negligible at a distance as close as $25\text{ }\mu\text{m}$ [53]. In addition, the coherence time for atoms near superconductors was measured to reach 20.5 s [57]. Mukai, T., *et al.* demonstrated the controllable reduction of the persistent current in milliseconds by locally heating the superconductor with a focused laser beam [42]. For our group, initially, we characterized the magnetic micro-trap formed by the magnetic field induced vortex current on a type-II superconducting strip [58]. By using the memory property of this superconductor [59], we showed that this vortex current is programmable and can form complex spatial current density pattern leading to complex trapping potentials [60]. We even achieved a self-sustaining trap on a superconducting square structure, that is a magnetic potential formed solely from the vortex current alone without any use of external magnetic field [61]. Even though the use of superconductors present these advantages compared to normal conductors, there are certain specific properties need to be taken into account. Due to the presence of Meissner effect, the trap was measured to have shorter trap to surface distance and shallower trap depth compared to current-carrying normal conducting wires [48]. This can give rise to atoms loss through evaporation [48]. In type-II superconductor, at temperatures below the dendritic instability temperature T_a (niobium, $T_a = 7\text{ K}$ [62]; MgB_2 , $T_a = 10\text{ K}$ [62]; YBCO, $T_a = 35\text{ K}$ [58]), vortex avalanches can lead to a loss of atoms [62].

So far, our discussion is focused on the magnetic field of the superconducting atom chips. There are proposals to interface neutral atoms [63] or polar molecules [64] to the superconducting solid state system via electric or electromagnetic field coupling [65]. However, in order to achieve this coupling, any stray electric fields from the environment have to be minimized or eliminated. One particular source of stray electric field is the electric field produced by the adsorbates from the atoms adsorbed on the atom chip surface either through chemisorption or physisorption. By characterizing the perturbations to the magnetic trap potential, McGuirk, J. M., *et al.* [66] measured the electric field of rubidium atoms deposited and get adsorbed on metal (titanium), semiconductor (silicon) and insulator (BK7) substrates. The same group also demonstrated the measurement of the vector nature of the electric field by driving the trapped atoms into a mechanical dipole oscillation for adsorbates on fused silica, yttrium, hafnium and lutetium surfaces [67]. In addition, electromagnetic induced transparency (EIT) spectroscopy involving the Rydberg states of ultracold atoms was also employed to map out the spatial profile of an electric field in proximity to a gold (Tauschinsky, A., *et al.* [68]), fused silica (Abel, R. P., *et al.* [69]) and copper (Hattermann, H., *et al.* [70]) surface. In most of the experimental studies, metallic substrates were used [66, 67, 68, 70]. Only a few experi-

ments were conducted with semiconductors and insulating substrates at room and higher temperatures [66, 67, 69]. These experiments demonstrated the formation of adsorbates due to chemisorption on metallic substrates [66, 67, 68, 70]. At room temperature, physisorption with van der Waals interaction played little role in generating an electric field on an insulating substrate [66]. Thus, insulators are proposed to be used in atom chip experiments to overcome adsorbates electric fields [66]. However, this breaks down at cryogenic temperature for superconducting atom chip experiment where physisorption plays a more dominant role. This physisorption process at cryogenic temperature was primarily investigated by our group [71].

My PhD thesis is divided into two parts. The trapping of ultracold atoms using type-II superconductor is described in Part I while the investigation of electric field emitted by adsorbates is described in Part II. Chap. 2 describes our experimental setup including the optical apparatus, vacuum system, electronics, computer control, image acquisition etc. Part I has two chapters. The theory for superconductors is covered in Chap. 3. This chapter introduces also the critical state model to describe the mesoscopic behaviour of induced vortices inside the superconductor. Furthermore, it shows our simulation using this model to describe the various complex trapping geometries. These theoretical descriptions serve as a background knowledge for our experimental realization of magnetic micro-traps using the vortex current as described in Chap. 4. This chapter discusses the manipulation and characterization of the properties of various trapping potentials on a superconducting strip and square structure. Part II has three chapters. Chap. 5 refers to the theory of Rydberg atoms. This chapter begins with the description of the various properties of the Rydberg atoms. Then, I proceed to describe the numerical technique to simulate the rubidium wavefunction. The simulated wavefunctions are then applied to the calculation of the radial matrix elements which are then used for the generation of the rubidium Stark map. Chap. 6 discusses the theory for chemisorption, physisorption and the temperature dependence behaviour of adsorption phenomenon. The last chapter, Chap. 7 shows the experimental results obtained from the measurement of the electric field at room to cryogenic temperatures. A theoretical model based on Langmuir equation is built to explain the temperature dependence. I conclude this thesis and write the future prospects in Chap. 8.

Chapter 2

Experimental setup

2.1 Introduction

This chapter describes the experimental setup of our atom chip experiment. Sec. 2.2 discusses the various structures on our atom chip. Subsec. 2.2.1 shows the relevant electrical properties of our YBCO superconductor. Subsec. 2.2.2 the fabrication procedure of our atom chip, patterning of the YBCO structures will be discussed. With the use of the optimized fabrication scheme, Subsec. 2.2.3 discusses the results by characterizing the surface roughness of the YBCO structures. The mounting of the atom chip on the cold finger is described in Sec. 2.3.

Our vacuum system is mentioned in Sec. 2.4 and 2.5. It is followed by a discussion of our laser system in Sec. 2.6. Sec. 2.7 discusses the circuit design of the power supplies.

The imaging setup, the images acquisition procedure and the images processing are discussed in Sec. 2.8. The computer control is discussed in Sec. 2.9. The last section, Sec. 2.11 shows the experimental sequences of acquiring a magneto-optical (MOT) trap, molasses cooling and the transport of ultracold atoms to the atom chip.

2.2 Atom chip design

The atom chip was manufactured following our design by an external company (Theva Duennschichttechnik). The structures in our atom chip are based on a high temperature superconductor, yttrium barium copper oxide (abbreviated as YBCO and its molecular formula is $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$) patterned onto a 20 mm $\times$ 20 mm yttria-stabilized zirconia (YSZ) single crystalline substrate. The schematic diagram of our various structures (gray) are shown in Fig. 2.1a. The square structure in Fig. 2.1b has a length of 1 mm. Our Z-type structures have various sizes. Shown in Fig. 2.1c, the sizes are 400 μm , 200 μm and 50 μm and all three have an axial length of 5 mm. The three Z-wires are separated from each other with a gap size of 20 μm . There is a dimple wire with a width of 200 μm crossing perpendicularly to all the Z-wires. All the YBCO structures on our chip are fabricated with a thickness of 800 nm.

Figure 2.1: a) Schematic diagram of our superconducting atom chip structures. Dashed box refers to the superconducting square structure and Z-structures used in our experiment. [Zoom in] b) 1 mm $\times$ 1 mm superconducting square structure d) Z-wires of different sizes together with a dimple wire on our atom chip.

Figure 2.2: Plot of the critical current density versus temperature of our YBCO using a four-point resistance measurement method. The wire used has a width of $26 \mu\text{m}$. The superconductivity begins at 87.1 K. Graph adapted from thesis by Wu, X. [73].

2.2.1 Superconducting material properties

The YBCO used in our experiment has a critical temperature, T_c of 87 K. The T_c of YBCO is higher than the boiling point of liquid nitrogen, 77 K at atmospheric pressure. This characteristic enables the use of liquid nitrogen as a refrigerator coolant. Other type-II superconductor also used in superconducting atom chip experiments like niobium ($T_c = 9.2$ K) [47, 48] and MgB_2 ($T_c = 35$ K) [42] require liquid helium. With the use of liquid nitrogen, we do not need a closed cycle refrigerators. This reduces the cost significantly and minimizes the complications of the experimental setup.

The critical current density, J_c of our YBCO at 83 K is 1 MA cm^{-2} . The measured critical current density with respect to temperature using four-point resistance measurement method is plotted in Fig. 2.2. With the various sizes of our Z-wires as shown in Fig. 2.1c, the critical currents are calculated to be 3.2 A, 1.6 A and 0.4 A. In the real experiment, there exists a temperature gradient across the chip. Thus, the critical current can be lower than calculated. These calculated critical currents have similar magnitude as a typical atom chip using normal conductor [72], serving as a promising tool for magnetic microtrap for ultracold atoms.

In addition to J_c , there are two critical magnetic fields, lower critical field, B_{c1} and upper critical field, B_{c2} associated to a type-II superconductor. The values for B_{c1} and B_{c2} of YBCO depend on its chemical elements composition and its temperature. For anisotropic high temperature superconductor, B_{c1} is dependent on the field direction with respect to the crystal structure plane. Taking an example of YBCO from reference [74], $B_{c1}(B_{//ab}, T \rightarrow 0) = 25 \text{ mT}$ and $B_{c1}(B_{//c}, T \rightarrow 0) = 90 \text{ mT}$ while B_{c2} is of the order of 100 T [75]. $B_{c1}(B_{//ab})$ and $B_{c1}(B_{//c})$ are the temperature dependent lower critical fields along the ab -plane and c -axis of the YBCO crystal structure. For our YBCO,

$B_{c1} \approx 25$ G at 83 K [58].

YBCO is extensively studied in the field of high temperature superconductors with its potential application in electrical power transmission [76]. Up to this day, there are some high temperature superconductors with better properties than YBCO with respect to the parameters discussed above. Nevertheless, the choice of our superconductor is decided by its availability commercially at a reasonable price due to the existing well established fabrication technology.

2.2.2 Fabrication of atom chip

There are two main procedures in manufacturing the superconducting atom chip. The first procedure is the deposition of the superconducting materials. The second procedure is the structure patterning. The YBCO is grown as an epitaxial thin layer on a YSZ substrate. With this, the crystal direction of the deposited superconducting materials form a defined orientation with respect to the YSZ crystalline substrate. To grow homogeneously, the lattice parameters of YBCO and YSZ are matched. The growth of YBCO has to be carefully controlled in order to match the temperature expansion coefficient of the YBCO thin film and substrate.

Different methods are combined and used to pattern the YBCO layer. Optical lithography technique for the deposition of photoresist and application of a mask before being etched using wet chemical etching. However, the spatial resolution for the optical lithography technique is only $1\ \mu\text{m}$ (E-beam lithography and ion-milling techniques can design patterns of sizes less than $1\ \mu\text{m}$ [77]). We have also employed femtosecond laser ablation to decrease the edge roughness. Focused laser pulses ($2\ \mu\text{J}$, 130 fs, centre wavelength of 800 nm) are used to remove some local regions of the YBCO layers. the pulse length of the laser has to be short to prevent heating. Heating of YBCO may lead to oxygen loss and changes its superconducting properties.

2.2.3 Surface roughness of the YBCO structure

The YBCO structures patterned with optical lithographic methods are smoother compared to the one using femtosecond pulse laser ablation. Still, the laser ablation technique is used locally to smoothen the imperfect rough regions of the lithographic patterned structures.

Surface roughness of the current carrying wires on the atom chips are found to lead to potential corrugation. This is shown in the fragmentation of thermal cloud and BEC trapped at close distance to the surface of the current carrying wires [19]. Different schemes are used to overcome the potential roughness [31, 32]. In our case, we manipulate and optimize the fabrication techniques and processes to decrease the surface roughness as mentioned in Subsec. 2.2.2.

To characterize the surface roughness of the YBCO structures, we perform an atomic force microscope (AFM) scan of our chip. Fig. 2.3a is an AFM image obtained by scanning a $10\ \mu\text{m} \times 10\ \mu\text{m}$ surface area of the YBCO wire. The surface roughness has a root-mean-square value of 2.3 nm. This low value is obtained because YBCO is grown

Figure 2.3: AFM images with scan performed on a $10\text{ }\mu\text{m} \times 10\text{ }\mu\text{m}$ surface area of our a) YBCO wire. b) At the edge of the YBCO wire. (The straight lines seen in the image are formed due to scanning of the AFM tip.) Graphs adapted from [45].

epitaxially on the substrate which can prevent the formation of grain structures. Fig. 2.3b shows an AFM image obtained with the scan performed at the edge of our YBCO wire. The thickness of the YBCO structure is 800 nm.

2.3 Mounting our atom chip to the cold finger

Fig. 2.4a shows the wire clusters clamped with a few copper rings to the walls of the cold finger. Since the wire clusters are to be connected to external current sources at room temperature, this design can minimize heat transfer to the chip. The ends of the wire clusters are inserted to the gold pins and clamped tightly.

Fig. 2.4b shows the mounted atom chip to the cold finger. The atomic chip is held above a chip holder (copper block). 4 stainless steel screws are used to screw the 4 copper chip clamps which result in pressing the atom chip on the copper block. To ensure good thermal contact, an indium layer is sandwiched in between. This copper block is then tightened directly to the cold finger using another 4 stainless steel screws. A structure made of Macor is used to insulate the electrical contacts from the cold finger Fig. 2.4a. These electrical contacts are then connected to the gold pads on the atom chip with thin gold wires.

2.4 Vacuum system

Our vacuum system consists of two ultra-high vacuum (UHV) pumps and one ionization gauge. We use one ion pump (Varian - VacIon Plus 75, CF6" Starcell) and one titanium sublimation pump (TSP Filament Cartridge on a 2 3/4" CFF - 9160050). The ion pump is turned on continuously to maintain the vacuum pressure of 4×10^{-10} mbar. When the vacuum pressure has increased and the lifetime of the ultracold atoms shorten, the TSP is turned on by resistive heating the titanium filaments. This is usually done 1-2

Figure 2.4: a) Picture of the wire clusters with its ends inserted into the gold pins. These wire clusters are pressed against the wall of the cold finger with copper clamps. b) Picture of our atom chip mounted on the copper chip holder which is then screwed to the cold finger.

times a year to replenish the titanium layers in the vacuum chamber's walls [78]. These titanium layers serve as pumping surface for adsorbing the residual gases (H_2 , N_2 , O_2 , etc) inefficiently pumped by the ion pump. The ionization gauge (Agilent Technologies - UHV-24P Bayard-Alpert Ion Gauges) measures the vacuum pressure. Our ionization gauge is at a distance from our atom chip. Thus, the vacuum pressure reading is no longer accurate as the value recorded contradicts lifetime measurements. Our measurement shows about 30 s lifetime for ultracold atoms in the magnetic trap. However, the vacuum pressure reading shows 5×10^{-11} mbar. For a background pressure of 10^{-9} mbar, one can expect a lifetime of a few seconds [79, 3]. If the background pressure is 10^{-11} mbar, the lifetime should be more than a minute [80].

2.5 Rubidium dispenser and LIAD

Our rubidium dispenser (Alvasources - AS-6-Rb-240-C) is purchased from Alvatec company. Usually, we apply 3.6 - 4.0 A to the dispenser to release rubidium to the vacuum¹. Once the vacuum pressure reaches $\approx 2.5 \times 10^{-9}$ mbar, the current is turned off. After some time, the rubidium atoms will gradually stick to the glass cell and the vacuum pressure will drop to its initial reading of $\approx 5 \times 10^{-11}$ mbar.

To prepare an ultracold atomic ensemble, we use light induced atomic desorption

¹Schematic of the circuit design in controlling the dispenser current is shown in Appendix A.

(LIAD), this offers a fast loading time and quick recovery of the vacuum pressure. During LIAD we illuminate the vacuum glass cell with UV light and the alkali atoms desorb from the surface [81]. LIAD is efficiently working for glass, pyrex, sapphire, stainless steel, and some organic coated films [82, 83, 84]. In our experimental setup, as the rubidium atoms are most likely sticking at the wall at the bottom of our vacuum glass cell, we place 6 UV LED array (Roithner LaserTechnik - RLT-MIL410-12V-30, violet, 410-415 nm, 12 x 120 mW at 4.2A, 3.7V, 30°) near this surface. They are switched on during the MOT loading process.

2.6 Cooling and repumper laser system

The laser systems in our experiment for cooling and repumping are commercial systems, Toptica - TA-100-780-02008 and Toptica - DL100. The cooling laser consists of two parts, the external cavity diode laser (ECDL) in Littrow configuration and a tapered amplifier (TA) to increase the power of ECDL to 1 W. The repumper laser is also an ECDL in Littrow configuration.

Shown in the picture in Fig. 2.5, the cooling and repumper laser system is placed at the ground level breadboard. The upper level breadboard is the optical setup for imaging beam, pumping beam, beat signal and spectroscopy for cooling and repumper laser. The schematic diagrams for the optical setup at the ground level and upper level are shown in Fig. 2.6 and Fig. 2.7. The laser output from the cooling laser system is split with a polarizing beam splitter (PBS). The transmitted beam is going through an AOM (AOM1, +2×10⁵ MHz) in double-pass configuration. This cooling beam that has been frequency doubled is then coupled to a polarizing maintaining fibre mounted to FP1. The reflected beam is then directed to the upper level.

The cooling laser beam after FP1 has a power of ~ 210 mW. As shown in Fig. 2.20a, it is directed to a series of half-wave plates and walk-off polarizers (WOP) to distribute equally the power of the cooling laser into six beam paths. Numbers 1-6 are labelled to denote the 6 beams. The out-coupled beams from 1-6 are transferred using a non-polarization maintaining fibre to the cage system holding the optical elements shown in Fig. 2.8b. Each beam is individually out-coupled using a fibre adaptor (FA) plate (Thorlabs - SM1FCA). A quarter-wave plate, a half-wave plate and a PBS ensures maximum output power followed by a quarter-wave plate to select an appropriate circular polarization for the MOT. A lens (125 mm focus, 50 mm diameter) is inserted between the PBS and the quarter-wave plate to collimate the diverging MOT beam before sending it off to the vacuum.

The output beam of the repumper laser is split with a PBS and then reflected through an AOM (AOM2, +80 MHz) that is used for switching purpose. The 1st order of the repumper beam is overlapped with the cooling beam and then coupled to FP1. The transmitted repumper beam from the PBS is then reflected to the upper level breadboard for spectroscopy.

As shown in Fig. 2.7, the cooling laser from ground level is split into different beams propagating in different directions with a few PBS. The cooling beam is initially split with a PBS. The reflected beam passes through an AOM (AOM5, +87 MHz) and it

Figure 2.5: Picture of the optical setup of our cooling and repumper laser system. The labels in red refer to some of the optical or electronic components in Fig. 2.6 and 2.7 respectively.

is modulated by a frequency supplied by a lock-in amplifier. Then, it is directed to a saturation spectroscopy setup (TEM Messtechnik - CoSy 5.0). The output signal from the CoSy box is a modulated absorption signal. A lock-in amplifier is used to demodulate this signal. Afterwards, the cooling beam is locked to the crossover peak, $|5S_{1/2}, F = 2\rangle$ to $|5P_{3/2}, F' = 2, 3\rangle$ of the D2 transition of ^{87}Rb with the use of a PI controller which provides feedback to the PZT of the cooling laser ECDL. On the other hand, the transmitted beam from the PBS is used for imaging purpose. It passes through an AOM (AOM3, $+2 \times 110$ MHz) in double-pass configuration and coupled to FP3. Part of the cooling beam is split for optical pumping. It passes through an AOM (AOM4, -46 MHz) and its -1st order is coupled to FP4.

The reflected repumper beam is guided to a saturated absorption spectroscopy. The transmitted repumper beam from the PBS is then overlapped with the cooling beam to create a superposition of the cooling and repumper beam. This overlapped beam is then coupled to FP2. The result for this overlap is a beat signal.

With the frequency of the cooling laser stabilized, the frequency of the repumper laser is locked with respect to it using trombone locking. The electronic scheme is shown in Fig. 2.9. The two beams are focused to a GaAs fast photodiode (Hamamatsu - G4176-03) producing an electronic 6.84 GHz beat signal. A bias tee (Pulsar BT-26463/1s) is placed between photo detector and amplifier. It is used to bias the Photodiode and a high pass filter (500 MHz - 10 GHz). This signal is amplified in a series of +10 dB amplifiers (Mini-Circuits - ZJL-7G). For monitoring purpose, a power splitter (Mini-Circuits - ZAPD-20-S+) is used to split part the signal to a spectrum analyzer. A second power splitter (Mini-Circuits - ZAPD-20-S+) is used, where the two outputs are recombined in a mixer (Mini-Circuits - ZFM-2000+). However, one of the outputs from the power splitter is passed through a variable phase shifter (ATM - PNR P1505 SER

Figure 2.6: Schematic diagram of the optical setup for the cooling and repumper laser system. These optical elements are placed at the ground level breadboard. Abbreviations for the optical and electronic elements are shown in Appendix B.

Figure 2.7: Schematic diagram of the optical setup used for imaging, optical pumping, saturated absorption spectroscopy (cooling laser and repumper) and repumping. These optical elements are placed at the upper level breadboard. Abbreviations for the optical elements are shown in Appendix B.

H781312z-01) for introducing an extra phase shift. The mixer mixes the two signals and produces an interference pattern. This signal is then fed to a PI controller. The error signal output from PI controller is feedback to the PZT of the repumper laser such that it can be locked to the transition from $|5S_{1/2}, F = 1\rangle$ to $|5P_{3/2}, F' = 2\rangle$.

The radio frequency used for the AOM and its corresponding addressing of the atomic level structure is shown in Fig. 2.10. Only the atomic transitions from $|5S_{1/2}, F = 2\rangle$ to $|5P_{3/2}, F' = 2\rangle$ and $|5P_{3/2}, F' = 3\rangle$ of ^{87}Rb are shown as other quantum states are non-relevant to our optical setup.

2.7 Power supply

We use five high current power supplies for our MOT coils, transfer coils and bias coils in x , y and z -direction ² (Fig. 2.20b). The electronic circuits to control these power supplies are shown in Fig. 2.11.

The electronic design for the current control of the MOT coils is shown schematically in Fig. 2.11a. The voltage of the power supply can be varied by the computer control. The value for this voltage is set such that output current can be driven by the power

²The jargons used in our experiment for x , y and z -direction are A-C, B-D, and E-F direction. These jargons are used in labelling the equipment and computer programme.

Figure 2.8: a) Schematic diagram showing the splitting of cooling beam into 6 beams. b) Schematic diagram showing the expansion of the MOT beam and formation of circular polarization for the 3 pairs of counter-propagating beams. Abbreviations for the optical elements are shown in Appendix B.

supply without limited by the voltage. The current for the MOT coils are adjusted using a MOSFET (IXYS - IXFN230N10) while the switching of the coils is done through an IGBT (PRX - CM600HA-24A). The MOSFET driver that controls the gate of the MOSFET is basically a comparator with a PI function that compares the Hall probe voltage and the computer analogue voltage. Thus, any resistance increase due to heating of the coils can be compensated accordingly by the MOSFET driver to maintain a constant current through the coils. Current to the coils is switched by an IGBT rather than through the power supply as the switching of the coils is faster. The IGBT is controlled through a TTL signal supplied by the computer control. To prevent the high back-emf during fast switching, a diode in series to a $1\ \Omega$ resistor is connected in parallel to the coils to form a closed loop. For the same reason, two varistors are used to connect them to the collector and emitter of IGBT. With this circuit design, the emitter of the IGBT has a floating voltage. This will not post a problem for a typical ultracold experiment where the crucial parameter is only magnetic field. However, in our Rydberg spectroscopy experiment as discussed in Chap. 7, the Rydberg state used is sensitive to electric field. The floating voltage will emit an electric field that shifts our Rydberg state out of resonance. To solve this, the emitter is grounded with a series of $1\ \text{k}\Omega$ resistor and a fuse. There is a maximum current limit even if the analogue voltage of the power supply is maximized. For some experiments, a higher current is required. For this reason, the MOT coils in red dashed box can be manually switched to a set of car battery current source. This is discussed later in Sec. 2.7.1.

The electronic circuits for the current control of transfer coils and bias coils are shown in Fig. 2.11b and 2.11c respectively. Compared to the electronic circuit for the MOT

Figure 2.9: Schematic diagram for the electronic circuits used for trombone locking of the repumper laser.

coils, the electronic circuit for transfer coils has an extra relay placed in series with the diode and $1\ \Omega$ resistor. A relay is used because our transfer coils are placed in adjacent to the MOT coils. The switch off of the MOT coil after MOT loading will induce eddy currents. This will inadvertently give rise to a magnetic field that is detrimental to the molasses. This relay is switched on during this time such that no eddy current can be produced in the transfer coils induced by the MOT coils. However, this relay will be switched off when a current is run through the transfer coils. In contrast to the electronic circuit for current control of MOT coils, the voltage for this power supply is manually fixed. It is manually fixed to its minimum such that the power supply is able to supply any current based on our experimental conditions without limited by the voltage.

The electronic circuits for the current control of the bias coils along x , y and z -direction are similar as the transfer coils except that there are no relays. The relevant parameters for the magnetic field generated by the MOT, transfer and bias coils are shown in Appendix C.

2.7.1 Current source using car batteries

Fig. 2.12 shows the current source to the MOT coils from car batteries. There are 6 batteries which can be connected in series. Depending on our needs, the batteries can be connected manually to provide various voltage sources of 12, 24, 36, 48, 60 and 72 V. For safety purpose, two relays (Tyco Electronics - EV200HAANA) and one MOSFET are required to close the circuit loop. The two relays are to be closed first using a Relay Control unit (32 V voltage supply). Afterwards, the pulse generator generates a single pulse (~ 300 ms) to close the circuit loop with the MOSFET. Even though the voltage of the car batteries are in discrete steps of 12 V, by supplying different voltage (Control

Figure 2.10: Frequency scheme for the cooling beam, imaging beam, optical pumping beam and frequency locking at the crossover transition of ^{87}Rb ($|5S_{1/2}, F=2\rangle$ to $|5P_{3/2}, F'=2,3\rangle$) implemented with AOMs.

Pulse Amplitude) to the solid state relay (SSR) (Crydom - DC60S5), we are able to generate variable current source from 0 - 270 A. The calibration between the hall probe voltage and the current is shown in Appendix D. Due to high temperature increase after each current run, a thermo-electric cooler (TEC) is used to cool the MOSFET. In case the relays remain closed for some reasons, an emergency stop (ES) is used to break the circuit.

2.8 Imaging system

As shown in Fig. 2.13, our absorption image setup consists of two achromatic lenses to minimize spherical and chromatic aberration. L1 has a focal length at the position of the atomic cloud while L2 is focused to the charge-coupled device (CCD) camera. There are three absorption imaging setups in our system. The first absorption imaging setup (L1 = 76.2 mm, diameter = 25.4 mm; L2 = 100 mm, diameter = 50 mm; Pixelfly QE Camera) is used to image the MOT, molasses and atomic cloud at the lower magnetic trap. The second (L1 = 75 mm, diameter 30 mm; L2 = 100 mm, diameter = 50 mm; Theta system - SIS1-s285M) absorption imaging setup images the atomic cloud ($\sim 50 \mu\text{m} \rightarrow \sim 1 \text{ mm}$ distance) on the Z-wires and square structure along y (axial direction of the Z-wires) with a small reflection angle on the chip structure. The third camera (L1 = 100 mm, diameter = 25.4 mm; L2 = 100 mm, diameter = 25.4 mm; Pixelfly QE Camera) is used to image along x -direction (transverse direction of the Z-wires) but with a high grazing angle. This is required as the presence of transfer coils block the imaging beam if it is

Figure 2.11: Schematic of the electronic circuits for current control of a) MOT, b) transfer and c) bias coils along x , y and z -direction.

Figure 2.12: Circuit diagram for the high current supply using car batteries supplied to the MOT coils. 6 batteries are used to be connected in series to generate voltage of 12, 24, 36, 48, 60 and 72 V.

aligned parallel to the chip surface.

2.8.1 Image analysis

We capture two images. The first image (real image), $I_{real}(x, z)$ is taken with the presence of atomic cloud for 100 μs . The second image (background image), $I_{bkg}(x, z)$ is taken one second after the first image is taken. At this time, the atomic cloud is gone and the second image is basically taking the background together with the imaging beam. A logarithmic plot of the ratio between these two images gives an optical depth, $OD(x, z)$ image [85, 86]. It is described by the following equation,

$$OD(x, z) = \ln \frac{I_{bkg}(x, z)}{I_{real}(x, z)} \quad (2.1)$$

For imaging beam intensity that is below saturation intensity, OD can be written as

$$OD(x, z) = n(x, z)\sigma \quad (2.2)$$

σ is the absorption cross section with detuning as the dependent parameter. For resonance case, detuning = 0. With $\lambda = 780$ nm, it is

$$\sigma = \sigma_0 = \frac{3\lambda^2}{2\pi} = 0.29 \mu m^2 \quad (2.3)$$

$n(x, z)$ is the atomic column density and is obtained from integration along the direction of the imaging beam. In our case, $n(x, z) = \int n(x, y, z) dy$. We can extract useful information about the atomic cloud based on $n(x, z)$ like the spatial density distribution, atom number, phase space density, size etc. The total atom number, N can be obtained through the numerical sum over all pixels,

$$N = \frac{\Delta_x \Delta_z}{\sigma} \sum_{i,j} OD(x_i, z_j) \quad (2.4)$$

where Δ_x and Δ_z are the object sizes that correspond to one pixel. In our case, $OD(x, z)$ is further manipulated to obtain the atom number. The matrix, $OD(x, z)$ is summed over all row vectors and column vectors to obtain two one-dimensional vectors. They are denoted by $OD'_z(x)$ and $OD'_x(z)$ respectively. $OD'_z(x)$ and $OD'_x(z)$ are each fitted with a single Gaussian function to give us the one-dimensional density spatial distribution along x and z -direction. Fitted Gaussian functions for $OD'_z(x)$ and $OD'_x(z)$ are then used to obtain N_x and N_z respectively by using a similar formula as Eq. 2.4 but for a one-dimensional case. In our experiment, we choose either N_x or N_z depending on the fits. For our first absorption imaging setup, $\Delta_x = \Delta_z = 11 \mu m$. Our second absorption imaging setup, $\Delta_x = \Delta_z = 5.33 \mu m$. We process and analyse our images using MATLAB.

2.8.2 Noise filtering through image processing

The images taken using our absorption imaging system have noisy components (random grain noise and frequency noise). These are attributed to diffraction from the chip

Figure 2.13: Absorption imaging of our experiment with the imaging beam passing through an atomic cloud. This leaves a shadow in the light and this absorbing light is captured by the CCD camera as real image. A background image is taken 1 s later where the atomic cloud is absence.

surface, interference of the imaging beam with the optical elements, flying by of dust particles during imaging, and mechanical movement of the environment due to temperature and vibration. These several factors result in non-perfect cancellation of the real and background image in Eq. 2.1. In addition, when the atom number is low (~ 5000), the signal-to-noise ratio in our experiment is also low. Different image processing techniques like linear combination of background images [87], frequency-domain filtering and spatial filtering [88, 89] are utilized and combined to process our images.

Linear combination method

One noise reduction technique uses the algorithm described by Ockeloen, C. F. *et al* [87]. As an example, we have one background and one real image denoted as $I_{bkg\ 0}$ and $I_{real\ 0}$ to be processed. This algorithm requires several background images. The number of background images used are determined by the degree of improvement. In our case, 20 background images are sufficient. For best possible results, we only use those series of background images obtained at a time not too long (tens of minutes) from when the $I_{bkg\ 0}$ and $I_{real\ 0}$ are acquired.

The approach is to have a linear combination of the matrix, $A_{bkg\ l}$ of the non-interested regions (Usually the region near the atomic cloud is selected. Thus, $A_{bkg\ l}$ is the submatrix of $I_{bkg\ l}$) in these background images including $I_{bkg\ 0}$. For these regions, the corresponding matrix to $I_{real\ 0}$ is $B_{real\ 0}$. A coefficient, c_l is assigned to each of these background images. The linear combination is written as

$$A_{bkg} = \sum_{l=-10}^{10} c_l A_{bkg\ l} \quad (2.5)$$

All the coefficients are optimized using linear square fit method such that $|B_{real\ 0} - A_{bkg}|^2$ is minimized. These coefficients obtained are then used to generate the background image,

$I'_{bkg,0}$ written as

$$I'_{bkg,0} = \sum_{l=-10}^{10} c_l I_{bkg,l} \quad (2.6)$$

The final result is just

$$OD(x, z) = \ln \frac{I'_{bkg,0}(x, z)}{I_{real,0}(x, z)} \quad (2.7)$$

The effects of processing the images are displayed in Fig. 2.14. Fig. 2.14a shows an atomic cloud of 8000 atoms and a size of $30 \mu\text{m}$. In Fig. 2.14c, the atomic cloud has 8700 atoms and a size of $31 \mu\text{m}$. These pictures are processed with the described algorithm and the results are shown in Fig. 2.14b and Fig. 2.14d respectively. It is clear from the images, the fringes are successfully removed. With the increase of signal-to-noise ratio, the atomic clouds shapes are recovered.

Figure 2.14: Original optical depth images are processed using linear combination algorithm. They are transformed as a) $\rightarrow$ b) and c) $\rightarrow$ d). It is clearly seen the fringes are successfully removed using this algorithm.

Frequency domain filtering

The linear combination method as described in Sec. 2.8.2 is not so effective for big atomic cloud ($\sim \text{mm}$ size) as the region-of-interest is big. In addition, the use of a series of background images is also time consuming. One quick solution is to use Fourier transform to eliminate the period noises [89]. Since our images are 2D, the images are processed using 2D discrete Fourier transform (algorithm is fast Fourier transform (FFT)).

The original optical depth, $OD(x, z)$ image is shown in Fig. 2.15a. It is clear from this image there is some high frequency noise from the diffraction of the chip as well as from the environment. The 2D matrix $OD(x, z)$ is to be Fourier transformed to become $OD'(w_x, w_z)$. $OD'(w_x, w_z)$ is reorganized such that the low frequency component is at the centre and higher frequency components are positioned further away from the centre. The result is shown in Fig. 2.15b. Since the information of our image is mostly at the low frequency regime, a low-pass filter is applied. It is done by applying Fig. 2.15b a filtering disk of a certain radius. The radius is decided based on the frequency of the noise. In this case, the radius is chosen to be 95. With this filter disk, the matrix values outside the radius of this disk are to be eliminated as demonstrated in Fig. 2.15c. The final step

is to process Fig. 2.15c through inverse 2D Fourier transform. The result is shown in Fig. 2.15d. Our implementation of frequency filtering is based on the MATLAB m-file source code ³ from the book by Marques, O. [89].

Figure 2.15: a) Original optical depth image. b) Fourier transformed image. c) A filter disk is applied. d) Inverse Fourier transformed image. These images are processed based on the MATLAB m-file source code (Footnote 3) from the book by Marques, O. [89].

Spatial filtering

Spatial filtering is an image operation on a pixel value $I(y_i, z_i)$ of an image I such that its final pixel value is dependent on its initial neighbour pixel value at (y_i, z_i) [89]. For the simplest case of spatial filtering, we use average filtering. This technique is found to be useful in removing grain noise in our application. The number of neighbourhood pixels

³ www.ogemarques.com/view-tutorial/?tutorial=11_2

involved is decided based on situation. Averaging using more neighbourhood pixels may introduce blurring to the image while too few pixels may be ineffective in eliminating noise.

2.9 LabVIEW computer control system

There are two types of control cards installed in our computer system to control the experiment. They are analogue card (National Instruments - PCI-6713, 12 bits) and digital card (SpinCore PulseBlaster (PB)). We use two analogue cards with 8 channels each that can provide variable voltages from 0 to 10 V. The digital card has 24 digital channels that can provide TTL pulse (3.3 V) for switching purpose.

To interface the LabVIEW cards with the experimental hardware, two programmes developed in NIST, Gaithersburg are adopted and modified. One of these programmes is called main programme while the other is called sub-programme. The sub-programme is a graphical user interface (GUI) with all the experimental procedures programmed in sequential order. It gives instructions to the corresponding analogue and digital channels to output voltage at different times. However, this sub-programme will only be executed when it is pre-run and called by the main programme. As an example, a picture for the sub-programme is shown in Fig. 2.16 displaying a sequential loading of MOT → CMOT → molasses → magnetic trap. Its ramping stages are also shown in Fig. 2.17. The picture for the main programme is shown in Fig. 2.18.

Shown by Fig. 2.16 and 2.17, the sub-programme has three main sections, Master PB Staging List, Master AO Staging List and Master AO Ramp List. Master PB Staging List in Fig. 2.17 shows the sequential order of a single experimental run. The light green LEDs denotes TTL pulses sent while the dark green LEDs represent no TTL pulses sent in its corresponding channels by the digital card. The analogue outputs are managed by Master AO Staging List and Master AO Ramp List sections shown in Fig. 2.16 and 2.17 respectively. Master AO Staging List outputs constant voltage from the two analogue cards in its corresponding channels. The activation of n th row of Master AO Staging List corresponds to the n th row of light green LED in the AO trig column of the Master PB Staging List. In addition, instead of giving constant output voltages for the channels, Master AO Ramp List is used to ramp the analogue voltages. Fig. 2.17a shows the ramping of the current to the MOT coils from 1.40 - 3.40 V using our 1st analogue card and its 7th channel in the compressed MOT Ramp stage (ramp index 3 means 3rd row of Fig. 2.16) for a time duration of 2×10.0 ms⁴. Other channels are supplying either constant voltage (Cooling AOM VCO = 5.80 V; MOT supply analogue control = 3.00 V) or 0 voltage. For Fig. 2.17b, it is changing the frequency of the cooling beam by ramping the voltage from 1.60 - 1.00 V during molasses stage for a time duration of 2×2.0 ms.

2.10 Assembled experimental setup

To show the completed assembly of various components including vacuum parts, cryogenic system, laser, cameras and computer controls as discussed from Sec. 2.2 to 2.9, a picture

⁴Due to internal programming error, the actual time duration is actually 2 times than the displayed time

Chapter 2. Experimental setup

2.10. Assembled experimental setup

Figure 2.16: LabVIEW sub-programme showing the sequence to load atoms into the magnetic trap.

Figure 2.17: a) Ramping up the current to the MOT coils at compressed MOT Ramp stage (3rd row) shown in Fig. 2.16. b) Ramping the detuning of the cooling beam at molasses stage (6th row) shown in Fig. 2.16 (Footnote 2).

of the experimental system is shown in Fig. 2.19. A computer is used for the computer control using the LabVIEW cards (Sec. 2.9). Two computers are used for the image acquisition and analysis using the three cameras as discussed in Sec. 2.8. The ion pump, TSP and ionization gauge (Sec. 2.4) are put on the rack surface. Below this rack, there are MOT, transfer, bias and compensation coils (Sec. 2.7) surrounding the vacuum glass cell. Inside the vacuum glass cell is our superconducting atom chip (Sec. 2.2) mounted on the cold finger. Our atom chip is cooled through the use of liquid nitrogen supplied from a liquid gas cylinder (Taylor Wharton - XL 160).

2.11 Cooling and trapping procedures

We follow the standard procedures to produce the ^{87}Rb MOT by using three pairs of counter-propagating circular polarized laser beams and a pair of anti-Helmholtz coils [90]. This is illustrated in Fig. 2.20a. However, the atomic cloud obtained using standard MOT technique is of low density [91, 92]. To increase the atomic density, a technique called compressed MOT (CMOT) is used by suddenly increasing the magnetic field of the MOT in a time [93]. This results in an atomic cloud of smaller volume. However, the drawback is the temperature of the atomic cloud is increased. So, after MOT and CMOT procedures, we continue with molasses cooling to decrease the temperature to 10

Figure 2.18: LabVIEW main programme to execute the instructions of the sub-programme.

μK . Since molasses require a condition of zero magnetic field, a few compensation coils (not drawn in Fig. 2.20) are set up to provide constant magnetic field along x , y , and z -direction to cancel the earth magnetic field.

It is followed by optical pumping to pump the ultracold atoms from $|F = 2, m_F = -2, -1, 0 \text{ and } 1\rangle$ to $|F = 2, m_F = 2\rangle$ quantum state using both pumping beam and repumper. One can also pump it to $|F = 2, m_F = -1\rangle$ state by using optical pumping beam alone with the absence of repumper. With the atoms in either of these trappable quantum states, they are to be loaded to a magnetic trap. It is done by turning on the magnetic field from the MOT coils abruptly. In this case, the MOT coils function as a pair of anti-Helmholtz coils producing a quadrupole field that forms a magnetic trapping potential for the ultracold atoms. Since this ultracold atomic cloud is 35 mm away from the chip structure, we call this magnetic trap as lower magnetic trap. The atomic cloud at the lower magnetic trap is transferred closer to the chip structure by using transfer coils (elliptical shape). While the magnetic field from the lower magnetic trap is still on, the magnetic field from the transfer coils is ramped up adiabatically. Afterwards, the magnetic field for the lower magnetic trap is ramped down adiabatically while maintaining the magnetic field from the transfer coils. Finally, with this setting, the atomic cloud is transferred closer (≈ 2 mm distance) to the chip structure. We call the atomic cloud trapped using transfer coils alone as upper magnetic trap. Atomic cloud can be transferred closer to the Z-wires or square structure by using 3 pairs of bias coils to introduce homogeneous bias fields to the existing quadrupole field. The schematic diagram for all the coils used in the experiment are shown by the drawing in Fig. 2.20b. Afterwards, all the magnetic fields can be turned off for the atomic cloud to experience a time-of-flight (TOF). After this TOF, the atomic cloud is to be imaged by the CCD

Figure 2.19: Picture of the superconducting atom chip experimental setup. Labels in yellow or blue show the components.

camera. However, the magnetic fields can be left on for in-situ imaging.

2.11.1 Temperature measurement

Measurement of the temperature, T of our atomic cloud is done through time-of-flight (TOF) measurement. The expansion of the size, $r(t)$ of the atomic cloud has a relation with TOF, t as shown by the following equation,

$$r^2(t) = r^2(0) + \frac{2kT}{m}t^2 \quad (2.8)$$

where k is the Boltzmann constant and m is the mass of the atom [94]. Shown in Fig. 2.21a and 2.21b are the plots of the radius square of the cloud size v.s. the square of the TOF for molasses and lower magnetic trap. Eq. 2.8 shows that the curve is linear and the gradient is related to its temperature by a fixed factor $\frac{2k}{m}$. The fit to the plot shows the temperature of our molasses to be $9.6 \pm 0.3 \mu\text{K}$. The temperature of our lower magnetic trap is $239 \pm 7 \mu\text{K}$. In order to make the temperature measured to be reliable, the magnetic fields have to be turned off completely or else the atomic cloud will experience a magnetic force. However, this is not possible with our superconducting vortex trap where the vortex current can't be switched off. Alternative temperature measurement technique called RF spectroscopy can be used [6]. Details of this measurement technique will be discussed in Chap. 4.2.2.

2.11.2 Lifetime measurement

Lifetime of the atomic cloud in a trapping potential can be determined by measuring the atom number v.s. the trapping time. A plot of the atom number v.s. the trapping time in our lower magnetic trap is shown in Fig. 2.22. An exponential fit to the curve shows the lifetime is 29 ± 3 s. In most of the cases, the trapping time is either limited by the background gas collisions at low density, or by three body recombination at high density [80]. Since our atomic cloud is at a low density regime, the lifetime is mainly determined by the vacuum pressure. With this lifetime, we estimate our vacuum pressure to be 4×10^{-10} mbar ⁵.

⁵Assuming a 1 mK trap depth, collision of ^{87}Rb with room temperature He background gases, and using the loss rate equation shown by Arpornthip, T. *et al* [95].

a)

b)

Figure 2.20: a) Drawing of our standard MOT setup with three pairs of counter-propagating 780 nm laser beams and a pair of anti-Helmholtz coils. For clarity purpose, bias coils and transfer coils are not drawn in this picture. Arrows are drawn to point to one side of the counter-propagating beams. 35 mm is written to indicate the distance between this atomic cloud and the atomic chip. b) A pair of MOT coils (copper colour) and transfer coils (vermilion colour, elliptical shape) which are used as magnetic trap for the ultracold atoms. Three pairs of bias coils producing homogeneous bias fields along x , y and z -direction (light blue colour) are added to the quadrupole field from the transfer coils to transfer atoms closer to the interested region on the chip structure. Due to space constraints, arrows are only drawn and pointed to one side of the bias coils.

Figure 2.21: Plot (blue) of the radius square of the atomic cloud v.s. TOF square for our a) molasses and b) lower magnetic trap. The linear fits (red) to the two curves give a gradient which corresponds to the temperature of $9.6 \pm 0.3 \mu\text{K}$ and $239 \pm 7 \mu\text{K}$ respectively.

Figure 2.22: Plot of the atom number in the lower magnetic trap v.s. trapping time (blue) with a fitted exponential curve (red). The fitted lifetime is $29 \pm 3 \text{ s}$.

Part I

Chapter 3

Theory of superconductor

3.1 Introduction

We begin this chapter with Sec. 3.2 discussing the basic ideas of superconductors: from Meissner effect, London equation, Pippard model to Ginzburg-Landau theory. In later sections, we focus on type-II superconductors. We elaborate a mesoscopic model (critical state model) describing the behaviour of vortices as supercurrents for an applied external magnetic field pulse B_{ext} bigger than the first critical field B_{c1} in Sec. 3.3. As discussed in Sec. 3.4, other than magnetic field pulse, vortices can also be induced using current pulses from an external source. We simulate the sheet current density J for the supercurrents inside various superconducting structures (strip, disk and ring) and obtain its magnetic field distribution based on Biot-Savart law. These magnetic field distributions are manipulated and programmed in such a way that various useful magnetic traps can be realized.

3.2 Superconductors

Superconductivity was first discovered by Heike Kamerlingh Onnes in the year 1911 where the electrical resistance of mercury became zero after it was cooled down to a critical temperature T_c of 4.2 K by using liquid helium [96, 97]. Later, most metals and alloys were found to display superconductivity at different critical temperature. In 1933, Meissner, W. and Ochsenfeld, R. discovered the property of Meissner effect in superconductor [98]. It is a phenomenon where the superconductor becomes a perfect diamagnet, repelling all the magnetic flux that are applied to it.

London equation

The Meissner effect of superconductor was described phenomenologically by the brothers Fritz and Heinz London based on the two-fluid model used for superfluid ^4He [99]. There are two equations called first London equation and second London equation [100] written

as

$$\mathbf{E} = \frac{\partial}{\partial t}(\Lambda \mathbf{J}_s) \quad (3.1)$$

$$\mathbf{B} = -\nabla \times (\Lambda \mathbf{J}_s) \quad (3.2)$$

respectively where Λ is the London parameter

$$\Lambda = \frac{m'}{n'q'^2} \quad (3.3)$$

with m' , n' and q' denote the mass, density and charge of the carriers. It is to be proven by BCS theory that the electrical conductivity carriers are Cooper pairs (two electrons bound together), thus $m' = 2m_e$, $q' = 2e$ and $n' = n_s/2$. m_e , e and n_s refer to the mass of the electron, elementary charge and the density of the superconducting electrons.

The first London equation relates electric field $\mathbf{E}$ to superconducting current density $\mathbf{J}_s$, a similar analogy as Ohm's law but with the absence of resistance. The second London equation relates magnetic field $\mathbf{B}$ and $\mathbf{J}_s$, showing the superconductor to exhibit Meissner effect. The negative sign also indicates that it has a diamagnetic behaviour.

Assume a semi-infinite superconductor which occupies the space of $x > 0$. For an applied magnetic field, analysis of the second London equation reveals that the $\mathbf{B}$ is decaying exponentially over a distance, written as

$$\mathbf{B}(x) = \mathbf{B}(0) \exp\left(-\frac{x}{\lambda_L}\right) \quad (3.4)$$

with λ_L denotes the penetration depth. It is further expressed as

$$\lambda_L = \sqrt{\frac{\Lambda}{\mu_0}} = \sqrt{\frac{m_e}{\mu_0 n_s e^2}} \quad (3.5)$$

The penetration depth signifies the region of space away from the superconducting surface where the screening current can reside. Since λ_L depends on n_s , λ_L is temperature dependent. λ_L is found to have the empirical relationship with temperature T [101] as

$$\lambda_L(T) = \frac{\lambda_L(0)}{\sqrt{1 - \left(\frac{T}{T_c}\right)^4}} \quad (3.6)$$

By equating Eq. 3.5 and Eq. 3.6, $n_s(T)$ is derived and expressed as

$$n_s(T) = n_s(0) \left[1 - \left(\frac{T}{T_c}\right)^4\right] \quad (3.7)$$

Pippard non-local model

However, London theory is a local theory. In other words, $\mathbf{J}_s$ in Eq. 3.2 only depends on $\mathbf{A}$ (related to $\mathbf{B}$ as $\mathbf{B} = \nabla \times \mathbf{A}$) at the same spatial position. It was found the

penetration depth λ measured in experiment differs from London penetration depth λ_L and it is also dependent on impurity contents. To explain these phenomena, Pippard introduced another characteristic length called coherence length by imposing non-local condition of $\mathbf{J}_s$ on $\mathbf{A}$ in London equation. The intrinsic coherence length ξ_0 is derived using Heisenberg uncertainty principle and is expressed as

$$\xi_0 \approx \frac{\hbar v_F}{\pi \Delta(0)} \approx \alpha \frac{\hbar v_F}{k_B T_c} \quad (3.8)$$

$\Delta(0)$ is the energy of the superconducting gap forms at the Fermi surface at zero temperature. α has a value of 0.18 (from BCS model) and v_F is the Fermi velocity [102]. To account for the effects of defects, the mean free path l term is introduced and added to the ξ_0 to obtain the effective coherence length ξ written as following

$$\frac{1}{\xi} = \frac{1}{\xi_0} + \frac{1}{l} \quad (3.9)$$

The purity of a superconductor is characterized by comparing ξ_0 and l . The superconductor is said to be in the clean limit if $\xi_0 \ll l$ and dirty limit if $\xi_0 \gg l$. ξ has a physical meaning that the spatial variation of superconducting electron density n_s can only occur over a finite distance but not any arbitrary small distance. In the two extreme cases of $\xi_0 \gg \lambda_L$ and $\xi_0 \ll \lambda_L$, the penetration depth λ is expressed as

$$\lambda = \begin{cases} \sqrt[3]{0.62 \lambda_L^2 \xi_0} & \text{for } \xi_0 \gg \lambda_L, \\ \lambda_L \sqrt{\xi_0 / \xi} & \text{for } \xi_0 \ll \lambda_L. \end{cases} \quad (3.10)$$

$$(3.11)$$

Ginzburg-Landau theory

In 1950, Ginzburg and Landau described the superconducting state through an order parameter $\psi(\mathbf{r})$ [103, 104]. The supercurrent density is $n_s(\mathbf{r}) = |\psi(\mathbf{r})|^2$ and vanishes as temperature of the superconductor approaches T_c . In this formalism, the coherence length ξ is expressed as

$$\xi(T) = \sqrt{\frac{\hbar^2}{2m'|\alpha(T)|}} \quad (3.12)$$

where α is the temperature dependent term of the expansion of free energy. It is similar to the coherence length introduced by Pippard except it is temperature dependent. The second parameter is the penetration depth which is similar as London's penetration depth but with n_s replaced with $|\psi_0|^2$. It is written as

$$\lambda(T) = \sqrt{\frac{m'}{\mu_0 q^2 |\psi_0|^2}} \quad (3.13)$$

One important classification of the type of superconductor is based on the ratio of

$\xi(T)$ and $\lambda(T)$. The ratio is called Ginzburg Landau parameter and is defined as

$$\kappa = \frac{\lambda(T)}{\xi(T)} \quad (3.14)$$

It is approximately constant and is independent of temperature. Ginzburg-Landau theory characterizes a superconductor that has the ratio of $\kappa < 1/\sqrt{2}$ as type-I superconductor and $\kappa > 1/\sqrt{2}$ as type-II superconductor.

For type-I superconductor, when the temperature of the superconductor is varied from above T_c to below T_c , the state transits from normal conducting to superconducting. For type-II superconductor, at a temperature below T_c and for an applied magnetic field B_{ext} lower than B_{c1} (lower critical field), it is a superconductor that displays the same Meissner behaviour as type-I superconductor. Based on Ginzburg-Landau formalism, B_{c1} is written as

$$B_{c1} \approx \frac{\phi_0}{4\pi\lambda(T)^2} \ln \kappa \quad (3.15)$$

However, when the B_{ext} is raised above B_{c1} but falls below B_{c2} (upper critical field), it transits from the Meissner state to the mixed state/vortex state. At the mixed state, there are fields penetration into the superconductor in the form of vortices (Abrikosov vortices) where each single vortex has a unit of single flux quantum $\phi_0 = h/(2e)$. Thus, the mixed state is a superconducting state which has a mixture of a superconducting (vortex free region) region and a normal conducting region (vortex region). If the superconductor has a few or no defects, the Abrikosov vortices will enter from the edges of superconductor. Due to interaction among vortices, they will arrange themselves in a triangular lattice. When defects are present, the vortices will be pinned to the defects and remain there because it is energetically more stable at these sites [105]. A further increase of B_{ext} will result in decreasing distance between the vortices.

If the applied magnetic field is above B_{c2} , the superconductor becomes normal conducting. This occurs when the distance between vortex is equal to the vortex core size ξ , thus deriving the formula for B_{c2} as

$$B_{c2} = \frac{\phi_0}{2\pi\xi(T)^2} \quad (3.16)$$

3.2.1 Mesoscopic model for vortices

As compared to type-I superconductor where the shielding current only flows in the region of $\lambda(T)$ away from the surface, a type-II superconductor in the mixed state can have shielding current flows larger than $\lambda(T)$. For temperature close to T_c , the penetration by B_{ext} is more uniform. In addition, at this temperature, dendritic avalanches for the flux penetration are also avoided. These conditions allow the use of a mesoscopic model called critical state model to describe the collective behaviour of vortices as supercurrent density modelled by Bean, C. P. [106] and Brandt, E. H. [107]. The model developed by Bean, C. P. [106] assumes the field penetrated region is at its critical current value. It is less accurate compared to the model by Brandt, E. H. [107] where it considers a spatial variation of the current density. We will only use the model described by Bean, C. P.

Figure 3.1: This figure shows the physical parameters of the superconductor. d is the thickness and $2w$ is the width of the strip. B_{ext} is the magnetic field applied perpendicularly to the strip for the creation of remanent state. Picture adapted from the paper by Zhang, B., *et al.* [43].

[106] to account for the qualitative behaviour of the supercurrents of our superconductor. For numerical simulations, we will use the model developed by Brandt, E. H. [107].

Usually, the type-II superconductor can have some defects which serve as pinning centres for the vortices. Thus, there is a competition between the pinning and mutual repulsion due to Lorentz force among the vortices. In this case thermal fluctuation plays an important role in providing energy for the depinning to occur. An equilibrium is established during the application of B_{ext} for the pinning process to the pinning centres and mutual repulsion between vortices. The equilibrium is likened to a glass phase without long-range order [43]. After B_{ext} is removed, a new equilibrium phase is established for the remaining vortices. These remaining vortices produce a magnetization and this equilibrium state is called remanent state.

The vortices equilibrium phase can be modified further applying a transport current. This is due to interaction between the Lorentz force of the transport current with the vortices, thus redistributing the vortex equilibrium position [108].

3.3 Critical state model (magnetic field pulse)

In our model, we restrict the applied external magnetic field B_{ext} to $B_{c1} < B_{ext} < B_{c2}$. The magnetic field direction is perpendicular to the surface of the type-II superconducting thin strip and it is assumed the superconductor is in its virgin state (not subject to any magnetic fields). According to Bean's critical state model, the region where the flux penetrates will have a current density equal to the critical current density j_c . Since the thickness of the superconductor is much smaller than the width of the superconducting wire (the YBCO Z-wire has thickness $d = 800$ nm, width $2w = 400$ μ m as shown in Fig. 2.1c), we define the sheet current density as $J(x) = j(x)d$ where $j(x)$ is the local current

density. The sheet current density $J(x, B_{ext}, J_c)$ is given by [107, 43]

$$J(x, B_{ext}, J_c) = \begin{cases} \frac{2J_c}{\pi} \tan^{-1} \left[\frac{x}{w} \sqrt{\frac{\omega^2 - b^2}{b^2 - x^2}} \right] & \text{if } |x| \leq b; \\ J_c \frac{x}{|x|} & \text{if } b \leq |x| \leq w; \end{cases} \quad (3.17)$$

b is the flux free region and is expressed as

$$b = \frac{\omega}{\cosh \frac{B_{ext}}{B_c}} \quad (3.18)$$

where B_c is the thermodynamic critical field that can be written as

$$B_c = \frac{\mu_0 J_c}{\pi} \quad (3.19)$$

J_c the critical sheet current density which can be obtained from the multiplication of j_c with d as shown as

$$J_c = j_c d \quad (3.20)$$

A schematic diagram to illustrate this model is shown in Fig. 3.1. In this figure, a superconducting strip along y -direction is drawn. Its thickness is labelled as d . The centre of this strip is at $x = 0$ and its width spans from $x = -w$ to $x = w$. The surface is at $z = 0$. An external magnetic field B_{ext} is applied perpendicularly to the z -direction creating the supercurrent. The inset shows the time sequence of this external magnetic field pulse.

Physically, $w - b$ denotes the vortex penetrated region after applying the magnetic field B_{ext} and b is the vortices free region. A maximum magnetic field B_p exists such that $b = 0$. B_p can be calculated easily by equating Eq. 3.18 with $b = 0$. B_p can be derived as

$$B_p = \frac{\mu_0 j_c d}{\pi} \ln \frac{2w}{d} \quad (3.21)$$

3.3.1 Current distribution of supercurrents

For an applied magnetic field B_{ext} pulse, the rising edge of the magnetic field will introduce vortices while the decreasing edge of the magnetic field will introduce anti-vortices entering at the strip edge. So, the total current density is [107, 43]

$$J_{total} = J(x, B_{ext}, J_c) - J(x, B_{ext}, 2J_c) \quad (3.22)$$

where the first term accounts the vortices due to the rising magnetic field while the second term accounts for the anti-vortices from the lowering of the magnetic field. It is also noticed that in this model J_{total} does not depend on the time duration between the rising and lowering part of the magnetic field. The pattern of the rising and lowering parts is also irrelevant in affecting the distribution of J_{total} .

In our simulations, we set the parameters $j_c = 2.08 \times 10^6$ A/cm², $2w = 400$ μ m and $d = 1$ μ m. Unless stated otherwise, these parameters are used for the simulation of the magnetic field distribution shown in Secs. 3.3.2 and 3.3.3. With these parameters,

$B_p = 499$ G and the thermodynamic critical field $B_c = 83.3$ G. The supercurrents in the central region are usually small and most of the supercurrents flow in the region $0.4w < x < w$ ($80 \mu\text{m} < x < 200 \mu\text{m}$) and $-0.4w > x > -w$ ($-80 \mu\text{m} > x > -200 \mu\text{m}$). Integration over half of the strip yields the magnitude of current to be $\pm 0.37j_c dw$. For an example of $B_{ext} = 2.4B_c = 200$ G, the supercurrent is calculated to be 1.55 A. Using Eq. 3.18, this supercurrent is flowing at the region of $36 \rightarrow 200 \mu\text{m}$ while $0 \rightarrow 36 \mu\text{m}$ region is free from vortices.

To illustrate the sheet current density distribution, a graph is plotted in Fig. 3.2a using the critical state model discussed so far for the magnetic field pulse of $B_{ext} = 0 \rightarrow 200 \text{ G} \rightarrow 0$. The rising part of the pulse gives a sheet current density shown by the red dashed line while the lowering part of the pulse is shown by the blue dashed line. The sum of the two sheet current densities gives rise to the total sheet current density shown by black solid line. If B_{ext} is pointed towards negative direction, the sheet current density distribution is just inverted at $x = 0$.

From the results shown in the Fig. 3.2a, the contribution of the currents from the region smaller than b are small and thus negligible. The currents mostly concentrate on the region of $w - b$ and they are of higher magnitude. With this plot, one intuitive useful concept can be derived from the interpretation of the graph. A magnetic field pulse B_{ext} of the range $B_{c1} < B_{ext} < B_{c2}$ will create a pair of supercurrents of the same magnitude but flowing in the opposite direction at the two edges of the superconductor strip. One can imagine this pair of supercurrents as two current carrying virtual wires inside a physical wire. Intuitively, one can visualize the size of these virtual wires as $w - b$. The magnitude and direction of the currents of these virtual wires can be controlled by the magnitude and direction of the B_{ext} .

3.3.2 Magnetic field distribution from supercurrents

With the $J(x, B_{ext}, J_c)$ calculated for a B_{ext} using the critical state model, one can continue to simulate the magnetic field distribution from these supercurrents using Biot-Savart law.

With a single magnetic field pulse of $B_{ext} = 0 \rightarrow 2.4B_c \rightarrow 0$ ($B_{ext} = 0 \rightarrow 200 \text{ G} \rightarrow 0$), we plot the magnetic field component in the z -direction B_z (blue solid line) and x -direction B_x (blue solid line) v.s. x at $z = w$ produced by these supercurrents as shown in Fig. 3.2b and 3.2c. These plots are then compared to the magnetic fields (rec circles) produced by two counter-propagating current carrying wires positioned at $x = \pm w$ and transport currents of $\pm 0.37j_c dw$. The patterns for the magnetic field distribution of B_z and B_x are found to be the same for the superconducting strip and the two wires. This verifies the concept discussed earlier that a magnetic field pulse applied to a type-II superconducting strip within $B_{c1} < B_{ext} < B_{c2}$ is analogous to two counter-propagating current carrying wires.

In Fig. 3.2d we show B_z with respect to z at $x = 0$. B_z rises to a maximum value before it decreases to 0 closer to the chip surface. In contrast to the conducting wires, B_z saturates to a constant value. The difference is explained by the Meissner effect where the magnetic field is zero inside the superconductor.

Figure 3.2: Simulation for external magnetic field pulse of $B_{ext} = 0 \rightarrow 2.4B_c \rightarrow 0$ a) Sheet current density distribution b) B_x v.s. x at $z = w$ from supercurrents (blue solid line) and two wires (red circles). c) B_z v.s. x at $z = w$ from supercurrents (blue solid line) and two wires (red circles). d) B_x v.s. z at $x = 0$ from supercurrents (blue solid line) and two wires (red circles). e) Single trap is formed at the edge with $B_{bias}^x = -0.173B_c$ f) Single trap is formed above the centre with $B_{bias}^z = -0.2B_c$. Graphs adapted from the paper by Zhang, B., *et al.* [43].

3.3.3 Programmable magnetic traps using supercurrents

To form a magnetic trap for the ultracold atoms, the magnetic potential must have a field minimum. For the magnetic fields generated by the supercurrents, one can apply an external magnetic field B_{bias} to add or cancel the existing field to generate a field minimum. However, in some special cases, a field minimum can be achieved without B_{bias} . This can be done with a tailored sequence of external magnetic field pulses. It is noted that our model is based on an infinite long strip along y -direction, so there is no confinement for the trapped atoms along this direction. However, in experiments, one can design a Z-shape or U-shape structure for confinement in this direction. The following subsections will discuss and show the simulation results of different trapping geometries for the magnetic traps by varying the magnetic field pulses in magnitude, direction and sequential order.

3.3.4 Single trap

For a single magnetic field pulse B_{ext} , we simulate 2 trapping geometries. In the following, we discuss the procedure to obtain a magnetic with a single pulse of $B_{ext} = 0 \rightarrow 2.4B_c \rightarrow 0$ ($B_{ext} = 0 \rightarrow 200 \text{ G} \rightarrow 0$). The current sheet distribution and its magnetic fields were discussed in details earlier in Sec. 3.3.1 and 3.3.2.

With B_{bias}^x - above the edge of the strip

By applying a homogeneous bias field B_{bias}^x along x -direction, a magnetic trap above the edge of the strip can be obtained. Depending on whether B_{bias}^x is applied to positive or negative x -direction, the trap can be formed at either side of the two edges.

In Fig. 3.2e, the contour plot of the magnetic field (absolute value of magnetic field) is simulated with a $B_{bias}^x = -0.173B_c = -14.4 \text{ G}$. The distance of this trap from the surface is $0.8w = 160 \mu\text{m}$. One can vary the trap to surface distance by varying the strength of the B_{bias}^x .

With B_{bias}^z - above the centre of the strip

From Fig. 3.2d, it is obvious by applying a homogeneous bias field B_{bias}^z in negative z -direction, a field minimum can be created. One can vary the position of this field minimum by varying the strength of this B_{bias}^z . For $B_{bias}^z = -0.2B_c = -16.6 \text{ G}$, the trap position is at $z = w = 200 \mu\text{m}$. The contour plot of this magnetic potential is shown in Fig. 3.2f.

3.3.5 Self-sustaining trap

A possible novel configuration of magnetic trap results from using type-II superconductor is self-sustaining trap. It is a type of trap which its magnetic potential is produced only by the supercurrents and does not require any external magnetic field sources to create the field minimum in the trapping potential. In the following, we will discuss the procedure

to create these self-sustaining traps by programming the supercurrents with external magnetic field pulses.

Single trap - above the centre of the strip

By applying two magnetic field pulses sequence $B_{ext} = 0 \rightarrow 2.4B_c \rightarrow -1.2B_c \rightarrow 0$ ($B_{ext} = 0 \rightarrow 200 \text{ G} \rightarrow -100 \text{ G} \rightarrow 0$) to a virgin state, its sheet current density distribution is shown in Fig. 3.3a. The contour plot of its magnetic field distribution is shown in Fig. 3.3b. It can be seen from this plot there exists a self-sustaining trap above the centre of the strip. It is of $0.6w = 120 \mu\text{m}$ distance away from the surface.

Double traps - above the edges of the strip

By applying three magnetic field pulses sequence $B_{ext} = 0 \rightarrow 2.4B_c \rightarrow -1.2B_c \rightarrow 0.7B_c \rightarrow 0$ ($B_{ext} = 0 \rightarrow 200 \text{ G} \rightarrow -100 \text{ G} \rightarrow 58.3 \text{ G} \rightarrow 0$) to a virgin state, its sheet current density distribution is shown in Fig. 3.3c. The contour plot of its magnetic field distribution is shown in Fig. 3.3d. It can be seen from this plot there exists two self-sustaining traps above the opposite edges of the strip. They are situated $0.25w = 50 \mu\text{m}$ distance away from the surface.

3.4 Critical state model (current pulse)

Other than a magnetic field pulse, a transport current I_a will also be able to induce vortices in a type-II superconductor. The sheet current distribution $J(x, I_a, J_c)$ depends on the magnitude of I_a and is expressed as follows [107, 43],

$$J(x, I_a, J_c) = \begin{cases} \frac{2J_c}{\pi} \tan^{-1} \sqrt{\frac{\omega^2 - b^2}{b^2 - x^2}} & \text{if } |x| \leq b; \\ J_c & \text{if } b \leq |x| \leq w; \end{cases} \quad (3.23)$$

b is the flux free region. b is shown as

$$b = w \sqrt{1 - \frac{I_a^2}{I_p^2}} \quad (3.24)$$

I_p is the maximum transport current and given by

$$I_p = 2wJ_c \quad (3.25)$$

Using the same j_c , w and d parameters as discussed earlier, we obtain $I_p = 8.32 \text{ A}$.

Figure 3.3: Two external magnetic field pulses of $B_{ext} = 0 \rightarrow 2.4B_c \rightarrow -1.2B_c \rightarrow 0$ are applied to the superconducting strip. a) Sheet current density distribution. b) A self-sustaining trap is formed above the centre of the strip. c) Sheet current density distribution for $B_{ext} = 0 \rightarrow 2.4B_c \rightarrow -1.2B_c \rightarrow 0.7B_c \rightarrow 0$. d) Two self-sustaining traps formed above the edges with $0.25w$ distance away. Graphs adapted from the paper by Zhang, B., *et al.* [43].

3.4.1 Current distribution of supercurrents

For a current I_a applied to a superconducting strip and with a decrease to I_f that falls between $-I_p < I_f < I_p$, the total sheet current density [107, 43] is written as

$$J(x, I_f, J_c) = J(x, I_a, J_c) - J(x, I_a - I_f, 2J_c) \quad (3.26)$$

The rising part of the I_a is represented by $J(x, I_a, J_c)$ while the decreasing part of I_a to I_f is given by $-J(x, I_a - I_f, 2J_c)$. Similar as Eq. 3.22, both the first term and second term refer to the current from vortices and anti-vortices entering from the edges.

We model a special case for an applied current pulse of magnitude $I_a = 0 \rightarrow 0.5I_p \rightarrow 0$ ($I_a = 0 \rightarrow 4.16 \text{ A} \rightarrow 0$). After the application of this pulse, the vortices pattern is simulated with its sheet current density distribution shown in Fig. 3.4a. The blue dashed line is the sheet current density of $I_a = 0 \rightarrow 0.5I_p$. The red dashed line is for $I_a = 0.5I_p \rightarrow 0$. From the addition of these two curves, the total sheet current density is given by the red solid line. The currents in the region $w > x > 0.935w$ ($200 \mu\text{m} > x > 187 \mu\text{m}$) and $-200 \mu\text{m} < x < -187 \mu\text{m}$ are of the same magnitude and flow in negative direction. The currents in the inner region $0 < x < 0.935w$ ($0 < x < 187 \mu\text{m}$) and $0 > x > -0.935w$ ($0 > x > -187 \mu\text{m}$) flow in the positive direction. This current distribution resembles a four-wire configuration where the inner wires and outer wires have currents flowing in opposite direction but having the same magnitude.

3.4.2 Magnetic field distribution from supercurrents

With the $J(x, I_f, J_c)$ calculated for a I_a using the critical state model, like before, one can simulate the magnetic field distribution from these supercurrents using Biot-Savart law.

We plot the magnetic field component in the x -direction B_x (blue solid line) and z -direction B_z (blue solid line) v.s. x at $z = w$ produced by these supercurrents in Fig. 3.4b and 3.4c. These plots are then compared to the magnetic fields produced by the four current carrying wires (red circles) positioned at $x = \pm 0.737w$ and $x = \pm w$. These inner two wires conduct currents of $0.04j_c dw$ while the outer two wires have opposite currents $-0.04j_c dw$. The patterns for the magnetic field distribution of B_z and B_x are found to agree well with the superconducting strip. This shows that a current pulse applied to a type-II superconductor is analogous to a four wire configuration with the inner two wires having opposite current to the outer two wires.

We also plot B_x v.s. z at $x = 0$. There is a deviation between the B_x produced by supercurrents and by the four wires. This is due to Meissner effect of superconductor at near distance.

3.4.3 Programmable magnetic traps using supercurrents

By applying a homogeneous bias field to the existing magnetic fields produced by the supercurrents, magnetic traps can be created in a similar procedure as discussed in Sec. 3.3.3. We discuss the magnetic trap from the magnetic field produced from a current pulse of $I_a = 0 \rightarrow 0.5I_p \rightarrow 0$.

Figure 3.4: Simulation for current pulse of $I_a = 0 \rightarrow 0.5I_p \rightarrow 0$ a) Sheet current density distribution b) B_x v.s. x at $z = w$ from supercurrents (blue solid line) and four wires (red circles). c) B_z v.s. x at $z = w$ from supercurrents (blue solid line) and four wires (red circles). d) B_x v.s. z at $x = 0$ from supercurrents (blue solid line) and four wires (red circles). e) Single trap is formed with $B_{bias}^x = 0.006B_c = -0.5$ G f) Double traps formed with $B_{bias}^x = 0.0016B_c = 0.13$ G. Graphs adapted from the paper by Zhang, B., *et al.* [43].

Figure 3.5: Schematic diagram illustrating the B_{ext} applied to the type-II superconducting strip, disk and ring structures. Pictures adapted from the paper by Zhang, B., *et al.* [44].

Single trap - above the centre of the strip

By applying a homogeneous bias field in negative x -direction, a single trap above the centre of the strip can be formed. We choose $B_{bias}^x = 0.006B_c = -0.5$ G and shows its contour plot of the magnetic field distribution in Fig. 3.4e. The trap is formed at the position of $z = w = 200$ μm .

Double traps - above the edges of the strip

By applying a homogeneous bias field in positive x -direction, two traps above the edges of the strip can be formed. We choose $B_{bias}^x = 0.0016B_c = 0.13$ G and show its contour plot of the magnetic field distribution in Fig. 3.4f. The traps are formed at position of $z = w = 200$ μm . The lateral positions for these two traps are $x = \pm 400$ μm .

3.5 Numerical simulation of sheet current density (strip, disk and ring)

Eq. 3.17 is an analytical solution of the sheet current density for a field penetrating a type-II superconducting strip [107]. This equation shows the penetrated region has its sheet current density at its critical value. In this section, we employ a numerical approach to calculate the sheet current density according to Brandt, E. H. [109, 110]. In Sec. 3.5.1, we numerically model the current distribution and compare it to the analytical solution in Eq. 3.17 for an infinite thin superconducting strip. This comparison allows us to validate the numerical method as it should not deviate very much from the analytical solution. The numerical method is then applied to a superconducting disk and ring. The relevant size parameters for the strip (left), disk (centre) and ring (right) structures are labelled accordingly in the drawing shown in Fig. 3.5.

For an applied external magnetic field B_{ext} , the sheet current density for the strip is

$$J(x, t) = \tau \left[2\pi x \frac{dB_{ext}}{dt} + \int_0^a \frac{\partial J(u, t)}{\partial t} M(x, u) du \right] \quad (3.27)$$

For the disk and ring, its sheet current density is

$$J(r, t) = \tau \left[\pi r \frac{dB_{ext}}{dt} + \int_{r_0}^a \frac{\partial J(u, t)}{\partial t} Q(r, u) du \right] \quad (3.28)$$

where

$$\tau = \frac{\mu_0 a d}{2\pi \rho(J)} \quad (3.29)$$

$$\rho(J) = \rho_c \left(\frac{J}{J_c} \right)^{n-1} \quad (3.30)$$

$$n = \frac{U_0}{k_b T} \quad (3.31)$$

$$M(x, u) = \ln \left| \frac{x - u}{x + u} \right| \quad (3.32)$$

$$Q(r, u) = -\frac{u}{r} \int_{r_0/u}^{r/u} \left[\frac{E\left(\frac{\sqrt{4\omega}}{1+\omega}\right)}{1-\omega} + \frac{K\left(\frac{\sqrt{4\omega}}{1+\omega}\right)}{1+\omega} \right] \omega d\omega \quad (3.33)$$

ρ is the nonlinear resistivity with its critical value as ρ_c . U_0 is the characteristic activation energy of the superconductor [111]. $E\left(\frac{\sqrt{4\omega}}{1+\omega}\right)$ and $K\left(\frac{\sqrt{4\omega}}{1+\omega}\right)$ are the first and second kind of elliptical integrals.

We choose $n = 19$ for our simulation. Even though n depends on the choice of materials, this does not change the qualitative behaviour of the sheet current density distribution. Eq. 3.27 and 3.28 are to be integrated numerically by Riemann sum.

3.5.1 Strip structure

Fig. 3.5 (left) illustrates our simulated strip structure. The strip has a size of $2a$ and thickness d and lies on the xy -plane. B_{ext} is applied along the z -direction to induce the supercurrents.

Comparing analytical and numerical method of sheet current density

For $B_{ext} = 0 \rightarrow \mu_0 J_c \rightarrow 0$, Eq. 3.27 is solved numerically and its sheet current density (blue solid line) is shown in Fig. 3.6. The general features do not differ from the analytical result (red dashed line) using Eq. 3.17. Eq. 3.17 dictates that the field penetrated region has a sheet current density of J_c but this is not the case for Eq. 3.27.

3.5.2 Disk structure

Fig. 3.5 (centre) shows our simulated disk structure has radius a and thickness d . The disk structure lies parallel to the xy -plane. B_{ext} is to be applied along the z -direction to induce the supercurrents in the superconductor.

Figure 3.6: The sheet current density solved numerically (blue solid line) and analytically (red dashed line) for $B_{ext} = 0 \rightarrow \mu_0 J_c \rightarrow 0$. Graph adapted from the paper by Zhang, B., *et al.* [44].

a)

b)

Figure 3.7: $B_{ext} = 0 \rightarrow 1.2\mu_0 J_c \rightarrow 0$ applied to a disk structure. a) Plot of B_z v.s. z at $r = 0$. b) Single trap is formed at $z_0 = 1.2a$ for $B_{bias}^z = -0.05\mu_0 J_c$. [Inset] Sheet current distribution for the disk along the radial direction. Graph adapted from the paper by Zhang, B., *et al.* [44].

Single trap

To simulate the sheet current density for a superconducting disk, we use $B_{ext} = 0 \rightarrow 1.2\mu_0 J_c \rightarrow 0$. Using Biot-Savart law, we plot its B_z v.s. z at $r = 0$ in Fig. 3.7a.

By applying a homogeneous bias field $B_{bias}^z = -0.05\mu_0 J_c$, a single trap is formed above the centre of the disk at $z_0 = 1.2a$. The contour plot of this trap is shown in Fig. 3.7b. The inset shows the sheet current distribution for the disk along the radial direction.

Self-sustaining trap

In addition, it is also possible to create a self-sustaining trap by loading two external magnetic field pulses. We simulate this magnetic trap by applying $B_{ext} = 0 \rightarrow 0.8\mu_0 J_c \rightarrow -0.4\mu_0 J_c \rightarrow 0$. The contour plot of this magnetic trap is shown in Fig. 3.8.

Figure 3.8: Formation of a self-sustaining trap with $B_{ext} = 0 \rightarrow 0.8\mu_0 J_c \rightarrow -0.4\mu_0 J_c \rightarrow 0$. Graph adapted from the paper by Zhang, B., *et al.* [44].

Single trap $\rightarrow$ ring trap

By applying $B_{ext} = 0 \rightarrow 1.2\mu_0 J_c \rightarrow -0.6\mu_0 J_c \rightarrow 0$, together with an additional B_{bias}^z , a quadrupole trap can be formed above the centre of the disk. This B_{bias}^z also shifts the field zero close to the chip surface further. Thus, there are two traps above the centre as shown in Fig. 3.9a with $B_{bias}^z = 0.03\mu_0 J_c$. By increasing the B_{bias}^z , the two traps are getting closer to each other until they merge. Fig. 3.9b shows the merging at $B_{bias}^z = 0.035\mu_0 J_c$. By increasing B_{bias}^z further, the merged trap is brought closer to the surface and becomes a ring trap. Fig. 3.9c shows this ring trap with a $B_{bias}^z = 0.06\mu_0 J_c$.

3.5.3 Ring structure

Fig. 3.5 (right) illustrates the simulated ring structure has an inner radius of $0.4a$ and an outer radius of a . The ring structure lies parallel to the xy -plane. B_{ext} is to be applied along the z -direction to introduce the supercurrents to the superconductor.

Single trap $\rightarrow$ ring trap (using a single magnetic field pulse)

For an external magnetic field $B_{ext} = 0 \rightarrow 0.8\mu_0 J_c \rightarrow 0$ applied to a ring superconductor, B_z v.s. z is plotted and shown in Fig. 3.10a. The sheet current density distribution is shown in the inset of Fig. 3.10b. For a $B_{bias}^z = -0.028\mu_0 J_c$, a quadrupole trap is formed at $z_0 \sim 0.7a$. The contour plot of this magnetic trap is shown in Fig. 3.10b. Increasing B_{bias}^z further will shift this quadrupole trap closer to the chip surface and deform it into a ring trap. Fig. 3.10c shows this ring trap with a $B_{bias}^z = -0.06\mu_0 J_c$.

Ring trap (using two external magnetic field pulses)

Fig. 3.10c shows a ring trap from a single magnetic field pulse. Using two magnetic field pulses will result in a ring trap of larger radius and further distance from the surface. Fig. 3.11 shows a ring trap generated by two pulses $B_{ext} = 0 \rightarrow \mu_0 J_c \rightarrow -0.5\mu_0 J_c \rightarrow 0$, and an additional $B_{bias}^z = 0.005\mu_0 J_c$.

Figure 3.9: $B_{ext} = 0 \rightarrow 1.2\mu_0 J_c \rightarrow -0.6\mu_0 J_c \rightarrow 0$ applied to a disk structure. a) Two traps are formed above the centre of the disk with $B_{bias}^z = 0.03\mu_0 J_c$ b) Two traps are merged with $B_{bias}^z = 0.035\mu_0 J_c$ c) Transformation to a ring trap with increasing bias field to $B_{bias}^z = 0.06\mu_0 J_c$ Graphs adapted from the paper by Zhang, B., *et al.* [44].

Figure 3.10: $B_{ext} = 0 \rightarrow 0.8\mu_0 J_c \rightarrow 0$ applied to a ring structure a) B_z v.s. z b) With $B_{bias}^z = -0.028\mu_0 J_c$, a quadrupole trap is formed at $z_0 \sim 0.7a$. [Inset] Sheet current density distribution along the radial direction. c) Formation of a ring trap with $B_{bias}^z = -0.06\mu_0 J_c$. Graphs adapted from the paper by Zhang, B., *et al.* [44].

Figure 3.11: With an $B_{ext} = 0 \rightarrow \mu_0 J_c \rightarrow -0.5\mu_0 J_c \rightarrow 0$, an additional $B_{bias}^z = 0.005\mu_0 J_c$ produces a ring trap. Graph adapted from the paper by Zhang, B., *et al.* [44].

3.6 Discussion

We have studied a variety of magnetic trapping potentials for ultracold atoms using the remanent state of type-II superconducting strip, disk and ring structures. We have numerically designed various magnetic potentials including a single quadrupole potential, double trapping potentials, self-sufficient potential and ring potential. We have also demonstrated that some potentials can be transformed from one geometry to another by varying the strength of homogeneous bias field. Thus, we have shown the rich versatility and reconfigurability of using vortex distribution in type-II superconductors.

The exclusion of the use of external current source for the self-sufficient trap eliminates the technical noise. In addition, the ring potential has a possibility to mimic superconducting quantum interference device (SQUID) [112, 113] as an atom circuit. In this case, the persistent flow of ultracold atoms can be induced in the ring trap as an analogue of the persistent current of the SQUID.

From an alternative perspective, the trapped atoms can be used as a probe of the vortices inside the superconductor. The properties of these traps such as position and potential geometries are sensitive dependent on the detailed behaviour of the vortices. Thus, these properties can be in turn used as a probe of the behaviour of the vortices like flux creeps or stability of vortices [114].

Chapter 4

Superconducting Vortex Trap

4.1 Introduction

This chapter will describe in details the various types of possible trapping potentials that can be realized using type-II superconducting strip and square structures. We begin with Sec. 4.2 to elaborate on the realization of a simple vortex trap by applying a parallel bias field B_{bias}^x on a superconducting strip that has been magnetized with a single external magnetic field pulse B_{ext} . Various parameters like lifetime, temperature and trap to surface distance with respect to B_{bias}^x of this trap are characterized. In addition, we also use the parameter trap to surface distance as an indicator to show the lifetime of the vortices inside the superconductor are longer than 2 hours. In addition we also use this parameter to show the hysteresis behaviour of the type-II superconductor by varying the magnitude and direction of B_{ext} . To show the effect of a transport current on the vortices inside the superconductor, we also use the trap to surface distance parameter to investigate its change with respect to the amplitude and direction of current. Sec. 4.2.5 discusses the realization of more complex trapping geometries with the use of multiple external magnetic field pulses on a superconducting strip. The complex potentials include a double trap and a self-sustaining trap above the strip. The next section, Sec. 4.3 discusses the trapping potential that can be formed on a superconducting square. Similar as the superconducting strip, we first show a simple realization of a trapping potential with a perpendicular bias field B_{bias}^z applied to the square that has been magnetized with a single pulse B_{ext} . We then highlight a possible realization of a self-sustaining trap without the use of any external magnetic fields. Similar to the superconducting strip, the square only requires the magnetization of the superconductor with two external magnetic field pulses B_{ext} of different magnitude but opposite directed. We measure its lifetime and estimate its trap depth. We characterize further this trap by fixing the amplitude of the first pulse. The parameters atom number and trap to surface distance are investigated by varying the amplitude of the second pulse. Motivated by the simulation where we transform a single trap to a ring trap using a superconducting disk Sec. 3.5.2, we experimentally investigate this transformation with a superconducting square. However, due to the non-radial symmetry of the square structure, we transform a single trap to four traps which is analogous to a four-way beam splitter.

Figure 4.1: a) For the external magnetic field $B_{ext} < B_{c1}$, the superconductor is in Meissner state b) For $B_{ext} > B_{c1}$, the superconductor is in vortex state c) After B_{ext} is removed, the superconductor is in remanent state d) Simulation of our vortex trap with an applied homogeneous bias field B_{bias}^x e) Absorption image of our vortex trap. Pictures adapted from the paper by Müller, T., *et al.* [58].

4.2 Magnetic trap using superconducting strip

In our superconducting atom chip experiment, ultracold atoms for ^{87}Rb are prepared in a standard laser cooling and trapping technique before being transferred to the magnetic trap. The details of these procedures are discussed in Chap. 2. The field gradient of the lower magnetic trap can be varied from 20 – 35 G/cm and the distance of this trap from the superconducting atom chip is 35 mm. Thus, the magnetic field B_{ext} on the surface of the chip from the magnetic trap is 69 – 121 G. These values are above the first critical field $B_{c1} = 25$ G but below second critical field B_{c2} of our YBCO type-II superconductor at 83 K.

If B_{ext} is below B_{c1} , the superconductor will be at Meissner state where it will repel any external magnetic field as illustrated in Fig. 4.1a. Since $B_{ext} > B_{c1}$, magnetic flux penetrates through the superconductor. Vortices are induced and they move from the edges towards the centre of the strip. The superconductor is said to be in the mixed state. The effect of penetration is shown in Fig. 4.1b. After B_{ext} is removed, vortices remain pinned to the pinning sites in our superconductor. This state is called remanent state and is shown in Fig. 4.1c.

The ultracold atoms from the lower magnetic trap are trapped for 160 ms before being transferred to the upper magnetic trap. We transfer the atoms by ramping down the magnetic field from the lower magnetic trap and simultaneously ramp up the magnetic

field from the transfer coils. This process takes 460 ms. At the end of this process, there are $\approx 1.5 \times 10^7$ atoms left. These atoms are further transferred closer to the superconducting strip with an additional homogeneous bias field. The magnetic field on the chip surface from the upper magnetic trap coils is smaller than the field from the lower magnetic trap coils. So, our superconductor will only retain the memory of the highest field, that is the magnetic field from the lower magnetic trap. In some experiments to be discussed later, if the B_{ext} requires a higher magnetic field (higher than 69 – 121 G) that is incapable to be supplied by the MOT power supply, car batteries (discussed in Sec. 2.7.1) are to be used. The current pulse from the batteries are applied first after cooling down our atom chip and before running the experimental cycle. In this situation, the type-II superconductor will only retain the memory of the highest applied magnetic field by the car batteries and ignore the lower magnetic field applied by the lower magnetic trap in each experimental cycle.

When the ultracold atoms are brought close to the superconducting strip, the magnetic fields are turned off. Only a homogeneous bias field along the x -direction B_{bias}^x is supplied. With this, a quadrupole trap is formed as shown by the simulated result in Fig. 4.1d. Since our atom chip is attached to a holder, the trap is formed only at one side of the superconducting strip. Fig. 4.1e shows the absorption image of 6×10^5 ultracold atoms from the vortex trap on a superconducting strip taken after 1 ms time-of-flight. Unless stated otherwise, our experiments are carried out using the 400 μm width Z-wire shown in Fig. 2.1c)

4.2.1 Transfer atoms from the upper magnetic trap to the vortex trap

To show the distortion of the quadrupole field from the upper magnetic trap due to the presence of vortices, we capture the in-situ image of the atomic cloud brought closer to the superconducting strip. The absorption image is shown in Fig. 4.2a. The field minimum of the upper magnetic trap is positioned at $z = 800 \mu\text{m}$ distance and $x = 600 \mu\text{m}$ to the side of the superconducting strip (II). A second field minimum is formed at closer distance near the edge of strip (I). It is shown by the high density region at $z = 250 \mu\text{m}$ and $x = 300 \mu\text{m}$. The qualitative features of the absorption image are matched closely to the simulated magnetic field shown in Fig. 4.2b.

Since there are two field minima, the aim is to optimize the transfer of the atom number from one field minimum to the other. The strategy is to overlap the field from the upper magnetic trap to the vortex trap. It is approached by trying different possible paths transferring to the vortex trap such that the final atom number is optimized. Fig. 4.2c shows the optimized vortex trap with an applied B_{bias}^x . The trap has a strong radial confinement in the x and z -direction. However, due to the structure of the Z-wire, it has a weak axial confinement along the y -direction.

4.2.2 Characterization of the vortex trap

We can trap up to a maximum number of 1×10^6 atoms in the vortex trap. In addition to B_{bias}^x , we also apply B_{bias}^z and B_{bias}^y to improve the trap. B_{bias}^z shifts the atomic cloud

Figure 4.2: a) Optical depth image of atomic cloud brought closer to the superconducting strip. The two field minima are denoted by I and II. b) Simulated result match the general features of the optical depth image. c) Vortex trap is formed after turning off the quadrupole field but maintaining the applied homogeneous bias field B_{bias}^x . Pictures adapted from the paper by Müller, T., *et al.* [58].

sideways. B_{bias}^y lifts the value of the trap bottom to minimize the Majorana spin flip loss of the atoms. The values for the homogeneous bias fields are typically at $B_{bias}^x = 40$ G, $B_{bias}^z = 7.7$ G and $B_{bias}^y = 3.3$ G.

Lifetime measurement

We first characterize the trap by measuring its lifetime. Shown in Fig. 4.3 is the graph for the total atom number v.s. its trapping time. A double exponential fit (red line) to the data (blue dots) shows two exponential decay time constants. They are 2.1 ± 0.2 s and 32 ± 2 s. The initial short decay time is due to evaporation of the hot atoms which have energy above the trap depth of the magnetic potential [33]. The second decay time constant is longer and should approximate the lifetime limited by the vacuum pressure. As expected, the measured decay time constant is close to the decay time obtained for the lower magnetic trap shown in Fig. 2.22¹. This measurement was performed with the applied bias fields of $B_{bias}^x = 14.0$ G, $B_{bias}^y = 3.7$ G, and $B_{bias}^z = 7.8$ G.

¹Our publications [58, 60] report lifetime of a few seconds. The longer lifetime here was measured after a new titanium layer was fired to the vacuum using our titanium-sublimation pump.

Figure 4.3: Lifetime measurement of our atomic cloud trapped using Z-wire. A double exponential fit (red line) to the data (blue dots) shows two exponential decay time constants. They are 2.1 ± 0.2 s and 32 ± 2 s.

Temperature measurement

Typically the temperature of an ultracold gas in conventional traps is measured via time-of-flight (TOF) measurement. However, it requires a zero magnetic field during the cloud expansion. In the trap generated by vortices the magnetic field cannot be switched off and with this TOF measurement cannot be used in characterizing the temperature of our atomic cloud. We employ a technique called RF (radio-frequency) spectroscopy following Fernholz, T., *et al.* [6]. An RF electromagnetic field induces spin-flip transition from trapped to untrapped state through multi-photon transitions. Only atoms with a Larmor frequency on resonance to the RF will make a spin-flip and leave the trapping region. The Larmor frequency is proportional to the absolute value of the magnetic field and with this to the potential height. A higher kinetic energy of an atom will always enable it to climb to higher potential. Thus, RF spectroscopy allows the measurement of kinetic energy distribution resembling a Maxwell-Boltzmann distribution.

For our vortex trap below the strip, an RF pulse in MHz range is irradiated for a time duration of 30 ms before the atom number is recorded. The comparison between the atoms loss due to RF pulse and the atom number without RF pulse allows us to obtain the fraction of the atom number loss. To demonstrate the versatility of RF spectroscopy, atomic cloud at various temperatures after evaporative cooling are investigated. Our results ² are plotted in Fig. 4.4. There are three data sets in this figure corresponding to the no RF evaporative cooling (red dots), evaporative cooling from 24 $\rightarrow$ 10 MHz (blue dots), and further cooling to 6 MHz (black dots). These three data sets are then fitted

²The various vortex traps discussed in this chapter trap the atomic cloud prepared in $|F = 2, m_F = 2\rangle$ state. However, for this RF spectroscopy results, it is performed on the $|F = 1, m_F = -1\rangle$ state. Nevertheless, it will not change the physics we want to highlight for this temperature measurement technique.

using 3-dimensional Maxwell-Boltzmann distribution function written as

$$dist(f) = a_1 + \frac{7.83 \times 10^{-6} a_2}{T^{3/2}} (f - f_0) \exp \left[-4.80 \times 10^{-5} \frac{f - f_0}{T} \right] \quad (4.1)$$

where a_1 , a_2 , f_0 and T are the fitting parameters. f_0 is introduced as an offset to the Maxwell-Boltzmann distribution function. Its value is equivalent to the magnitude of the trap bottom. T is the temperature of the atomic cloud. The resulting fits to these three data sets show an initial temperature of $218 \pm 23 \mu\text{K}$ (red line) being cooled to $66 \pm 4 \mu\text{K}$ (blue line) for an RF ramp from $24 \rightarrow 10$ MHz. A second ramp to 6 MHz continues to cool it further to $48 \pm 3 \mu\text{K}$ (black line). After this final cooling procedure, there are $\approx 4 \times 10^4$ atoms left.

Figure 4.4: RF spectroscopy performed on an atomic cloud trapped in the vortex trap using the superconducting strip. The results refer to no RF evaporative cooling (red dots), evaporative cooling from $24 \rightarrow 10$ MHz (blue dots), and a further cooling to 6 MHz (black dots). The fits show an initial temperature of $218 \pm 23 \mu\text{K}$ (red line) being cooled to $66 \pm 4 \mu\text{K}$ (blue line). The final procedure cools it further to $48 \pm 3 \mu\text{K}$ (black line).

Atom number v.s. B_{bias}^x

The atom number v.s. B_{bias}^x is an interesting topic to investigate as it shows the relation between the trap depth and B_{bias}^x . For this experiment, the superconducting strip is magnetized with $B_{ext} = 112$ G. The atom number is measured after the atomic cloud is released from the vortex trap with 1 ms TOF. The result is shown in Fig. 4.5. The atom number starts increasing from $B_{bias}^x = 4.6$ G. This minimum value approximates the minimum attractive force requires to overcome the gravitational field. Similar to a typical magnetic trap using a Z-wire [79], an increasing B_{bias}^x results in a closer distance to the strip. Closer distance gives rise to higher density and higher field gradient [79], which implies that the trap depth is increased and can trap much hotter atoms. The increase stops when B_{bias}^x reaches 40 G. The atom number drops with a further increased of B_{bias}^x as the cloud size approximates the trap to surface distance. This lead to collision of the atoms with the chip surface and results in an atom loss.

Figure 4.5: Graph showing the atom number v.s. B_{bias}^x . Atom number is increasing with B_{bias}^x but starts to drop when it is above 40 G. Graph adapted from the paper by Müller, T., *et al.* [58].

Trap to surface distance v.s. B_{bias}^x

The distance of the atomic cloud to the surface can be measured by performing in-situ imaging of the atomic cloud. We tilt the imaging beam and direct it towards the surface. The reflected beam is then directed to the CCD camera. Shown in Fig. 4.6 are a few examples of absorption images with two atomic clouds (one image, one real) taken along the axial and transverse direction of the Z-wire using two CCD cameras. Due to the vortices in the dimple wire, the atomic cloud is not spread over 5 mm length. Indeed, there are two atomic clouds each spread over half of the length. We show in the figure, by increasing the homogeneous bias field in the transverse direction, the atomic cloud can be brought closer to the surface. Fig. a) $\rightarrow$ b) $\rightarrow$ c) shows in the order of increasing bias field. We can relate the distance to the surface from the distance between the two atomic clouds in the optical depth image.

We perform the trap to surface distance v.s. B_{bias}^x measurement with two different values of external magnetic field pulse, $B_{ext} = 121$ G and 69 G. The results are shown by the curves in Fig. 4.7. As expected for the two curves, the distance is getting smaller with increasing B_{bias}^x . However, they saturate at a certain magnetic field. It occurs when the distance approximates the size of the flux region ($w - b$) discussed in Sec. 3.3. It is a similar effect to a typical atom chip experiment using fabricated wire where the smallest distance is limited by the finite size of the wire used (finite size effect) [1]. Overall, the data points agree well with the simulated curves (solid line 121 G, dashed line 69 G) where the size of the flux region is used as a free-scaling parameter. In the inset, we perform another measurement where B_{bias}^x is fixed at 10 G but with B_{ext} serves as a tuning parameter.

Stability of vortices

It is important to investigate if the vortex current (vortex relaxation) alter during the experimental run. Since the trap to surface distance is sensitive dependent on the vor-

Figure 4.6: Optical depth images obtained using 2 CCD cameras imaging along the axial and transverse direction of the Z-wire. Two atomic clouds are seen in the images due to reflection from the chip. By increasing the bias field along the x -direction, the atomic cloud is brought closer to the surface. The bias field is increasing in the order of a) $\rightarrow$ b) $\rightarrow$ c).

Figure 4.7: Graph showing two curves of the trap to surface distance v.s. B_{bias}^x for $B_{ext} = 121$ G and 69 G. Overall, the experimental results agree well with the simulation. [Inset] Trap to surface distance v.s. B_{ext} with B_{bias}^x fixed at 10 G. Graph adapted from the paper by Müller, T., *et al.* [58].

tices, a measurement of this parameter v.s. time can give valuable information about the behaviour of vortices. In Fig. 4.8, we plot the trap to surface distance at different time Δt . At each Δt , trap to surface distance for different values of $B_{bias}^x = 6.6, 13, 19, 26$ and 33 G are taken. Magnetization using external magnetic field pulse $B_{ext} = 76$ G is applied initially at time $\Delta t = 0$ s. At this time, 5 trap to surface distance data points corresponding to the 5 different B_{bias}^x are taken. The purpose for these measurements are to serve as a reference for subsequent measured readings. After these measurements, a single magnetization of $B_{ext} = 200$ G is applied once to the superconductor. Afterwards, 5 trap to surface distance data points are taken again. The trap to surface distance for various B_{bias}^x are measured to be higher due to higher magnitude of applied B_{ext} . Same measurements are repeated for subsequent time. All the plots with different B_{bias}^x suggest the vortices are stable over the time scale of 140 minutes. Our results are consistent with the measurements of the relaxation of vortices using different techniques [115].

4.2.3 Effect of transport current on vortices

Using a similar technique as discussed in Sec. 4.2.2, we measure the trap to surface distance for an applied transport current after the superconductor is magnetized with external magnetic field pulse B_{ext} . The transport current can cause motion and dissipation of vortices. The effects are complex and depend on the pinning energy at the defects [43]. We do not model these effects and only present the experimental results to show there exists a detectable change by the atoms.

We apply $B_{ext} = 200$ G and the transport current is chosen from values between -0.5 A and 0.5 A [43]. The experimental procedure is done initially by trapping the

Figure 4.8: Measurement of the stability of vortices over time. Trap to surface distance are plotted v.s. time with different B_{bias}^x . At $\Delta t = 0$ s, $B_{ext} = 76$ G is applied to magnetize the superconductor. Subsequent measurements are carried out with $B_{ext} = 200$ G. Graph adapted from the paper by Müller, T., *et al.* [60].

atoms for 100 ms with the magnetic field generated by the supercurrents. Afterwards, the transport current is applied to the superconducting strip for 200 ms. It is to be switched off after this time and the atoms which are still held in its trap are trapped for 100 ms longer. This 100 ms waiting time is necessary to ensure the magnetic field generated by the transport current is no longer affecting the atomic cloud. Nevertheless, after this 100 ms, the transport current has already altered the vortices distribution and will reflect accordingly in the magnetic potential for the atomic cloud.

Fig. 4.9a and 4.9b show the vertical and horizontal position of the atomic cloud with respect to transport current. The vertical position is measured with respect to the chip surface whereas the horizontal position is measured from an arbitrary reference position. Fig. 4.9a shows the trap to surface distance is reduced for higher applied transport current irrespective with the current direction. The decrease of the distance shows that the supercurrents inside the superconductor are reduced, resulting in lowering of the overall magnetic field. Fig. 4.9b shows a significant shift in horizontal direction for transport current in negative direction. However, the horizontal shift is minimal for positive transport current. From this, we infer in addition to a decrease of supercurrents, there is also a redistribution of the vortices, changing the overall supercurrents distribution.

4.2.4 Hysteresis behaviour of type-II superconductor

We study the hysteresis behaviour of type-II superconductors by measuring the trap to surface distance with respect to a sequential order of varied magnetic field used for magnetization. Fig. 4.10a shows the plot of the results of this study. The arrows with a number indicated on top show the increasing or decreasing direction of the applied magnetic field. The number 1 correspond to $0 \rightarrow 220$ G, number 2 is $220 \rightarrow 0$ G, number

Figure 4.9: Plots of the vertical position (a)) and horizontal position (b)) of the atomic cloud with respect to the transport current. Graphs adapted from paper by Zhang, B., *et al.* [43].

3 is $0 \rightarrow -220$ G (reversed), number 4 is $-220 \rightarrow 0$ G, and number 5 is $0 \rightarrow 220$ G.

Taking as an example loading sequence 1. Initially, the superconductor starts from a virgin state. Then using the standard laser cooling and trapping technique, the atoms are transferred near to the superconductor. By turning on the homogeneous bias field, the trapped position is recorded. However, due to the lower magnetic trap used, a field of 76 G is unavoidable at the chip position, which induces supercurrents. Thus, there exists a trap even with the magnetic field pulse of 0 G. In the next measurement, the experiment is repeated as before but by applying a magnetic field pulse of 50 G to the superconductor beforehand. Similarly, ultracold atoms are prepared using standard laser cooling and trapping technique and then transported near the superconductor with the same parameters. Then, the trapped atoms position is again measured. The same procedure is repeated for the applied magnetic field pulses of 100 G, 120 G, 150 G and 200 G.

In the loading sequence 2, magnetic field pulses are applied in a decreasing order. Beginning with 150 G, decreasing to 100 G, 50 G until 0 G where for each prepared magnetization, the trapped atom position is recorded. Due to the memory effect of the highest applied field of 200 G, the trapped positions are unchanged for the decreased applied magnetic field.

In loading sequence 3, a magnetic field is increased from 0 G to 200 G but in opposite direction. Antivortices are introduced to the superconductor and lead to an overall decrease of the magnetization. Thus, the trap to surface distance is reduced. The polarity of the reversed magnetization can be changed completely with an applied field B_R . B_R depends on the thermodynamic critical field B_c (see Sec. 3.19) and also on the previous highest applied magnetic field which in our case is 200 G. However, due to 76 G of positive magnetization from lower magnetic trap in each experimental run, it is still possible to trap atoms from the partial positive magnetization of the superconductor.

In the loading sequence 4, the magnetic field decrease again in the order of -200 G, -150 G, -50 G, until 0 G and increased to 20 G, 50 G and 70 G. Again, due to memory of the highest applied magnetic field pulse of -200 G, no pronounced change of the magnetic trap position for these series of magnetic field pulses. No change occurs for positive

Figure 4.10: a) Hysteresis behaviour of the superconductor is investigated by measuring the trapped position of the atomic cloud with respect to the magnetic field pulse applied to the superconductor. The numbers 1, 2, 3, 4, and 5 above the arrows drawn denote the sequential order of the applied pulses. The number 1 correspond to $0 \rightarrow 220$ G, number 2 is $220 \rightarrow 0$ G, number 3 is $0 \rightarrow -220$ G (reversed), number 4 is $-220 \rightarrow 0$ G, and number 5 is $0 \rightarrow 220$ G. b) Simulated hysteresis behaviour using the mesoscopic model discussed in Sec. 3.2.1. The homogeneous bias field is $B_{bias}^x = 0.17B_c$.

magnetization of 20 G, 50 G and 70 G due to the unavoidable 76 G magnetization of the lower magnetic trap. Same as in loading sequence 3, due to reverse magnetization, there are two vortex traps above the edges of the superconducting strip. However, we always take the position of the atomic cloud on the same side as the positive magnetization.

In loading sequence 5, the magnetic field pulse is increased from 80 G, 90 G, 100 G, 152 G to 200 G. The change of the trap position is pronounced in this sequence, since the magnetization is above 76 G. The overlap of the curve for loading sequence 1 and 5 above 100 G shows that the magnetization of the superconductor can be reversed.

The overall qualitative behaviour of the hysteresis can be simulated based on the mesoscopic model discussed in Sec. 3.2.1. The results of our simulation is shown in Fig. 4.10b and agrees well with the experimental results.

4.2.5 Programmable vortex trap using superconducting strip

The characterization of the vortex trap discussed in Sec. 4.2.2 is based on the single trap formed due to a single magnetic field pulse. One can partially reverse the magnetization of the superconductor as discussed in Sec. 4.2.4 to create different types of trapping potentials.

Double traps above superconducting strip

Fig. 4.11a is the in-situ absorption image of the double trap for an applied homogeneous bias field after the application of multiple magnetic field pulses. The pulse sequence is $220 \rightarrow -220 \rightarrow 76$ G. The application of the first 220 G magnetic field pulse is to show that it can be overwritten by the second -220 G magnetic field pulse. Thus, only the

Figure 4.11: a) In-situ absorption image of the double traps for an applied homogeneous bias field after the application of magnetic field pulse sequence of $220 \rightarrow -220 \rightarrow 76$ G. b) Simulation of the contour map of the magnetic field distribution for these double traps. Graphs adapted from the paper by Müller, T., *et al.* [60].

Figure 4.12: a) Self-sustaining trap after the loading magnetic field pulse sequence of $-220 \rightarrow 76$ G b) Simulation of the contour map of the magnetic field distribution for the self-sustaining trap. Graphs adapted from the paper by Müller, T., *et al.* [60].

-220 G and 76 G pulses are relevant for the creation of double traps. The simulation of the experimental result is shown in Fig. 4.11b using the relevant parameters for these two traps.

Self-sustaining trap above the superconducting strip

It is possible to create a trapping potential without any external magnetic bias field. Such a trap is formed entirely by the persistent supercurrents. We show this self-sustaining trap by applying a magnetic field pulse sequence $-220 \rightarrow 76$ G. In principle, a self-sustaining is also possible with magnetic field pulse sequence of $220 \rightarrow -76$ G. However, due to the unavoidable magnetization from the magnetic trap of each experimental run, we choose $-220 \rightarrow 76$ G. The absorption image and the simulation are shown in Fig. 4.12a and 4.12b respectively.

4.3 Magnetic trap using superconducting square

There are different trapping potentials that can be realized with a superconducting square structure. As shown in Fig. 4.13, applying an external magnetic field pulse B_{ext} between B_{c1} and B_{c2} , a certain current distribution is induced. From a far field approximation, the supercurrent can be visualized as a single persistent current, I circling around the

Figure 4.13: Schematic diagram for an applied external magnetic field pulse B_{ext} inducing a supercurrent I inside a type-II superconducting square structure. [Inset] A perpendicular homogeneous bias field $B_{bias}^z = 24.5$ G applied to the superconducting square that has been magnetized with $B_{ext} = 278$ G forms a single trap above. Graph adapted from the paper by Siercke, M., *et al.* [61].

edge of the square. The inner current as discussed in Sec. 3.3.1 is considered to be small and with a negligible contribution to the trapping potential in the far field. A trap can be generated by applying a perpendicular bias field $B_{bias}^z = 24.5$ G to a superconducting square that has been magnetized with $B_{ext} = 278$ G. The absorption image of this trap is shown in the inset of Fig. 4.13.

4.3.1 Self-sustaining trap above the superconducting square

It is possible to realize a self-sustaining trap based only on the vortex current inside the superconductor. To achieve this, we apply the pulse sequence $B_{ext} = 600 \rightarrow -226$ G to the superconducting square. In the beginning, the atomic cloud is brought closer to the superconducting square using the quadrupole field generated by the transfer coils (upper magnetic trap). This field will merge with the magnetic field generated from the supercurrents in the superconducting square. If the atomic cloud is brought close enough to the superconductor, the atoms will follow the spatial density distribution of the supercurrents. The absorption image of this spatial density distribution of the atomic cloud is shown in Fig. 4.14. Inside the figure, the magnetization from the supercurrents based on Bean's model [106] is drawn. It matches qualitatively the features of our atomic cloud at this distance. x -axis of the plot is the position of our square structure in units of its length L .

When the upper magnetic trap is turned off, a quadrupole magnetic potential is generated solely by the supercurrents. This self-sustaining trap has a sufficient trap depth to trap the atoms. Fig. 4.15 shows the absorption image of this trap with 10^3 atoms at a trap to surface distance of $155 \mu\text{m}$. To maximize the atom number in this trap, the field minimum at the upper magnetic trap is brought to the field minimum of the self-sufficient trap in 300 ms before all the magnetic fields are turned off. This is done to directly load the atoms to the new trapping potential. However, the location of the field minimum of the self-sustaining trap is modified due to the presence of the quadrupole

Figure 4.14: Absorption image of the atomic cloud brought closer the superconducting square that has been magnetized with external magnetic field pulses of $B_{ext} = 600 \rightarrow -226$ G. At close distance, the spatial density of the atomic cloud follows the magnetization pattern by the supercurrents. [Inset] A plot of the magnetization of the superconductor based on the magnetic field pulses parameter using Bean's model [106]. Picture adapted from the paper by Siercke, M., *et al.* [61].

Figure 4.15: Absorption image of the ultracold atoms in the self-sustaining trap formed solely by the persistent currents inside the superconducting square. The atomic cloud at the bottom is a mirror image due to tilted imaging beam to the reflective surface. Picture adapted from the paper by Siercke, M., *et al.* [61].

field of the superimposed magnetic trap. The atomic cloud in the upper magnetic trap has a temperature of 200 μK . The loading efficiency with 10^3 atoms loaded to the self-sustaining trap indicates a shallow trap potential. We can estimate the trap depth of self-sustaining trap using the kinetic energy distribution of the atoms at a temperature of 200 μK . This estimate gives us a trap depth of 10 μK . In the Fig. 4.15, two atomic clouds are seen. These two atomic clouds are to be used to calibrate the trap to surface distance to the chip surface as described by Smith, D. A., *et al.* [86]. The atomic cloud at the bottom is a mirror image due to tilted imaging beam to the reflective surface.

Lifetime measurement

Fig. 4.16 shows the lifetime measurement of this self-sustaining trap. The double exponential fit to the data gives an initial decay time of 50 ms and a second longer decay time of 1.5 s. The initial fast decay is attributed to the evaporation of hot atoms. However, the longer decay time is due to Majorana spin flip loss rate [116]. This decay time τ_0 is consistent with the radius r of the atomic cloud shown as [116]

$$\tau_0 \approx \frac{m}{\hbar} r^2 \quad (4.2)$$

There are several schemes to overcome this spin flip loss rate. Since the loss rate is solely dependent on the size of the atomic cloud, a straightforward approach is to increase the trapping potential to a larger size. It can be done by using a larger superconducting square structure or increasing its critical current density with much lower cryogenic temperature. Alternative we can use RF dressing to remove its field minimum [117] or to apply a rotating bias field to the existing quadrupole field to average the field minimum to a non-zero value [116].

Variation of trap to surface distance

The variation of the relative magnitude between the first and second external magnetic field pulse will change the trap to surface distance. We fix the first external magnetic field pulse at 600 G, and investigate the trap to surface distance by varying the magnitude of the second pulse. Fig. 4.17 shows the trap distance with respect to the pulse height. As expected, an increase of the second pulse amplitude brings the field minimum of the self-sustaining trap closer to the chip surface. Also in the same figure is the relation between the atom number and the magnitude of the second pulse.

Transformation from a single trap to four traps

As discussed in Sec. 3.5.2, for two opposite magnetic field pulses applied the superconducting disk structure, it is possible to create a single trap and transform it into a ring trap. It can be done by applying an increasing perpendicular bias field B_{bias}^z to the disk superconductor. However, due to the four sides of our square structure, instead of a ring trap, four trapping potentials above the corners are formed. To study this, the superconductor is magnetized with the double pulse sequences same as the self-sustaining trap as discussed in Sec. 4.3.1, that is $B_{ext} = 600 \rightarrow -226$ G. The schematic diagram to

Figure 4.16: a) Lifetime measurement of the self-sustaining trap gives two decay time constants of 50 ms and 1.5 s. The fast decay is due to escape of the hot atoms above the trap depth of the trapping potential while the longer decay time is due to Majorana spin flip loss. Graph adapted from the paper by Siercke, M., *et al.* [61].

Figure 4.17: Plot of the atom number (blue diamonds) and trap to surface distance (black squares) with respect to the amplitude of the second pulse of the self-sustaining trap. Graph adapted from the paper by Siercke, M., *et al.* [61].

Figure 4.18: a) Schematic diagram illustrating the alteration from a single trap to four traps with the use of a perpendicular bias field B_{bias}^z . b) When B_{bias}^z is small, only a single trap is formed above the square. c) An increase of B_{bias}^z transforms the single trap to the four traps above the corners of the square structure. Only two traps are seen due to integrated direction of the imaging beam. Pictures adapted from the paper by Siercke, M., *et al.* [61].

illustrates the evolution from a single trap to four traps with applied B_{bias}^z is show in Fig. 4.18a. Shown in Fig. 4.18b is the absorption image of the atomic cloud held by the upper magnetic trap with an additional perpendicular bias field B_{bias}^z . When B_{bias}^z is small, only a single trap is formed above the square. An increase of B_{bias}^z further cancels the stronger near field of the chip surface. With this, it transforms the single trap to four traps. The absorption image of these four traps is shown in Fig. 4.18c. Due to the direction of the imaging beam, only two traps are visible.

Part II

Chapter 5

Rydberg atoms

5.1 Introduction

Rydberg atoms have electrons in highly excited states but are not yet ionized [118]. Rydberg atoms have enhanced properties compared to ground state atoms such as increased sensitivity to electric field, stronger dipole-dipole interactions, longer radiative lifetime and larger orbital radius. These properties find applications for example in qubit operation [119], simulation of crystal lattice structure [120], studies of van der Waals interaction [121], electrometry [122] and Förster resonance [123]. In this chapter, I will highlight several properties of Rydberg atoms and its scaling laws with principal quantum number n . This is followed by modelling for the radial wavefunctions of rubidium atoms which can be solved numerically using the Numerov's method. The numerically solved radial wavefunctions allow us to calculate the radial matrix elements which are then used to generate Stark maps. The obtained Stark maps enable us to find the corresponding Stark shift of a quantum state in the presence of electric field. A final section is dedicated to electromagnetic induced transparency (EIT) [124, 125]. This section will introduce Rydberg EIT which makes use of the enhanced electric field sensitivity of Rydberg state as an electrometer.

5.2 Properties of Rydberg atoms

One particular kind of Rydberg atoms are alkali Rydberg atoms. They have one valence electron, a nucleus and a closed shell electrons which makes them similar to hydrogen atoms. The binding energy, orbital radius, polarizability, radiative lifetime, etc scale with n for these Rydberg atoms. These scalings are listed in Table 5.1.

Property	n dependence
Binding energy	n^{-2}
Energy between adjacent n states	n^{-3}
Orbital radius	n^2
Geometric cross section	n^4
Dipole moment $\langle nd er nf \rangle$	n^2
Polarizability	n^7
Radiative lifetime	n^3
Fine-structure interval	n^{-3}

Table 5.1: Properties of Rydberg atoms [118].

5.3 Radial wavefunctions

To solve the radial wavefunctions, we employ the time independent Schrödinger equation which is written in the form

$$-\frac{\hbar^2}{2m}\nabla^2\psi + V\psi = E\psi \quad (5.1)$$

By using Laplacian in the spherical coordinates [126], this time-independent Schrödinger equation is

$$-\frac{\hbar^2}{2m}\left[\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial\psi}{\partial r}\right) + \frac{1}{r^2\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial\psi}{\partial\theta}\right) + \frac{1}{r^2\sin^2\theta}\left(\frac{\partial^2\psi}{\partial\phi^2}\right)\right] + V\psi = E\psi \quad (5.2)$$

For a special case where the potential V depends only on r , the solutions of the wavefunction can be separable into the multiplication of radial part and spherical part

$$\psi(r, \theta, \phi) = R(r)Y_{l,m}(\theta, \phi) \quad (5.3)$$

The radial part of the differential equation can become

$$\left[-\frac{\hbar^2}{2m}\left(\frac{d^2}{dr^2} + \frac{2}{r}\frac{d}{dr}\right) + \frac{\hbar^2}{2m}\frac{l(l+1)}{r^2} + V(r)\right]R(r) = ER(r) \quad (5.4)$$

As a matter of convention, it is convenient to write R as a function of $a_0 = \frac{4\pi\epsilon_0\hbar^2}{me^2}$. m is the reduced mass of the quantum system, and when $m = m_e$, a_0 is Bohr radius. By substituting above equation with $r \rightarrow ra_0$, the equation becomes

$$\left[-\left(\frac{d^2}{dr^2} + \frac{2}{r}\frac{d}{dr}\right) + \frac{l(l+1)}{r^2} + V(r)\right]R(r) = ER(r) \quad (5.5)$$

The energy E for hydrogen in this transformed coordinates is

$$E = -\frac{1}{2n^2} \quad (5.6)$$

while the potential V is

$$V = -\frac{1}{r} \quad (5.7)$$

One can easily recover the original energy by multiplying with $\frac{\hbar^2}{ma_0^2}$. Similarly, the potential can be retrieved by a resubstitution of $r \rightarrow r/a_0$ to Eq. 5.7 and multiplying it with $\frac{\hbar^2}{ma_0^2}$. From now on, the potential and energy are in unit of a_0 unless specified otherwise.

5.3.1 Numerov's method

To numerically obtain the radial wavefunction, Numerov's method is used. Schrödinger equation is modified and transformed to an equation called Numerov's equation such that it can be numerically solved. This Numerov's equation is a second order ordinary differential equation of the form

$$\frac{d^2 X}{dx^2} = g(x)X \quad (5.8)$$

where the first order term is absent [127]. If $X(x-h)$ and $X(x)$ are known, calculation of the next iterative term $X(x+h)$ can be obtained from the relation

$$X(x+h) = \frac{[2 + 10h^2g(x)/12]X(x) - [1 - h^2g(x-h)/12]X(x-h)}{1 - h^2g(x+h)/12} + O(h^6) \quad (5.9)$$

5.3.2 Numerov's method for hydrogen and alkali metal atoms

There are two types of scaling called square-root scaling [128] and logarithmic scaling [129]. They can be applied to Eq. 5.5 to obtain the differential equation in Numerov's form shown by Eq. 5.8.

Hydrogen atom

For hydrogen atom with potential of $V = -1/r$, square-root scaling substitutes

$$x = \sqrt{r} \quad (5.10a)$$

$$X(x) = Rr^{3/4} \quad (5.10b)$$

to Eq. 5.5 to produce $g(x)$ of the form

$$g(x) = -8 - 8Ex^2 + \frac{(2l + \frac{1}{2})(2l + \frac{3}{2})}{x^2} \quad (5.11)$$

For logarithmic scaling, the substitutions are

$$x = \ln(r) \quad (5.12a)$$

$$X(x) = R\sqrt{r} \quad (5.12b)$$

and produce $g(x)$ of the form

$$g(x) = 2e^{2x} \left(-\frac{1}{r} - E \right) + \left(l + \frac{1}{2} \right)^2 \quad (5.13)$$

The studies by Pritchard, J.D. suggest logarithmic scaling produces much higher errors for Rydberg state with higher n [130]. So, for our numerical simulation of radial wavefunctions, we use square-root scaling.

Alkali metal atoms

For alkali metal atoms, the same numerical techniques to find the radial wavefunction of hydrogen as discussed earlier are employed. A substitution of Eqs. 5.10 to the Schrödinger Eq. 5.5 results in a more generic $g(x)$ written as

$$g(x) = -8x^2V - 8Ex^2 + \frac{(2l + \frac{1}{2})(2l + \frac{3}{2})}{x^2} \quad (5.14)$$

which is readily solved once the potential V and energy E are known.

Potential V

Alkali atoms differ from hydrogen atom where the potential V is purely between a single proton and a single electron Coulombic interaction $V = -1/r$. For alkali atoms, the valence electron with low orbital angular momentum l has a chance of penetrating the closed shell electrons and interacting with the unshielded nuclear charges. Thus, the electron will experience a stronger Coulombic potential compared to those with higher l at short distance [118]. In addition, there is an effect of valence electron polarizing the ionic core due to its electric field at short distance [131]. The potential V of an alkali atom by taking into account of core penetration and core polarization effects has been calculated by Marinescu, *et al.* [131]. It is called parametric model potential and is given by

$$V_l(r) = -\frac{Z_l(r)}{r} - \frac{\alpha_c}{2r^4}(1 - e^{-(r/r_c)^6}) \quad (5.15a)$$

$$Z_l(r) = 1 + (Z - 1)e^{-a_1r} - r(a_3 + a_4r)e^{-a_2r} \quad (5.15b)$$

The first term of V_l describes the core penetration effect for different l . Z is the nuclear charge of the corresponding alkali metal atoms. The l -dependent parameters a_1, a_2, a_3, a_4 and r_c [131] for rubidium atoms are listed in Table 5.2. The effect of core polarization is described in the second term of V_l . α_c is the static polarizability of the positive ion core. For rubidium atoms, α_c has a value of 9.0760. r_c is the cutoff radius introduced to account for unphysical short range contribution to this core polarization potential [131]. By including the spin orbit coupling potential,

$$V_{so} = \frac{\alpha^2}{2r^3} \mathbf{L} \cdot \mathbf{S} \quad (5.16)$$

	a_1	a_2	a_3	a_4	r_c
$l = 0$	3.69628474	1.64915255	-9.86069196	0.19579987	1.66242117
$l = 1$	4.44088978	1.92828831	-16.79597770	-0.81633314	1.50195124
$l = 2$	3.78717363	1.57027864	-11.65588970	0.52942835	4.86851938
$l \geq 3$	2.39848933	1.76810544	-12.07106780	0.77256589	4.79831327

Table 5.2: Parameters of rubidium atoms used for the core penetration term in the parametric model potential. Parameters for other species of alkali metal atoms, please refer to publication by Marinescu, *et al.* [131]

where $\mathbf{L} \cdot \mathbf{S} = \frac{1}{2} [j(j+1) - l(l+1) - s(s+1)]$ and α is the fine structure constant. The total potential V is just $V = V_{so} + V_l$

Energy E

The core polarization and core penetration effects are particularly dominant among those quantum states with low angular momentum. These effects lead to the depression of its energy and can be accounted for by introducing quantum defect term δ_{nlj} related to n through Rydberg-Ritz expression [118]. These equations are written as

$$E = -\frac{1}{2n^*} \quad (5.17a)$$

$$n^* = n - \delta_{nlj} \quad (5.17b)$$

$$\delta_{nlj} = \delta_0 + \frac{\delta_2}{(n - \delta_0)^2} + \frac{\delta_4}{(n - \delta_0)^4} + \frac{\delta_6}{(n - \delta_0)^6} + \frac{\delta_8}{(n - \delta_0)^8} + \dots \quad (5.17c)$$

where n^* is the effective principal quantum number. $\delta_0, \delta_2, \delta_4, \delta_6$ and δ_8 are l and j dependent quantum defect constants and are experimentally determined. It is sufficient to have the first two terms of δ_{nlj} for high n states ($n > 20$) [118, 132, 133]. For our simulations, all these parameters are obtained from different sources and listed in table 5.3.

5.3.3 Rydberg wavefunctions of rubidium atoms

For the numerical calculation of the radial wavefunctions of rubidium, we follow the approach by Zimmerman, M. L., *et al.* [129]. Numerov's equation as express in Eq. 5.8 is integrated by using the $g(x)$ written in Eq. 5.14. It is done by solving the iterative equation (Eq. 5.9) beginning with higher radius $r_s = 2n(n+15)$ and choosing a step size of $h = 0.01$. This iterative equation is simulated iteratively from higher radius to smaller radius. It is truncated at an inner radius of some finite values due to transient and diverging behaviour of the radial wavefunction at short distance. This inner radius is chosen to be $\alpha_c^{1/3}$ for low l state ($l \leq 3$). For high l state, the inner truncation is determined such that the radial wavefunction does not begin diverging. The two initial conditions $X(x-h)$ and $X(x)$ are known to be zero at large distance, and for our applications, we choose sufficiently low values $X(x-h) = 2 \times 10^{-8}$ and $X(x) = 1 \times 10^{-8}$ to approximate this zero value. The choice of whether $X(x-h) > X(x)$ or $X(x-h) < X(x)$ is arbitrary

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5.3. Radial wavefunctions

L_J	δ_0	δ_2	δ_4	δ_6	δ_8
$S_{1/2}$	3.1311807(8) [132]	0.1787(2) [132]	-1.8 [118]	0 [118]	0 [118]
$P_{1/2}$	2.6548849(10) [133]	0.2900(6) [133]	-7.904 [118]	116.437 [118]	-405.907 [118]
$P_{3/2}$	2.6416737(10) [133]	0.2950(7) [133]	-0.97495 [118]	14.6001 [118]	-44.7265 [118]
$D_{3/2}$	1.3480948(11) [132]	-0.6054(4) [132]	-1.50517 [118]	-2.4206 [118]	19.736 [118]
$D_{5/2}$	1.3464622(11) [132]	-0.5940(4) [132]	-1.50517 [118]	-2.4206 [118]	19.736 [118]
$F_{5/2}$	0.0165192(9) [134]	-0.085(9) [134]	-0.36005 [118]	3.2390 [118]	0 [118]
$F_{7/2}$	0.0165437(7) [134]	-0.086(7) [134]	-0.36005 [118]	3.2390 [118]	0 [118]
$l > 3$	0	0	0	0	0

Table 5.3: Quantum defect constants for an n Rydberg state with angular momentum $l = \{0, 1, 2, 3\}$. For $l > 0$, the quantum defect constants are zero. Constants referred to references Li, W. H., *et al.*, Han, J. N., *et al.*, and Gallagher, T.F. [133, 134, 118] are experimentally determined for ^{85}Rb . As the constants do not differ significantly for ^{85}Rb and ^{87}Rb isotopes [132], thus the constants of ^{85}Rb are used in our calculations.

as it will only invert the radial wavefunction along the $R(r) = 0$ axis. The obtained $X(x)$ and x are to be inverted to obtain the radial wavefunction $R(r)$ and its position r .

$$r = x^2 \quad (5.18a)$$

$$R = Xr^{-3/4} \quad (5.18b)$$

To produce the normalized radial wavefunction, R is further multiplied with a normalization constant n_c

$$n_c = \frac{1}{\sqrt{\int r^2 R^*(r) R(r) dr}} \quad (5.19)$$

To show an example, a normalized radial wavefunction of quantum state $|n = 35, l = 18, j = 35/2\rangle$ is plotted in Fig. 5.1.

Radial matrix elements

After the normalized radial wavefunction is simulated, the calculation of the radial matrix element $\langle n, l, j | r | n', l', j' \rangle$ between two quantum states is straightforward where

$$\langle n, l, j | r | n', l', j' \rangle = \int r^2 R_{n,l,j}^* r R_{n',l',j'} dr \quad (5.20)$$

Fig. 5.2 illustrates the function of the integrand of a radial matrix element where the numerical integration is performed with trapezoidal sum of the area under the curve. Relevant calculated radial matrix elements are listed in the Table 5.4. They are in exact agreement with the reference by Pritchard, J. D. [130].

Figure 5.1: Normalized radial wavefunction for rubidium atoms of quantum state $|n = 35, l = 18, j = 35/2\rangle$ simulated using Numerov's method.

5.4 Stark map of rubidium atoms

The total Hamiltonian $\hat{H}$ for an atom in the presence of electric field can be written as

$$\hat{H} = \hat{H}_0 + \hat{H}' \quad (5.21a)$$

$$\hat{H}' = -\mathbf{p} \cdot \mathbf{E}' \quad (5.21b)$$

$$\mathbf{p} = -e\mathbf{r} \quad (5.21c)$$

$\langle n, l, j r n', l', j' \rangle$	calculated value (a_0)
$\langle 46, 2, 5/2 r 48, 1, 3/2 \rangle$	1551
$\langle 46, 2, 5/2 r 44, 3, 7/2 \rangle$	1586
$\langle 46, 2, 5/2 r 47, 1, 3/2 \rangle$	2708
$\langle 46, 2, 5/2 r 45, 3, 7/2 \rangle$	2694
$\langle 50, 2, 5/2 r 52, 1, 3/2 \rangle$	1844
$\langle 50, 2, 5/2 r 48, 3, 7/2 \rangle$	1893
$\langle 35, 2, 5/2 r 37, 1, 3/2 \rangle$	877
$\langle 35, 2, 5/2 r 33, 3, 7/2 \rangle$	882

Table 5.4: Calculated radial matrix elements using the simulated Rydberg wavefunctions. These values are found to have an exact match to the reference by Pritchard, J. D. [130].

Figure 5.2: A plot of the integrand for the radial matrix element $\langle 46, 2, 5/2 | r | 48, 1, 3/2 \rangle$. Numerical integration by summing the area under the curve shows that $\langle 46, 2, 5/2 | r | 48, 1, 3/2 \rangle = 1551a_0$.

where $\hat{H}_0$ is the unperturbed Hamiltonian of the atomic quantum system. $\hat{H}'$ is the Stark Hamiltonian showing a direct coupling between an atom with dipole moment $\mathbf{p}$ and the external electric field $\mathbf{E}'$. Assuming the electric field is pointing along the $\hat{z}$ direction, $\hat{H}' = ezE' = er \cos \theta E'$.

We obtain the Stark map by diagonalizing the matrix of the total Hamiltonian considering the coupling of all individual quantum states among each other from a total number of i quantum states. For our application, choosing $\Delta n = \pm 5$ is sufficient. The matrix of the total Hamiltonian to be diagonalized is given by

$$\begin{pmatrix} E_1 & 0 & 0 & \cdots & 0 \\ 0 & E_2 & 0 & \cdots & 0 \\ 0 & 0 & E_3 & \cdots & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & 0 & \cdots & E_i \end{pmatrix} + eE' \begin{pmatrix} 0 & \langle 1|z|2 \rangle & \langle 1|z|3 \rangle & \cdots & \langle 1|z|i \rangle \\ \langle 2|z|1 \rangle & 0 & \langle 2|z|3 \rangle & \cdots & \langle 2|z|i \rangle \\ \langle 3|z|1 \rangle & \langle 3|z|2 \rangle & 0 & \cdots & \langle 3|z|i \rangle \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ \langle i|z|1 \rangle & \langle i|z|2 \rangle & \langle i|z|3 \rangle & \cdots & 0 \end{pmatrix} \quad (5.22)$$

The first matrix is a diagonal matrix with all its unperturbed energy of all quantum states. For rubidium atoms, the energies are shown in Eqs. 5.17. To obtain the energy in correct unit, Eq. 5.17a is further multiplied with a constant factor $\frac{\hbar^2}{ma_0^2}$ before inserting it into this diagonal matrix. For the second term of Matrix 5.22, the off-diagonal element

is mathematically represented by [129]

$$\begin{aligned} \langle n, l, j, m_j | z | n', l', j', m'_j \rangle &= \delta_{m_j, m'_j} \delta_{l, l' \pm 1} \langle n, l, j | r | n', l', j' \rangle \\ \times \sum_{m_l = m_j \pm \frac{1}{2}} \langle l, \frac{1}{2}, m_l, m_j - m_l | j, m_j \rangle \langle l', \frac{1}{2}, m_l, m_j - m_l | j', m_j \rangle \langle l, m_l | \cos \theta | l', m_l \rangle \end{aligned} \quad (5.23)$$

The first two terms after the summation notation are the Clebsch-Gordan coefficients. The $\langle l, m_l | \cos \theta | l', m_l \rangle$ term is angular integral term. It is written as

$$\langle l, m_l | \cos \theta | l - 1, m_l \rangle = \sqrt{\frac{l^2 - m_l^2}{(2l + 1)(2l - 1)}} \quad (5.24a)$$

$$\langle l, m_l | \cos \theta | l + 1, m_l \rangle = \sqrt{\frac{(l + 1)^2 - m_l^2}{(2l + 3)(2l + 1)}} \quad (5.24b)$$

The eigenvalues obtained as a result of diagonalization the Matrix 5.22 are the new energies of the corresponding quantum states due to an electric field E' . To map out the shift of energies within an electric field range, the diagonalization is performed successively for each value of electric field within this range.

To check the accuracy of our simulation, we compare our Stark maps for rubidium atom to the paper by Zimmerman, M. L., *et al.* [129]. For $|m_j| = 1/2, 3/2$ and $5/2$, the simulated Stark maps within the energy range $[-540, -440] \text{ cm}^{-1}$ and electric field range of $[0, 6000] \text{ V/cm}$ are shown in Fig. 5.3, 5.4 and 5.5 respectively. In these simulation, we consider the coupling of all quantum states between $n = 12$ to $n = 22$ where $\Delta n = \pm 5$ at $n = 17$.

The Rydberg EIT experiments discussed in Chap. 7 are performed with Rydberg $D_{5/2}$ state. The simulated Stark maps for the selected states $48D_{5/2}$ and $58D_{5/2}$ are shown in Fig. 5.6.

Figure 5.3: Stark map of rubidium, $|m_j| = 1/2$

Figure 5.4: Stark map of rubidium, $|m_j| = 3/2$

Figure 5.5: Stark map of rubidium, $|m_j| = 5/2$

Figure 5.6: a) Stark map for $48D_{5/2}$ state b) Stark map for $58D_{5/2}$ state. The y -axis is the energy referenced to the corresponding unperturbed state. The $|m_j|$ degeneracy are lifted in the presence of electric field. For $|m_j| = 1/2$, magnetic sublevel is shifted positively but turned to negative direction at certain electric field due to avoided crossing with adjacent quantum state of nearest l .

Chapter 6

Theory of adsorption

6.1 Introduction

There are two types of adsorption processes: chemical adsorption (chemisorption) and physical adsorption (physisorption). In the first and second section, I will discuss chemisorption and physisorption respectively. The following section will highlight the thermodynamics of adsorption phenomenon. In the Sec. 6.5, we compare chemisorption and physisorption. These four sections focus on the adsorption phenomena based on the theoretical and experimental works that have been carried out in the field of surface science.

6.2 Chemisorption

The interests in adsorption phenomena have grown since Langmuir and Kingdon discovered that caesium atoms adsorbed on tungsten decreases the work function of the substrate and desorb in the form of positive ions [135]. Since then, there are a growing number of experimental [136] and theoretical [137, 138] studies investigating the physical and chemical properties of different adsorbates-substrate systems [139]. The phenomenon first observed by Langmuir and Kingdon was chemisorption. In this case, the bond formed involves the charge transfer between the adsorbate and substrate which results in a chemical bond (ionic bond, covalent bond and metallic bond).

Depending on the ionization energy of the adatoms and the work function of substrate, the charge can be transferred from the adsorbate to the substrate or vice versa [140]. This phenomenon is illustrated in Fig. 6.1 where the ionization energy, I_E of the adatoms is higher or lower than the work function, ϕ of the substrate. For $I_E < \phi$, the electronic charges are transferred to the substrate, resulting in the formation of electropositive induced dipoles as shown in Fig. 6.1a. This effect is particularly dominant among alkali atoms (potassium, caesium, rubidium, etc.) as they have relatively low ionization energies compared to most metallic substrate [139]. Fig. 6.1b shows the opposite case where $I_E > \phi$. The electronic charges are transferred from the substrate to the adsorbates, turning the adsorbates to electronegative dipoles. This effect occurs mostly for adsorbates from group 16-17 (nitrogen, oxygen, chlorine, carbon monoxide, etc.) which are highly

Figure 6.1: a) $I_E < \phi$ case: Adsorbates are formed as a result of electronic charge transfer from the adsorbates to the substrate. b) $I_E > \phi$ case: Electronegative adsorbates are formed as a result of electronic charge transfer from the substrate to the adsorbates.

electronegative and have large Pauling scale. In both cases of $I_E < \phi$ and $I_E > \phi$, there is a change in the work function of the substrate. This work function change depends on the coverage which is defined as the ratio of the density of the adsorbates with respect to the substrate [139]. There is a decrease of the work function of the substrate for electropositive adsorbates (K/Pt(111) system [141], Cs/Ru(0001) system [142], Li/Mo(110) system [143], etc.) and an increase for electronegative adsorbates (N₂/W [144] system, O₂/W [144] system, Cl₂/Cu(001) [145] system, O₂/Pt [146] system, etc.) where both situations indicate two different directions of charges transfer.

To illustrate these two different phenomena, experimental results showing the work function change with respect to coverage are represented in Fig. 6.2. For potassium chemisorbed on platinum [139, 141] as shown in Fig. 6.2a, the clean substrate Pt(111) has an initial work function value of 5.9 eV prior to any adsorption. In the beginning of increasing coverage, the work function decreases almost linearly and comes to a minimum before it rises again towards a steady value. These features occur commonly for alkali metal/metal substrate system and also on semiconductor materials [139]. The linear and low coverage region is called charge-transfer region. Each adsorbate deposits approximately the same amount of electronic charges to the substrate which gives rise to the linear behaviour. This process continues until the repulsive dipole-dipole interactions among adsorbates come into play. This minimum in the Fig. 6.2a is explained by the minimization of the total energy due to repulsion among adsorbates. Some fraction of the electronic charges flow back from the Fermi level of the substrate to the adsorbate, leading to a depolarization effect with a reduction of the dipole moment of the adsorbates [138]. Furthermore, some experiments suggest that there is a phase transition at the minimum where the dense monolayer structure is nucleated (condensation) [148]. After the minimum, the work function rises towards the work function of the bulk alkali. For potassium, it is 2.3 eV. The region after the work function minimum is called metallic region as

Figure 6.2: a) Graph of work function change with respect to coverage for potassium chemisorbed on Pt(111). Pt(111) has an initial work function of 5.9 eV. It decreases linearly and comes to a minimum before it rises again towards the work function of the bulk potassium (2.3 eV). Graph adapted from the works by Kiskinova, M., *et al.* [139, 141] b) Graph of work function change with respect to coverage for chlorine chemisorbed on Fe(100). In this case, the work function rises in an approximate linear trend before coming to a saturated value. Graph adapted from the works by Hino, S., *et al.* [147]

the work function increases to a value characteristic of a metallic bulk sample [139, 149]. The minimum occurs at about half a monolayer while the increase is approaching the monolayer structure [150]. Overall, the work function value of the substrate and the work function value of the adsorbates serve as the asymptotic limits. Fig. 6.2b shows an example of an electronegative adsorbate, chlorine chemisorbed on Fe(100) [147]. In this case, the work function rises in an approximate linear trend before coming to a saturated value.

The relationship between the work function change and the dipole moment of adsorbates is described by the Topping model [151, 152]. According to this model, the work function changes from an initial value of ϕ_i to ϕ as

$$\phi = \phi_i - e \frac{p}{\epsilon_0} n_0 \quad (6.1)$$

where n_0 is the area density of the number of dipoles and p is the dipole moment of the adsorbates. With increasing coverage, additional adsorbates adsorbed on the surface exert an opposite directed dipole field on an existing dipole at a site i with an electric field, E_{ads} of the form

$$E_{ads} = -p \sum_{i,i \neq j} \frac{1}{r_{ij}^3} \quad (6.2)$$

This interaction leads to depolarization, and the adsorbate at site i changes its initial dipole moment from p_i to p . The magnitude of this change depends on its polarizability, α and its interaction with the dipole field E_{ads} from the neighbouring adsorbates. The

Figure 6.3: Graph of work function change with respect to coverage for lithium adsorbed on Mo(110) (dashed line). The fit (solid line) to the experimental result using Topping model shows $p_i = 0.56\text{\AA}$ and $\alpha = 10\text{\AA}^3$ (in CGS units). Graph adapted from the works by Kröger, J., *et al.* [143, 152]

resulting dipole moment is given by

$$p = p_i + \alpha E_{ads} \quad (6.3)$$

Substituting E_{ads} from Eq. 6.2 leads to p of the form

$$p = p_i \left(1 + \alpha \sum_{j,j \neq i} \frac{1}{r_{ij}^3} \right)^{-1} \quad (6.4)$$

We can simplify this equation by assuming the adsorbates form a hexagonal closed pack lattice structure.

$$\sum_{j,j \neq i} \frac{1}{r_{ij}^3} = 8.9n_0^{3/2} \quad (6.5)$$

By substituting Eq. 6.4 to Eq. 6.1, we can write ϕ in dependence of θ_{ads} as

$$\phi = \phi_i - e \frac{p_i}{\epsilon_0} \left[\frac{n_s \theta_{ads}}{1 + 8.9\alpha(n_s \theta_{ads})^{3/2}} \right] \quad (6.6)$$

where n_0 has been replaced by the density of the substrate atoms on the surface, n_s multiplied with the coverage of the adsorbates, θ_{ads} . In Fig. 6.3 the Topping model is fitted to the experimental data for lithium adsorbed on Mo(110), giving the initial dipole moment value, $p_i = 0.56\text{ e\AA}$ and the polarizability, $\alpha = 10\text{ \AA}^3$ (in CGS units) [143, 152].

Even though the theory based on the comparison between the ionization energy of the adsorbate and the work function of the substrate explains most of the observations in chemisorption process, there are some exceptions. For example, with adsorbates from the alkali earth metals group, calcium ($I_E = 6.09\text{ eV}$) and strontium ($I_E = 5.67\text{ eV}$) chemisorbed on tungsten ($\phi = 4.5\text{ eV}$) [153] and barium ($I_E = 5.2\text{ eV}$) chemisorbed on nickel ($\phi = 5.01\text{ eV}$) [154] turn out to lower the work function of the substrate even though

the adsorbates have higher ionization energy than the substrate. This phenomenon is explained by Gurney, R. W. [153]. For electronegative adsorbates, nitrogen ($I_E = 14.6$ eV) chemisorbed on most of the tungsten crystal structures ((210),(310),(611),(111),(311)) result in an increase of the work function except W(100) [155, 156, 157]. Theoretical calculations using density functional theory (DFT) by Michaelides, *et al.* suggest that a transfer of electronic charges to nitrogen adsorbates is consistent with a decrease of the work function of W(100), attributing the observation to the overspill of electrons to the vacuum [158].

In general, the coverage dependency of work function of the substrate can help studying the behaviour of adsorption phenomena as well as the adspecies interaction [136, 138, 139, 159, 141].

6.3 Physisorption

Physisorption is an adsorption phenomenon where the bonding between the adsorbate and the substrate is mediated by the van der Waals interaction [160]. A neutral atom can have a temporary fluctuating dipole moment p_1 from its fluctuating electronic charge due to quantum and thermal fluctuations [161]. This fluctuating dipole moment may generate a fluctuating electromagnetic field which renders an adjacent atom of distance r an induced fluctuating dipole which is related to p_1 as $p_2 \propto -p_1/r^3$ [162]. Van der Waals force is an attractive force that exists between the fluctuating dipole and the resultant induced fluctuating dipole. The interaction energy is long range and scales with $-1/r^6$ [163]. At shorter distance, Pauli repulsion between the closed shell electrons of the two atoms prevents them from approaching each other [164]. The potential obtained from the summation of the attractive van der Waals potential and the repulsive potential gives the Lennard-Jones potential shown as

$$V(r) = \epsilon \left[\left(\frac{r_m}{r} \right)^{12} - 2 \left(\frac{r_m}{r} \right)^6 \right] \quad (6.7)$$

where ϵ is the trap depth and r_m is the distance where the potential $V(r_m)$ is minimum. However, as illustrated in Fig. 6.4a, an atom at position $\mathbf{r}$ physisorbed on the substrate involves the interaction between its fluctuating dipole moment and the induced dipole moment of all atoms at different positions $\mathbf{r}_{\mathbf{n},\mathbf{i}}$ inside the substrate [152]. To add up all the potential using Eq. 6.7,

$$V(\mathbf{r}) = \epsilon \sum_{\mathbf{n},\mathbf{i}} \left[\left(\frac{r_m}{|\mathbf{r} - \mathbf{r}_{\mathbf{n},\mathbf{i}}|} \right)^{12} - 2 \left(\frac{r_m}{|\mathbf{r} - \mathbf{r}_{\mathbf{n},\mathbf{i}}|} \right)^6 \right] \quad (6.8)$$

where the equation is summed over all the unit cell $\mathbf{n}$ and all the atoms inside the unit cell denoted by $\mathbf{i}$. Working in cylindrical coordinate by substituting $|\mathbf{r} - \mathbf{r}_{\mathbf{n},\mathbf{i}}|^2 = (z + z')^2 + r'^2$ with z as an atom to surface distance, turning the sum into a double integral, and introducing N as number of atoms inside the substrate per unit volume, Eq. 6.8 becomes

$$V(z) = \epsilon \int_0^\infty \int_0^\infty \left\{ \frac{r_m^{12}}{[(z + z')^2 + r'^2]^6} - \frac{2r_m^6}{[(z + z')^2 + r'^2]^3} \right\} 2\pi r' N dr' dz' \quad (6.9)$$

Figure 6.4: a) A schematic diagram illustrating an atom with its temporary fluctuating dipole moment at position $\mathbf{r}$ interacting with an induced dipole moment at position $\mathbf{r}_{n,i}$ inside the substrate. b) A graph showing the short range repulsive term (red), long range attractive van der Waals term (red), and the total potential term (blue) in Eq. 6.10.

which solution is

$$V(z) = \frac{\pi N \epsilon}{3} \left\{ \frac{r_m^{12}}{15z^9} - \frac{r_m^6}{z^3} \right\} \quad (6.10)$$

It is clearly seen from Eq. 6.10 that the van der Waals attractive term in Leannard-Jones potential 6.7 has become proportional to $-1/z^3$ for physisorption.

However, Eq. 6.10 is obtained from pair-wise summation by assuming the atom-substrate interaction is additive by largely ignoring the interactions among the atoms inside the substrate. To account for this oversimplifying assumption in physisorbed system, a van der Waals potential of the form

$$V(z) = -\frac{C}{(z - z_0)^3} \quad (6.11)$$

is used. C coefficient is calculated considering the van der Waals interactions for atoms inside the substrate by Lifshitz, E. M. [165] and Mavroyannis, C [166]. $V(z)$ is typically varied in the range of $\text{\AA}$ so it is important also to know the position of the reference plane to obtain a reliable evaluation of the van der Waals potential. z_0 is the reference plane and is derived by Zaremba, E., *et al.* [167] for atoms interacting through dipole-dipole interactions inside a jellium metal and an insulating crystal.

The repulsive potential is calculated by Zaremba, E., *et al.* [168] based on Hartree-Fock approximation in the lowest order of the wave functions overlap between the atom and the metallic substrate. Combined with his previous works [167] of van der Waals interaction, Zaremba, E., *et al.* applied his theoretical studies to calculate the binding energies and equilibrium positions of a helium atom [168]. Shown in Fig. 6.5 are the results from the works by Zaremba, E., *et al.* [168, 167]. Table shown in Fig. 6.5a lists the work function (ϕ), van der Waals coefficient (C), binding energies ($-E_0$), position of the reference plane (Z_0), zero-point energy, equilibrium position ($\langle Z_0 \rangle$), and the first excitation energy ($E_{10} = E_1 - E_0$) for ^3He and ^4He calculated based on the metallic density of the substrate $n_s = (4/3\pi r_s^3)^{-1}$ as an input parameters. The C coefficient and equilibrium position $\langle Z_0 \rangle$ for ^4He as a function of related metallic density parameter, r_s are shown in Fig. 6.5b. Shown in Fig. 6.5c is the charge density distribution as a result

a)

Metal	r_s	Φ^a (eV)	C^b (eV Å ³)	Z_0^b (Å)	$-E_0$ (meV) (K)		Zero-point energy (meV) (K)		$\langle Z \rangle_0$ (Å)		ΔE_{10} (meV) (K)	
					³ He	⁴ He	³ He	⁴ He	³ He	⁴ He	³ He	⁴ He
Al	2.07	4.19	0.202	0.544	3.44(40.0)	3.65(42.4)	1.76(20.4)	1.55(18.0)	3.79	3.73	2.16(25.0)	2.05(23.8)
Mg	2.65	3.66	0.153	0.478	1.65(19.1)	1.78(20.6)	1.12(13.0)	0.99(11.5)	4.27	4.18	1.19(13.8)	1.17(13.5)
Li	3.28	3.1	0.117	0.428	0.77(8.9)	0.85(9.8)	0.70(8.1)	0.62(7.2)	4.91	4.77	0.63(7.3)	0.64(7.4)
Na	3.99	2.7	0.092	0.388	0.41(4.7)	0.46(5.3)	0.49(5.7)	0.44(5.1)	5.51	5.31	0.36(4.2)	0.38(4.4)
K	4.96	2.39	0.070	0.356	0.20(2.4)	0.24(2.7)	0.34(4.0)	0.30(3.5)	6.23	5.95	0.19(2.2)	0.21(2.5)
Rb	5.23	2.21	0.065	0.351	0.15(1.8)	0.18(2.1)	0.28(3.3)	0.25(2.9)	6.64	6.30	0.15(1.7)	0.17(1.9)
Cs	5.63	2.14	0.058	0.345	0.13(1.5)	0.15(1.8)	0.25(2.8)	0.23(2.6)	6.84	6.47	0.12(1.4)	0.14(1.6)
Cu	2.67	4.65	0.225	0.220	3.34(38.8)	3.55(41.2)	1.74(20.2)	1.53(17.8)	3.66	3.59	2.08(24.2)	1.98(23.0)
Ag	3.02	4.0	0.249	0.196	3.34(38.7)	3.53(41.0)	1.61(18.7)	1.42(16.5)	3.73	3.67	2.01(23.4)	1.90(22.1)
Au	3.01	5.22	0.274	0.155	5.61(65.1)	5.91(68.6)	2.39(27.7)	2.09(24.3)	3.23	3.17	3.13(36.3)	2.92(34.0)

b)

c)

Figure 6.5: a) Table lists the relevant parameters as a function of r_s b) Graph showing C coefficient and $\langle Z_0 \rangle$ as a function of r_s c) Figure of the charge density distribution as a result of a ^4He atom adsorbed on the substrate (solid line) and the charge density of a clean substrate. All the graphs are adapted from the works by Zaremba, E., *et al.* [168, 167]

of a helium atom adsorbed on the metallic substrate (solid line) while the unperturbed metallic density is shown by the dashed line.

As shown by Fig. 6.5c, due to the redistributed charges on the surface, Zaremba, E., *et al.* [168] found that there is an induced dipole moment directed inward for a helium atom adsorbed on the substrate, leading to an increase of the work function. Similar theoretical results have been found for the orientation of the dipole moment of hydrogen physisorbed on a perfectly charge imaging semi-infinite metal, a dipole moment directed inward at equilibrium position of the physisorbed potential [169]. However, the dipole moments of Xe atoms physisorbed on Pd(100) are found to be directed outward, which is shown experimentally by the decrease of the work function of the substrate with increasing coverage [170]. Works by Zaremba, E., *et al.* suggest the direction of the dipole moments formed is different for light and heavy atoms because of the balancing between the repulsive potential and attractive van der Waals potential [168, 169]

6.4 Thermodynamics of adsorption and desorption

In this section, we discuss the formulation of thermodynamic equilibrium achieved between the gas phase and the adsorbate phase from the perspective of quantum statistics. The thermodynamic equilibrium condition occurs when the chemical potential of the gas phase is equal to the chemical potential of the adsorbate phase. Chemical potential for an ideal gas phase and adsorbate phase are quoted from standard statistical mechanics textbook sources [171, 152, 172]. These equations lead to the derivation of the Langmuir equation which describe the thermodynamics of the adsorption.

For an ideal non-interacting gas molecules at temperature T and pressure P [152, 171], its chemical potential μ_{gas} can be written as

$$\mu_{gas} = k_B T \ln \left[\frac{P}{k_B T} \left(\frac{h^2}{2\pi m k_B T} \right)^{3/2} \frac{1}{Z_{rot} Z_{vib}} \right] + E_g \quad (6.12)$$

where Z_{rot} and Z_{vib} are the rotational and vibrational partition function. E_g is the ground state energy of the gases.

The behaviour for adsorbates on the substrate can be modelled with an array of ordered surface sites where the occupation number on each site with an adsorbate can be either 0 or 1. It is assumed the adsorption energy on each single site is unaffected by the presence or absence of an adsorbate on the neighboring sites. This model is called non-interacting lattice gas model. It can describe chemisorption since the adsorbates form a periodic structure given by the periodicity of the crystal surface. This situation is similar to Fermi-Dirac statistics for fermions which allows us to write the mean occupation number per site $\langle n \rangle$ using Fermi-Dirac distribution as

$$\langle n \rangle \equiv \theta_{ads} = \frac{1}{1 + e^{\frac{E_{ads} - \mu_{ads}}{k_B T}}} \quad (6.13)$$

$\langle n \rangle$ is equivalent to the coverage θ_{ads} . E_{ads} and μ_{ads} refer to the energy of the adsorbates and its chemical potential. By rearranging Eq. 6.13 we obtain the chemical potential as a function of coverage. Adding the vibrational partition function to account for vibrational degree of freedom of the adsorbates, the expression for μ_{ads} can be written as

$$\mu_{ads} = E_{ads} + k_B T \ln \left(\frac{\theta_{ads}}{1 - \theta_{ads}} \right) - k_B T \ln Z_{vib,ad} \quad (6.14)$$

Equilibrium between the gas phase and the adsorbate phase is established when the chemical potentials of the two phases are equal. Thus, equating Eq. 6.12 and Eq. 6.14 results in

$$\left(\frac{\theta_{ads}}{1 - \theta_{ads}} \right)^\alpha = e^{\frac{E_g - \alpha E_{ads}}{k_B T}} \frac{\hbar^3}{k_B T (2\pi m k_B T)^{3/2}} \frac{Z_{vib,ad}^\alpha}{Z_v Z_r} P \quad (6.15)$$

where α is introduced to account for dissociative adsorption. α is the number of individual atoms the molecule is separated into upon adsorption on the substrate. For $\alpha = 1$, Eq. 6.15 becomes the Langmuir equation. For the case of maintaining the temperature of the substrate but with the pressure varied, the graph is called Langmuir isotherm. On the other hand, if the pressure is kept constant but with the temperature varied, the graph

Figure 6.6: With $\Delta E_{ads} = 2$ eV a) Plots of Langmuir isotherms showing θ_{ads} as a function of P for different T_{subs} . b) Plots of Langmuir isobars showing θ_{ads} as a function of T_{subs} for different P .

obtained is called Langmuir isobar. Adsorption energy is defined as the subtraction between the two terms E_g and E_{ads} , that is $\Delta E_{ads} = E_g - \alpha E_{ads}$.

It is important to know that experiments are usually performed by varying the temperature of the substrate placed inside a vacuum system. However, the molecules in the gas phase are in contact with the walls of the vacuum chamber which is at room temperature. This suggests that the complete thermodynamic equilibrium condition is not achieved as required by Eq. 6.15. Only the temperature (T in the exponential term of Eq. 6.15) of the substrate is varied but the temperature (T in the denominator of the second term of Eq. 6.15) of the gas phase is maintained at room temperature. Eq. 6.15 is rewritten to replace the corresponding terms T to T_{gas} and T_{subs} to show the temperature of the gas and substrate respectively, assuming $\alpha = 1$ and negligible vibration and rotational energy

$$\frac{\theta_{ads}}{1 - \theta_{ads}} = e^{\frac{\Delta E_{ads}}{k_B T_{subs}}} \frac{\hbar^3}{k_B T_{gas} (2\pi m k_B T_{gas})^{3/2}} P \quad (6.16)$$

Shown in Fig. 6.6, Eq. 6.16 is plotted for the Langmuir isotherm and the Langmuir isobar case respectively with $\Delta E_{ads} = 2$ eV.

In the field of surface science, two techniques are typically used to obtain isosteric (constant coverage) heat of adsorption $Q(\theta_{ads})$. One technique is to apply Clausius-Clapeyron equation to Langmuir isobars and Langmuir isotherms. Another technique is called thermal desorption spectroscopy (TDS). From Langmuir isotherm and isobar curves, $Q(\theta_{ads})$ is determined using Clausius-Clapeyron equation shown as

$$Q(\theta_{ads}) = - \left. \frac{\partial(\ln P)}{\partial(1/k_B T)} \right|_{\theta_{ads}} \quad (6.17)$$

where $Q(\theta_{ads})$ is obtained from the gradient of the plot of $\ln P$ v.s. $1/k_B T$ at a particular θ_{ads} . $Q(\theta_{ads})$ is not exactly equal to ΔE_{ads} but has an extra contributing term from

Figure 6.7: Graph showing the isobar curves for the adsorption of CO on Ni(111) at different pressure. Langmuir isobar is added to the experimental isobar curves to illustrate the discrepancies between the experiments and the theory. Graph adapted from the works by Ibach, H., *et al.* [172, 152]

energy due to temperature,

$$Q(\theta_{ads}) = \Delta E_{ads} + \frac{5}{2}k_B T \quad (6.18)$$

where $\Delta E_{ads} = E_g - E_{ads}$ for non-dissociative adsorption.

TDS is an experimental technique that measures the desorption rate as the adsorbates overcome its binding energies when the temperature of the substrate is increased. The desorption rate is measured by a pressure gauge since the desorbed atoms give rise to a sudden increase of vacuum pressure. Thus, TDS is a measurement technique to determine desorption energy, E_d , the activation energies for adsorbates during the desorption process. Usually, the adsorption process does not require an activation energy [152], so activation energies obtained from TDS technique approximate the heat of adsorption, $Q(\theta_{ads})$. In an adsorption process where the adsorption energy is on the scale of $\sim \text{eV}$ (for example, chemisorption), $Q(\theta_{ads}) \approx \Delta E_{ads}$.

Shown in Fig. 6.7 are the isobar curves for adsorption of CO on Ni(111). There are some discrepancies between the experimental results and the Langmuir isobar curve. At ~ 325 K, binding energies are weaker and this results in an early desorption of adsorbates. The Langmuir isobar predicts the desorption at higher temperatures. At decreasing coverage, the binding energies are getting stronger and some adsorbates remain on the surface even though the Langmuir isobar predicts the adsorbates should have depleted at 400 K. The Langmuir equation is only applicable to adsorption and desorption phenomena of a monolayer structure, without taking into account the interactions among

the adsorbates, thus neglecting the coverage dependence of binding energies. Different models are built to account for the interactions among adsorbates. The Bragg-Williams model adds an extra coverage dependent term in Eq. 6.14 to describe coverage dependence change in adsorption energy [152]. For physisorption model, Brunauer-Emmett-Teller (BET) theory was developed by Brunauer, S., *et al.* [173] to discriminate the different binding energies at different layers of adsorbates.

6.4.1 Relationship between binding energy, adsorption energy and heat of adsorption

Even though binding energy, ϵ , adsorption energy, ΔE_{ads} and heat of adsorption, $Q(\theta_{ads})$ refer to the energy released as a result of bonds formed between the adsorbates and substrate, they refer to different scenarios [174].

To illustrate this description, an example of a curve (yellow) shown in Fig. 6.6b for $P = 1 \times 10^{-10}$ mbar and $\Delta E_{ads} = 2$ eV is used. From this curve, it is estimated the desorption occurs at about 500 K. A direct conversion of 500 K to thermal energy, $\frac{5}{2}k_B T$ corresponds to 108 meV which is far lower than ΔE_{ads} of 2 eV. This example shows that the adsorption energy and binding energy are describing different properties.

Adsorption energy, ΔE_{ads} refers to the difference between the total energy of the adsorbate system and the summation of the energies of the gas phase molecules and clean substrate. In an alternative perspective, ΔE_{ads} can be said to consist of the summation of two energy terms. The binding energy of purely adsorbate substrate interacting system are added to the interaction energy (E_{int}) for the interaction among the adsorbates themselves. Thus, binding energy can be computed as the difference between ΔE_{ads} and E_{int} [174].

However, in TDS experiments and measurements using Clausius-Clapeyron equation, we determine the desorption energy, E_d or the heat of adsorption, $Q(\theta_{ads})$ [174]. This energy is associated with a kinetic process but ϵ is a thermodynamic quantity at 0 K [174]. Shown by Eq. 6.18 is the relationship between $Q(\theta_{ads})$ and ΔE_{ads} . In the limit of zero coverage (one adsorbate), $Q(\theta_{ads} \rightarrow 0) = \epsilon$, $Q(\theta_{ads})$ approaches the binding energy of a single adsorbate-substrate system.

6.5 Chemisorption and physisorption

To directly compare chemisorption and physisorption, a list of the relevant properties of the two processes is tabulated. Table 6.1 shows the properties of heat of adsorption, bonding type, bonding strength, reversibility, layer structure, adsorption site specificity, surface reaction, temperature regime, pressure dependency and modelling [175, 176].

Property	Chemisorption	Physisorption
Heat of adsorption	High ($\sim$ eV)	Low ($\sim$ hundreds of meV)
Bonding type	Chemical bond (ionic, covalent, metallic)	Van der Waals bond
Bonding strength	Strong (short range)	Weak (long range)
Reversibility	Irreversible (original species non-recoverable upon desorption)	Reversible (original species recoverable upon desorption)
Mobility	Low immobility and do not diffuse	High mobility and easily diffuse
Layer structure	Monolayer	Multilayers
Adsorption site specificity	Highly specific on the structure of crystal surface	Not very specific on the structure of crystal surface
Surface reaction / reconstruction	Dissociative adsorption, condensation and surface reconstruction	No surface reaction
Temperature regime	Can occur at high temperature	Usually takes place at low temperature and adsorption decreases with increasing temperature
Pressure dependency	Higher pressure increases adsorption rate, lowering of pressure does not cause desorption. No more adsorption once all sites are filled.	Higher pressure increases adsorption rate and the number of adsorbates, lowering of pressure causes desorption.
Modelling	Langmuir equation	BET theory

Table 6.1: Comparison of the properties between chemisorption and physisorption [175, 176].

Chapter 7

Experimental investigation of adsorption phenomena

7.1 Introduction

We investigate the electric field produced by adsorbates by employing Rydberg spectroscopy. Sec. 7.2 explains our optical setup used for the Rydberg spectroscopy experiment. Subsec. 7.2.1 and 7.2.2 describes the 780 nm and 480 nm laser system which serve as a probe and coupling beam respectively. Subsec. 7.2.3 shows the locking scheme based on counter-propagating probe and coupling beam. Subsec. 7.2.4 explains the calibration of the frequency of the 480 nm laser beam. In Sec. 7.3, the experimental procedures for the Rydberg spectroscopy experiment are explained including the acquisition of a Rydberg spectrum and its post-processing. Sec. 7.4 introduces a simple model to fit the spatial profile of the electric field outside a layer of adsorbates. Sec. 7.5 and 7.6 discuss the Rydberg spectroscopy experiments performed at room temperature and cryogenic temperature respectively. Shown in these sections are the spectra together with the theoretical fits based on the model discussed in Sec. 7.4. In order to identify the atomic species of the adsorbates at cryogenic temperature, a few experiments are performed and the details are explained in Sec. 7.6. Sources for electric fields are attributed to adsorbates due to chemisorption at room temperature and physisorption at cryogenic temperature. We experimentally investigate the temperature dependency of the adsorbates. The explanation of the results are based on the Langmuir isobar equation as discussed in Sec. 7.7.

7.2 Rydberg spectroscopy experimental setups

As shown in Fig. 7.1, the experimental setup consists of two parts: 780 nm (orange box) and 480 nm (red box) laser systems (including EIT locking setups). The 780 nm laser system is frequency stabilized to the D2 transition of ^{87}Rb . The 480 nm laser system is stabilized using EIT locking.

7.2.1 780 nm laser system

The laser used in the 780 nm laser system is an external cavity diode laser (ECDL). There are different configurations like Littman or Littrow [177]. ECDL has an advantage of frequency selection and tunability using diffraction grating. It can also narrow down the linewidth from $\sim$ GHz to a few hundreds kHz. This is necessary for our application as the natural linewidth of ^{87}Rb is 6 MHz. The tunability behaviour allows us to probe the different hyperfine transition of the D2 line. In order to have greater stability, our ECDL is based on the design by Gilowski, *et al.* [178]. This design uses interference filter for frequency selection rather than diffraction grating.

We lock the ECDL via saturated absorption spectroscopy (SAS) to the crossover peak, $|F' = 2, 3\rangle$ of $5P_{3/2}$. First, the light passes through an optical isolator (OI) to prevent any optical back reflection into the diode. A half-wave plate splits laser beam at a polarizing beam splitter (PBS). The transmitted beam is coupled to the fibre for EIT locking while the reflected beam is directed to a rubidium vapour cell (Thorlabs GC25075-RB). This beam passes through the vapour cell and is retro-reflected by a mirror. A quarter waveplate is placed on the path such that the retro-reflected beam has orthogonal polarization and is transmitted through the same PBS onto a photodiode (PD).

The photodiode signal is processed using a PI and a lock-in setup. We modulate the current of the ECDL through the laser diode driver (LDD). Our lock-in amplifier has an internal signal generator which can generate a frequency of $f \sim 80$ kHz. f from lock-in 1 is divided with a frequency divider (FD) to $f/3$. The resultant signal of $f/3$ is attenuated with a 30 dB attenuator before being directed to the LDD for current modulation. The detected absorption signal from PD is mixed with a reference frequency of f from the same signal generator inside lock-in 1 to produce an error signal. This error signal is the 3rd derivative of the ^{87}Rb spectrum. Since the derivative of a peak is zero, the error signal obtained allows us to lock at the peak, the centre of an atomic transition. The locking is done with proportional integrate (PI) controller. The error signal is subject to proportional scaling and integration before being feedback to the piezoelectric transducer (PZT) of the ECDL. For our application, we lock the 780 nm laser system to the crossover peak, $|F' = 2, 3\rangle$ of $5P_{3/2}$.

7.2.2 480 nm laser system

Our 480 nm laser system is purchased from LEOS Solutions. It consists of a 960 nm laser system (from Sacher Lasertechnik Company) and a second harmonic generation (SHG) (LEOS Solutions).

The 960 nm laser system has two main components. It has a tunable ECDL in Littman configuration and a tapered amplifier (TA) system. Coarse tuning of the wavelength can be varied by adjusting the cavity length, that is translating and rotating the mirror inside the ECDL. In addition, tuning of the wavelength can also be adjusted by varying the temperature of the ECDL. At last, fine tuning of the wavelength can be done by adjusting the current supplying the laser diode and PZT determining the length of the external cavity. Characteristic for the TA is the tapered shape gain medium. Both front and rear facets are antireflection (AR) coated to decrease the reflectivity typically below 10^{-4} [179]. Laser light from ECDL is first passed through an OI. After the OI, two

mirrors are used to steer the laser beam to the TA. An aspheric lens is used to focus the beam to the TA. Another aspheric lens together with a cylindrical lens (CL) are used to collimate the output beam. The collimated beam is then passing through another OI to prevent any back reflection that might damage the TA.

This 960 nm laser system is able to produce an output laser power of ~ 1.5 W for SHG. This high power laser beam goes through a half-wave plate before being directed to the bow-tie cavity using a mirror. A lens is used to focus the laser beam to a non-linear crystal inside the cavity. The crystal used is lithium triborate (LBO). The bow-tie cavity has 4 high reflectivity mirrors at 960 nm but low reflectivity at 480 nm. This allows the frequency doubled beam of 480 nm gets transmitted through the mirror but the 960 nm beam circulates in the cavity. The bow-tie cavity is locked using Hansch-Couillaud method (not drawn) [180]. The output 480 nm beam from the cavity is to be collimated with two cylindrical lenses. When the laser system was first delivered by the company, the total output power from the cavity had a maximum of 400 mW. However, it degraded fast over time and now we only obtain ~ 140 mW after careful optimization.

7.2.3 EIT locking

The frequency locked 780 nm laser system will be used to EIT lock the 480 nm laser system. EIT locking is done by overlapping the 780 nm and the counter-propagating 480 nm laser beam. The 780 nm laser serves as a probe beam while the 480 nm laser beam serves as a coupling beam. First, the 780 nm laser beam is transferred to the EIT locking setup through a fibre. Then, it passes through a half-wave plate and a PBS. This is to control the power of the probe beam as high power will broaden the EIT signal. Two mirrors are used to steer the probe beam to the rubidium vapour cell (Photonics Technologies SC-RB-75-Q-BA) which has Brewster angled windows. On the other side, the output beam from 480 nm laser system is also passed through a half-wave plate and a PBS before being directed to the vapour cell using a mirror. The coupling beam passes through a dichroic mirror (DM1) which is transmitting 480 nm but reflecting 780 nm. The probe beam also passes through a dichroic mirror (DM2) but transmitting 780 nm and reflecting 480 nm. To make the overlapping between the two beams easy, two irises are used. Two lenses are used to focus the probe and coupling beams inside the vapour cell. The focused coupling beam increases the Rabi frequency and leads to a stronger EIT signal. The 480 nm laser power decreases quite appreciably after passing through several optical elements. The probe beam is then reflected by DM1 and detected by a photodiode (PD).

If the probe beam is on resonance to the D2 transition of ^{87}Rb , the scan of the coupling beam with PZT will show a dip in the absorption signal, indicating a transparency of the medium. Fig. 7.2a shows two dips (blue line) with the scan of the coupling beam. These two dips correspond to the $42D_{3/2}$ (0 MHz) and $42D_{5/2}$ (159.2 MHz) Rydberg state. Eq. 7.1 which is obtained from the summation of two gaussian functions are fitted (red line) to the experimental curve.

$$g(x) = a_0 + a_1 \exp - \left(\frac{x - x_{c1}}{\delta} \right)^2 + a_2 \exp - \left(\frac{x - x_{c2}}{\delta} \right)^2 \quad (7.1)$$

Figure 7.2: a) Absorption signal (blue line) with fitting from the summation of two gaussian functions (red line). Two EIT dips seen correspond to $42D_{3/2}$ (0 MHz) and $42D_{5/2}$ (159.2 MHz). Resulting fit gives a linewidth of 13.6 MHz. b) Error signal (blue dots) with fitting from the 3rd derivative of the summation of two gaussian functions (red line).

x_{c1} and x_{c2} are given by $42D_{3/2}$ (0 MHz) and $42D_{5/2}$ (159.2 MHz). The resulting fit gives a power broadened linewidth of $\delta = 13.6$ MHz.

The detected absorption signal serves as the input to the second lock-in amplifier (lock-in 2). The reference signal is the current modulation of the probe laser. These two signals are multiplied using a mixer inside lock-in 2 to produce an error signal. Fig. 7.2b shows this error signal (blue dots). As expected, even though the input signal shown in Fig. 7.2a is noisy, the lock-in amplifier is able to extract a clean signal. The error signal is fitted with the 3rd derivative of Eq. 7.1 (red line). This fit to the error signal finds reasonable agreement, thus showing the expected functioning behaviour of the lock-in amplifier. The 480 nm laser system is locked with a PI controller on the error signal from lock-in 2.

After locking to a particular Rydberg state, the 480 nm laser is then frequency shifted by an AOM to vary the coupling beam detuning for the Rydberg spectroscopy. The last lens pair is collimating the beam. After all optical elements, the maximum power of the coupling laser is ~ 40 mW.

7.2.4 Calibrating detuning of coupling beam

For the EIT locking, the rubidium vapour cell used is at room temperature. Thus, the Doppler effect has to be considered for the calibration of the coupling beam detuning for Rydberg spectroscopy experiment. As mentioned in Sec. 7.2.1, the 780 nm laser system is locked to the crossover peak, $|F' = 2, 3\rangle$ of $5P_{3/2}$. It is on resonance with rubidium atoms in the velocity class $\mathbf{v}$.

$$f_{780} + 133 \text{ MHz} = f_{780 \text{ lab}}(1 + v/c) \quad (7.2)$$

A + sign for the velocity component v is used to denote the opposite direction of the probe beam. These atoms see a Doppler shifted frequency of $f_{780 \text{ lab}} + 133 \text{ MHz}$ on resonance with the transition to $|F' = 3\rangle$. $f_{780 \text{ lab}}$ is the frequency from the ground state to the crossover peak in the lab frame. We can write v in the form

$$v = 133 \text{ MHz} \frac{c}{f_{780}} \quad (7.3)$$

To fulfil the EIT resonance condition, the coupling beam has to be on resonance with the same velocity class. The Doppler shifted frequency in the frame of atoms, $f_{480 \text{ atom}}$ for the coupling beam is written as

$$f_{480 \text{ atom}} = f_{480 \text{ lab}}(1 - v/c) \quad (7.4)$$

where $f_{480 \text{ lab}}$ is the frequency of the coupling beam in the lab frame. Substituting v from Eq. 7.3 to Eq. 7.4 gives

$$f_{480} = f_{480 \text{ lab}} - 133 \text{ MHz} \frac{f_{480 \text{ lab}}}{f_{780}} \quad (7.5)$$

The last term in Eq. 7.5 can be approximately converted to

$$- 133 \text{ MHz} \frac{f_{480 \text{ lab}}}{f_{780}} \approx -133 \text{ MHz} \frac{780 \text{ nm}}{480 \text{ nm}} = -216 \text{ MHz} \quad (7.6)$$

For the Rydberg spectroscopy on an ultracold ensemble, the Doppler effect is minimal. Thus, we have to shift the frequency by -216 MHz with an AOM shown in Fig. 7.1 in order to be on resonance.

Shown in Fig. 7.3b, in order to achieve $\delta_c = 0 \text{ MHz}$ in the Rydberg spectroscopy experiment, the EIT locking probe laser which is on resonance to the crossover peak, $|F' = 2, 3\rangle$ of $5P_{3/2}$, the coupling beam has to be shifted by -216 MHz . However, this correspondence may vary by a few MHz due to an internal small offset by the lock-in amplifier which affects the locking position. The reference position of $\delta_c = 0 \text{ MHz}$ in a Rydberg spectroscopy spectrum can be determined more precisely by using the simulated Stark map discussed in Sec. 5.4.

7.3 Rydberg spectroscopy experimental procedures

Fig. 7.3a shows the experimental setup for the Rydberg spectroscopy experiment. It shows the 780 nm probe beam and the counter-propagating 480 nm coupling beam. The 780 nm probe beam is the existing imaging beam in our absorption imaging setup. The coupling beam is from the laser system discussed in Sec. 7.2. In order to introduce the coupling beam to the absorption imaging setup, a dichroic mirror is placed in front of the camera. This dichroic mirror is transparent for 780 nm but reflective for 480 nm. Mirrors (not shown) are used to steer and overlap the coupling beam with the probe beam. In addition, a cylindrical lens (75 mm focus length) is located such that a line focus along z -direction is obtained at the position of the atomic cloud on the atom chip. This is necessary to probe the spatial dependence of the electric field along z -direction and is

Figure 7.3: a) Schematic diagram of the experimental setup for the Rydberg spectroscopy experiment. [Zoom in] Illustration of the electric field emitted by the adsorbates adsorbed on the YBCO square structure. Shown also is the absorption image of the atomic cloud and its EIT transparency when the coupling beam is present. b) Energy level diagram of ^{87}Rb . Probe beam couples $|5S_{1/2}, F = 2\rangle$ to $|5P_{3/2}, F' = 3\rangle$, while the coupling beam couples the intermediate state to an $nD_{5/2}$ Rydberg state.

perpendicular to the YBCO square structure. A zoom in picture of the atomic cloud is shown on the right. This picture illustrates the electric field emitted by the adsorbates chemisorbed on the YBCO substrate. Shown also is the optical depth image of the atomic cloud located below the YBCO square. Fig. 7.3b is the energy level diagram for ^{87}Rb with the probe beam couples the ground state $|5S_{1/2}, F = 2\rangle$ to the intermediate state $|5P_{3/2}, F' = 3\rangle$. Coupling beam with a detuning of δ_c couples the intermediate state to an $nD_{5/2}$ Rydberg state.

Graph analysis

With an appropriate choice of δ_c , a transparency region can be seen from the optical depth image of an atomic cloud. Fig. 7.4 shows a few examples with coupling beam detunings of 31.0 MHz (Fig. 7.4a), -4.2 MHz (Fig. 7.4b) and -20.6 MHz (Fig. 7.4c). The arrows drawn in each image point to the transparency region. These pictures illustrate that for a particular detuning and linewidth of the coupling laser, a certain electric field shifts the energy level on resonance so the atoms in this region are transparent for the probe beam.

In order to show the data in a more meaningful representation and decouple it from atom number fluctuation, the results of optical depth image for various δ_c have to be analysed further. Taking Fig. 7.4d as an example, two column vectors A and B of this optical depth image are selected. A is chosen to be at the focus region of the coupling laser. B is chosen to be $-160 \mu\text{m}$ away from A, a distance which is outside of the transparency region. Column vector A is then divided by column vector B to produce the EIT ratio column vector C. The density of the atomic cloud is homogeneous between A and B, then each element inside C will have a value between 0 and 1. EIT ratio of 0 means full EIT while EIT ratio of 1 means no EIT. Potential error sources are unequal density distribution across the atomic cloud or atom number loss over the experimental runs.

Afterwards, column vectors C obtained for a successive range of δ_c are put in an increasing order. The result is shown in Fig. 7.4e. In this graphical representation,

Figure 7.4: Images showing the optical depth of atomic cloud for various chosen values of coupling beam detunings. Arrows are pointed to the transparency region. a) $\delta_c = 31.0 \text{ MHz}$ b) $\delta_c = -4.2 \text{ MHz}$ c) $\delta_c = -20.6 \text{ MHz}$ d) Optical depth image of an atomic cloud d) Spectrum before modification e) Spectrum after modification

y -axis is the distance from the substrate and x -axis is the coupling beam detuning. Fig. 7.4e is further manipulated by a few procedures. First, the matrix elements of this image are inverted and scaled proportionally between 0 and 1 to produce an optical density ratio (ODR) image rather than an EIT ratio image. Since the -1st order of the AOM blue beam is used to control the δ_c , the coupling beam detuning's sign has to be flipped. Additionally, an offset (if necessary) is also added to the x -axis. This is to account for an offset in error signal present in lock-in 2 of Fig. 7.1 which affects its locking position. This image is subject further to noise reduction by using an image processing technique called average filtering. The final spectrum obtained is shown in Fig. 7.4f.

7.4 Model for adsorbate electric field

Figure 7.5: a) Drawing showing the electric field produced by the adsorbates b) Drawing showing the electric field, $E_{dipole}(z)$ from a dipole layer is equal to the summation of the electric fields produced by two surface charge square sheets of opposite signs and with separation t .

In order to understand the behaviour of the spectrum acquired through Rydberg spectroscopy discussed later in Sec. 7.5 and 7.6, the characteristics of the adsorbates' layer producing an electric field have to be identified. To model the electric field from the adsorbates' layer shown in Fig. 7.5a, it is assumed they behave as the electric field originated from a homogeneous dipole layer square sheet shown in Fig. 7.5b. This dipole layer square sheet is producing a total electric field, $E_{dipole}(z)$ that can be calculated from the summation of two electric fields originating from two constant surface charge density square sheets of opposite signs (Fig. 7.5b).

We denote σ^+ and σ^- as the positive and negative surface charge density, size of square sheet as l , and separation as t . The electric field $E_{\sigma^+}(z)$ and $E_{\sigma^-}(z)$ above the centre of a constant surface charge density square sheet σ are given by

$$E_{\sigma^+}(z) = \frac{2\sigma}{\pi\epsilon_0} \tan^{-1} \sqrt{1 + \frac{l}{2z^2}} - \frac{\sigma}{\pi\epsilon_0} \quad (7.7a)$$

$$E_{\sigma^-}(z) = -\frac{2\sigma}{\pi\epsilon_0} \tan^{-1} \sqrt{1 + \frac{l}{2(z+t)^2}} + \frac{\sigma}{\pi\epsilon_0} \quad (7.7b)$$

which the total electric field above the centre of a dipole layer in Fig. 7.5 is simply the

summation of Eq. 7.7,

$$E_{dipole}(z) = E_{\sigma+}(z) + E_{\sigma-}(z) \quad (7.8)$$

After substituting Eq. 7.7 to Eq. 7.8, the result is

$$E_{dipole}(z) = \frac{2\rho}{\pi\epsilon_0(1\text{\AA})} \left[\tan^{-1} \sqrt{1 + \frac{l}{2z^2}} - \tan^{-1} \sqrt{1 + \frac{l}{2(z + 1\text{\AA})^2}} \right] \quad (7.9)$$

Since we are interested in dipole density ρ ($\rho = \sigma t$), we fix the value of t to $1\text{\AA}$ and vary ρ . In our applications, t is chosen as $1\text{\AA}$. ρ and l in Eq. 7.9 are optimized such that the resultant electric field $E_{dipole}(z)$ reproduces the spectrum obtained by the experiment.

7.5 Rydberg spectroscopy at room temperature

Rydberg spectroscopy using $52D_{5/2}$, $58D_{5/2}$ and $61D_{5/2}$ states are performed at room temperature and its spectra are shown in Fig. 7.6a, 7.6b and 7.6c respectively. The general features of these graphs are the presence of 3 lines which are getting further separated as the distance is getting closer to the substrate. These 3 lines are the $|m_j| = 5/2, 3/2$ and $1/2$ manifolds of their $nD_{5/2}$ state. These manifolds are labelled accordingly (white) in the graphs. Their degeneracies are lifted by the presence of an electric field. The increase splitting between the $|m_j|$ states closer to the surface suggests also the electric field is increasing.

To infer the dipole density, we first use the Stark map discussed in Sec. 5.4 for the spectra shown in Fig. 7.6a, 7.6b and 7.6c to determine the electric field distribution. The electric field distribution is shown in Fig. 7.6d. We fit Eq. 7.9 to the experimental electric field and obtain for the dipole density $\rho_{chem} = 3.7 \times 10^{-12} \text{ C m}^{-1}$, an offset of 0.14 V/cm and $l = 1 \text{ mm}$.

As shown by the fitting parameters, the electric field is produced by a layer of adsorbates of size $l = 1 \text{ mm}$, a value which is the same as the physical size of our YBCO square structure. Meaning the electric field originates from the YBCO square. Rubidium atoms are chemisorbed on this square. As discussed in Sec. 6.2, the strength of the chemisorbed dipole depends on the difference between the work function of the substrate and the ionization energy of the atom. The work function of YBCO (4.5 eV) [181] differs from the rubidium ionization energy (4.2 eV), giving rise to strong chemisorbed dipoles, while the work function of our YSZ substrate (4.34 eV) [182] is closer to the ionization energy of a rubidium atom. Still, chemisorption onto the YSZ substrate may be responsible for the small offset field of 0.14 V/cm . Chemisorption involves a chemical bond of typical binding strength in the range of $\sim \text{eV}$ and should be chemically stable at room temperature. Fig. 7.6a, 7.6b and 7.6c were obtained from experiments conducted at different days within one week. This observation also indicates that the chemisorbed layer responsible for the electric field was chemically stable for at least one week at room temperature.

Figure 7.6: Spectra obtained from Rydberg spectroscopy using a) $52D_{5/2}$, b) $58D_{5/2}$ and c) $61D_{5/2}$ Rydberg states at room temperature. Labels in white show the corresponding $|m_j| = 5/2, 3/2, 1/2$ manifolds. These spectra are fitted (black dotted lines) based on theoretical Stark shifts using electric fields from d). d) Graph showing the spatial distribution of the electric field at room temperature caused by the chemisorbed adatoms.

7.6 Rydberg spectroscopy at cryogenic temperature

Rydberg spectroscopy experiments have been performed to identify the spatial profile of the electric field at room temperature as discussed in Sec. 7.5. It is interesting to investigate if the distribution of this electric field below the chip structure at room temperature is changed at cryogenic temperature. By cooling down the atomic chip to 83 K, Rydberg spectroscopy using Rydberg $44D_{5/2}$, $48D_{5/2}$ and $52D_{5/2}$ states are carried out. The resulting spectra are shown in Fig. 7.7a, Fig. 7.7b and Fig. 7.7c respectively. Only two lines are visible in these figures. They correspond to the $|m_j| = 3/2$ and $|m_j| = 1/2$ manifolds of their $nD_{5/2}$ state. The $|m_j| = 5/2$ manifold is not seen as the strength of the electric field shifts it out of our accessible coupling beam detuning region. While the field is still seen to increase close to the chip surface, there is a significant, relatively constant field at distances exceeding 2 mm. Such a change in electric field distribution is not expected to arise due to chemisorption, as it is already saturated at room temperature.

There are a few questions to address based on what have been observed in these spectra. Firstly, what causes the large splittings between $|m_j| = 3/2$ and $|m_j| = 1/2$ at cryogenic temperature? Are the splittings caused by magnetic fields (e.g. eddy currents from the cold finger, magnetic fields from the vortices inside the type-II superconductor, etc.) or electric fields (e.g. electronic noises from equipment, grounding of the substrate, etc.). Secondly, if they are electric fields or magnetic fields, what are their sources? These two questions are to be dealt with separately in the following two subsections.

Sources of splittings - Electric field or magnetic field?

To investigate whether the splittings between $|m_j| = 3/2$ and $|m_j| = 1/2$ are caused by electric fields or magnetic fields, one can perform Rydberg spectroscopy experiment using various Rydberg states of the same l_j but different n . From the spectra obtained, the level shift of its m_j states will be of the same magnitude if it is the Zeeman effect but will be of different magnitude if it is the Stark effect. Using this physical understanding, a few Rydberg spectroscopy experiments using Rydberg $D_{5/2}$ states are performed at different n . Fig 7.7e shows the separation between the $|m_j| = 3/2$ and $|m_j| = 1/2$ states as a function of n -state by using the splittings value at a distance of 2 mm away from the substrate. The obtained n dependency follows the Stark splittings between $|m_j| = 3/2$ and $|m_j| = 1/2$ at 1 V/cm, indicating that our observations result entirely from the coupling to the electric field.

Sources of electric field

From previous section (Sec. 7.6), it has been identified that the spectra are caused by electric fields. So, it is important to understand further what are the sources of these electric fields. One observation is that the spectra shown in Fig. 7.7a, Fig. 7.7b and Fig. 7.7c are at its saturated values. This means a certain number of experimental runs are required before the spectrum does not change any further. To show this, a series of spectra obtained using the $52D_{5/2}$ state are shown in Fig. 7.8a $\rightarrow$ d. These experiments are performed by scanning the coupling beam detuning from positive to negative direction.

Figure 7.7: Spectra obtained for a) $44D_{5/2}$ b) $48D_{5/2}$ and c) $52D_{5/2}$ Rydberg states at 83 K. Labels in white show the corresponding $|m_j| = 3/2, 1/2$ manifolds. The $|m_j| = 5/2$ is shifted out of the scanning region. These spectra are fitted (black dotted lines) with theoretical Stark shifts using total electric fields from d). d) Spatial distribution of the electric field at 83 K. Electric field from chemisorbed layer (black) is summed to the electric field from physisorbed layer (blue) to give the total electric field (red). e) Plot of the energy difference between $|m_j| = 1/2$ and $|m_j| = 3/2$ v.s. n of the Rydberg state (blue) at the distance of 2 mm. The data scale with the theoretical splittings (red) at an electric field of 1 V/cm.

Chapter 7. Experimental investigation of adsorption phenomena

7.6. Rydberg spectroscopy at cryogenic temperature

	1 st Experiment	2 nd Experiment	3 rd Experiment
Step 1	Cool down the atom chip and have experimental runs of a few hundred times with		Cool down the atom chip and
	LIAD : OFF Electronics : ON	LIAD : ON Electronics : OFF	Dispenser : ON
	↓	↓	↓
Step 2	scan the coupling detuning		
	↓	↓	↓
Observation	Increasing of splitting over runs and splitting comes to a saturation after certain number of runs.	Splitting is saturated value.	Splitting is closed to saturated value.
Conclusion	Sources of electric field are not from electronic equipment.	Sources of electric field are from LIAD.	Sources of electric field are from rubidium atoms.

Table 7.1: Table showing 3 sets of experiments performed with varied conditions to find the possible sources of electric field measured in Fig. 7.7. The comparisons between the observations from each experiment lead to the conclusion that it is the rubidium atoms that are responsible for the electric field. Rubidium atoms are desorbed from the vacuum glass cell wall during LIAD time and adsorbed on the atom chip substrate.

Arrows (black) are drawn to the position at 2 mm where the magnitude of the Stark shift for the corresponding $|m_j| = 3/2$ and $|m_j| = 1/2$ manifolds are written (white). Since Fig. 7.8a → d are obtained in the sequential order from positive to negative detunings, it is thus possible to know the corresponding Stark shifts at 2 mm versus its nth run. Fig 7.8e plots the electric fields v.s. the nth run of experiment by converting the Stark shifts for $|m_j| = 3/2$ and $|m_j| = 1/2$ states in Fig. 7.8a → d to electric fields based on the theoretical Stark maps. Rydberg electrometry is only capable of measuring the magnitude but not the direction of electric fields. The conclusion in Sec. 7.7 shows negative electric fields produced by the physisorbed layer which is the same sign as in Fig. 7.8e. It is apparent from these figures that the splittings between $|m_j| = 3/2$ and $|m_j| = 1/2$ increase with the number of the experimental runs and come to a saturation after a certain number.

To examine and investigate further, a few experiments are performed by varying some settings to identify the possible sources. Experiments that are run under different settings and conditions are listed in Table 7.1 and its results are compared to each other. As shown by Table 7.1, 3 sets of experiments using Rydberg $52D_{5/2}$ state were conducted. Before running each set of experiments, the atom chip was cooled down to its cryogenic temperature using liquid nitrogen and put on hold for a waiting time of 45 minutes. This was to make sure the temperature of the system reached a steady state and equilibrated with the environment. Afterwards, the first experiment was performed for a few hundred times with the LIAD turned off but all the electronics sources were turned on (e.g. currents used for the MOT, magnetic trap and homogeneous bias field) during the ex-

Figure 7.8: Shown are the spectra of Rydberg spectroscopy using $52D_{5/2}$ state with the coupling beam detunings scanned from positive to negative direction. Spectra are obtained in the sequence of a) $\rightarrow$ b) $\rightarrow$ c) $\rightarrow$ d). Arrows (black) are drawn to the position at 2 mm with the magnitude of Stark shift written for $|m_j| = 3/2$ and $|m_j| = 1/2$ manifolds (white). (Blue region at the edge Fig. a), b) and c) refer to data not taken. Colour same as background colour is used and to ensure same axis scaling for Fig. a) $\rightarrow$ d). The increased splitting with the experiment runs shows that the experimental run itself introduces an electric field. e) Graph for the converted detunings to electric fields v.s. nth experimental run.

perimental runs. These allows us to infer if the electronic equipment in the experimental setup were responsible for the electric fields measured. The second step was to perform the Rydberg spectroscopy experiment by scanning the coupling beam detuning. It turned out that increase splitting over the experiment runs were observed which led us to the conclusion that it was not the electronics that introduce the electric fields.

During the second experiment, the UV light for LIAD was turned on but electronics were turned off. The UV light is illuminating the vacuum glass cell which results in desorption of adsorbed rubidium atoms from the glass surface. This experiment allowed us to check if the desorbed rubidium atoms influence the measured electric field. The Rydberg spectroscopy showed that the splittings were at its maximum value, meaning the desorbed atoms from the LIAD adsorbed on the chip surface and increased the electric field. We can see an increase of the splittings over experimental runs as shown in Fig. 7.8 confirming each run introduces a certain amount of adsorbates on the atom chip substrate. This adsorption process continues until the atom chip is fully saturated with adsorbates. Inside our UHV system not only rubidium atoms are adsorbed on the glass cell, there are also other atomic species (e.g. H_2 , He, H_2O , N_2 , etc.) that can't be pumped out efficiently by the ion getter pump and the titanium sublimation pump [95]. Thus, there is a possibility these atomic species may contribute to the adsorption.

Figure 7.9: a) Rydberg spectrum of $52D_{5/2}$ state after running our dispenser with 3.84 A for 20 minutes to reach a pressure of 1.4×10^{-9} mbar. b) Rydberg spectrum of $52D_{5/2}$ state after running our dispenser with 3.66 A for 20 minutes to reach a pressure of 0.69×10^{-9} mbar.

To find the atom species responsible for adsorption on the atom chip, a third experiment was carried out with the rubidium dispenser turned on at a current of 3.84 A for 20 minutes when the ion gauge reached a pressure reading of 1.4×10^{-9} mbar. If the acquired Rydberg spectrum shows a minimum splitting ($\sim (11.6 - (-8.4)) = 20$ MHz)) as shown in Fig. 7.8a, it indicates that the adsorbates are not originated from rubidium atoms. However, if the spectrum shows the saturated splitting ($\sim (23.4 - (-15.4)) = 38.8$ MHz)) similar to Fig. 7.8d, the rubidium atoms are causing the electric field. Fig. 7.9a shows the Rydberg spectrum obtained after loading the rubidium dispenser. It has a splitting of 35.2 MHz, a value which is close to the 38.8 MHz. A possible explanation is 20 minutes of

rubidium dispenser loading time with the vacuum pressure that reached 1.4×10^{-9} mbar was insufficient to completely saturate the substrate with adsorbates. This explanation is supported by another experiment by applying a dispenser current of lower magnitude, 3.66 A for 20 minutes to reach a vacuum pressure of 0.69×10^{-9} mbar. As the pressure was lower, the adsorbates should be less. Indeed, the spectrum shown in Fig. 7.9b shows a splitting of 31.6 MHz, a value lower than 35.2 MHz obtained at higher pressure. These findings suggest that rubidium atoms are adsorbed on the substrate.

Overall, these series of experiments and observations point to the fact that at cryogenic temperature, rubidium atoms can overcome the temperature barrier and get adsorbed on the surface, changing the existing electric field at room temperature. From the discussion of the characteristics of chemisorption and physisorption in Sec. 6.5, it can be deduced this adsorption phenomenon is physisorption which usually occurs at low temperature.

Modelling the physisorbed layers

The electric fields close to the chip surface are due to adsorbates coinciding with the calculations of electric fields based on a dipole layer square sheet introduced in Sec. 7.4 for physisorbed adsorbates. The relatively constant field at distances far from the chip suggests a larger layer of adsorbates than the one present on the 1 mm YBCO square, indicating the presence of physisorption on both the YSZ substrate and YBCO. Since the chemisorbed layer is unchanged at lower temperature, its electric fields have to be included in the calculation. So, the total electric fields above the centre of the YBCO square, E_{total} is the summation of the electric fields from chemisorbed layer, E_{chem} and the newly formed physisorbed layer, E_{phys} at cryogenic temperature. It is written as

$$E_{total} = E_{chem} + E_{phys} \quad (7.10)$$

To explain the smaller electric field gradient at far distance, two physisorbed layers consist of physisorption on the YSZ substrate and on the YBCO are modelled. Both electric field distributions are denoted by $E_{phys\ YSZ}$ and $E_{phys\ YBCO}$.

$$E_{phys} = E_{phys\ YSZ} + E_{phys\ YBCO} \quad (7.11)$$

The resultant fit to the spectrum shows $E_{phys\ YSZ}$ is produced by a physisorbed layer with a dipole density of $\rho_{phys\ YSZ} = -1.7 \times 10^{-11}$ C m⁻¹ distributed on YSZ over a size of 13.3 mm. $E_{phys\ YBCO}$ is produced by dipole density of $\rho_{phys\ YBCO} = -2.3 \times 10^{-11}$ C m⁻¹ on YBCO of size 1 mm. While the localized contribution is explained by the presence of the YBCO square, the larger distribution of dipoles is comparable to the size of our YSZ substrate (20 mm). Fig. 7.7d shows a graph with the E_{chem} (black) obtained from Fig. 7.6d added to the electric fields from physisorbed layers, E_{phys} (blue) to produce the total electric field, E_{total} . Fittings from E_{total} to the corresponding $nD_{5/2}$ state in Fig. 7.7a, 7.7b and 7.7c are shown by the black dotted lines.

7.7 Temperature dependent spectrum

The electric field distribution, E_{chem} (Fig. 7.6d) at room temperature and at cryogenic temperature, E_{total} (Fig. 7.7d) should serve as the boundary conditions for these two temperature limits. In other words, if the temperature of the substrate is varied from cryogenic temperature to room temperature, E_{total} should evolve to E_{chem} . To study the temperature dependence of the adsorption phenomenon, δ_c is fixed at 27 MHz while the temperature of the substrate is increased from cryogenic temperature to room temperature¹. The result is shown in Fig. 7.10a.

There are two important features in Fig. 7.10a, the transformation of the broad EIT resonance to a narrower resonance at 200 K and the reversal of this narrow EIT resonance at 230 K. From this figure, there is a broad EIT resonance from position 0.8 mm to 2.8 mm at the temperature of 140 K. This broad EIT resonance is constant until it reaches 190 K. It transforms to a narrower EIT resonance at 200 K. As the chip temperature increases from 200 K, this resonance moves closer to the chip surface indicating a weakening of the adsorbate electric field. At approximately 230 K, the resonance vanishes implying the cancellation of the electric field. Above 230 K, the resonance reverts to the room temperature value. At 250 K, the spatial EIT resonance is positioned at 0.6 mm. Since the Stark shift of a Rydberg state only depends on the magnitude of the field, the transformation at 200 K and the reversal at 230 K suggest a change in the direction of the electric field rather than a discontinuity in the field change. As such, our data indicates that the dipole moment of the physisorbed rubidium atoms carries the opposite sign from those that are chemisorbed. Indeed, due to the difference in work function of the substrate and the ionization energy of rubidium atom, electrons are pulled from rubidium adatoms toward the substrate during chemisorption. On the other hand, as discussed in Sec. 6.3 for the case of physisorption, balancing between the repulsive potential and attractive van der Waals potential may lead to an opposite dipole moment compared to a chemisorbed adatom.

To theoretically model the temperature dependent spectrum in Fig. 7.10a, the Langmuir isobar in Eq. 6.16 is used to simulate the temperature dependency of the physisorbed layers. In this equation, temperature of the gas is at $T_{gas} = 293$ K and vacuum pressure is at $P = 4 \times 10^{-10}$ mbar. Shown in Fig. 7.10b are two curves for the temperature dependent behaviour of physisorbed layers on YSZ (blue) and on YBCO (red) substrate respectively. These are necessary to explain the cancelling of electric fields at two temperature regions, 200 K and 230 K. First, the change of the large, constant electric field at 200 K is caused by desorption of physisorbed layer on the large YSZ substrate. This is described by the Langmuir isobar curve (blue) with adsorption energy of 790 meV. Another desorption process follows for the physisorbed layer on the YBCO which is described by the Langmuir isobar curve (red) with adsorption energy of 880 meV. These desorption processes are graphically highlighted in Fig. 7.11.

With the physisorbed layers on YSZ and on YBCO described by Langmuir isobar equations, simulations of the temperature dependent electric fields above the YBCO square are straightforward. Using the fitted surface dipole densities obtained at cryogenic

¹We measured the resistance of the YBCO and related it to the temperature of our atom chip. The temperature and resistance is of linear proportional to each other as shown and measured by Wu, M. K. *et al.* [75]. The calibration is shown in Appendix E

Figure 7.10: a) Temperature dependent spectrum with $\delta_c = 27$ MHz. b) Langmuir isobar curves for physisorbed layers on YSZ substrate (blue) and on YBCO (red) with adsorption energy of 790 meV and 880 meV respectively. c) Electric fields distribution v.s. temperature with the behaviour of surface dipole density on YSZ and on YBCO changing in linear proportion to the Langmuir isobar curves according to Eq. 7.12. d) Simulated spectrum with c) subject to a gaussian equation such that the EIT resonance at δ_c decays in a gaussian form.

Figure 7.11: Pictures showing the physisorbed and chemisorbed layer on the atom chip. The different direction of the dipole moments for chemisorbed adatoms and physisorbed adatoms are also drawn. a) At cryogenic temperature, physisorbed layer of dipole density, $\rho_{phys\ YSZ}$ (black box) is distributed over the size of 13.3 mm on YSZ substrate. For YBCO substrate, the dipole density of its physisorbed layer (red box) has a value of $\rho_{phys\ YBCO}$. This physisorbed layer is adsorbed above the existing chemisorbed layer (purple box) of dipole density, ρ_{chem} . b) Physisorbed layer on YSZ desorbs first and its desorption dynamics is described by Langmuir isobar curve (blue) with adsorption energy 790 meV in Fig. 7.10b. For a non-saturated case in Fig. 7.12b, its adsorption energy is 703 meV. c) Desorption of physisorbed layer from YBCO (red box) is followed next as described by Langmuir isobar curve (red) with adsorption energy of 880 meV in Fig. 7.10b, leaving behind the chemisorbed layer as the temperature increases to room temperature. For non-saturated case, it is described in Fig. 7.12b with adsorption energy of 783 meV.

temperature, they are simulated to vary with respect to temperature according to the Langmuir isobars curves in Fig. 7.10b. Mathematically, it is described as

$$\rho_{phys}(T) = \rho_{phys}(T = 87 \text{ K}) \times \theta_{ads}(T) \quad (7.12)$$

where $\rho_{phys}(T = 87 \text{ K})$ is the surface dipole densities fitted to the Fig. 7.7a, Fig. 7.7b and Fig. 7.7c respectively. $\theta_{ads}(T)$ is basically the coverage dependence of temperature plotted in Fig. 7.10b. Let's choose surface dipole density for physisorbed atoms on YSZ as an example. $\rho_{phys \text{ YSZ}}$ is chosen to be $-1.6 \times 10^{-11} \text{ C m}^{-1}$ in our simulation and does not vary much at temperature below 160 K according to Fig. 7.10b (blue line). However, $\rho_{phys \text{ YSZ}} = 0$ at temperature above 240 K. At 200 K, the coverage becomes $\theta_{ads} = 0.5$. So, $\rho_{phys \text{ YSZ}}(T = 200 \text{ K}) = (0.5 \times -1.6) \times 10^{-11} \text{ C m}^{-1} = -0.8 \times 10^{-11} \text{ C m}^{-1}$. Similar approach is also applied to $\rho_{phys \text{ YBCO}} = -2.1 \times 10^{-11} \text{ C m}^{-1}$. $\rho_{phys \text{ YSZ}}$ and $\rho_{phys \text{ YBCO}}$ used in this simulation are chosen to be within 10% error from the one quoted earlier ($\rho_{phys \text{ YSZ}} = -1.7 \times 10^{-11} \text{ C m}^{-1}$ and $\rho_{phys \text{ YBCO}} = -2.3 \times 10^{-11} \text{ C m}^{-1}$) in Sec. 7.6 to account for changes in experimental conditions resulting in different adsorbates density. These slight changes of the dipole density will not change the general feature of the temperature dependent spectrum. The result of the simulated temperature dependent electric field is shown in Fig. 7.10c. The electric field is negative at cryogenic temperature but transforms to positive at room temperature. This is because at cryogenic temperature, the electric field from physisorbed atoms dominate with its dipole moments opposite to the chemisorbed adatoms. The smaller gradient at 140 K also accounts for the fact that physisorption occurs on a larger area, that is the YSZ substrate. This electric field is weakened at 200 K and 230 K. As expected, above 230 K, the electric field is approaching the E_{chem} shown in Fig. 7.6d as the physisorbed layers are completely desorbed.

Since δ_c chosen is positive, according to the theoretical Stark maps in Fig. 5.6, $|m_j| = 3/2$ and $5/2$ will not appear in the temperature dependent spectrum. Thus, δ_c of 27 MHz corresponds to the absolute value of the resonance electric field, $|E_0| = 1.07 \text{ V/cm}$ for the quantum manifold $|m_j| = 1/2$ in $52D_{5/2}$ quantum state. This indirectly shows that EIT resonance will only occur at a position where its electric field is $|1.07| \text{ V/cm}$ as shown in Fig. 7.10c. The linewidth of the blue laser is incorporated so that it can be translated to the electric field linewidth. For a linewidth of 7 MHz, this is equivalent to the electric field linewidth, δ_{E_0} of 0.35 V/cm. With these information at hand, the expected spectrum S_{sim} can be deduced from Fig. 7.10c. It is written as

$$S_{sim} = -\exp \left[\frac{(|E_{sim}| - |E_0|)^2}{2\delta_{E_0}^2} \right] \quad (7.13)$$

where E_{sim} is the matrix for the electric field distribution plotted in Fig. 7.10c. Since the Rydberg states are only sensitive to the absolute of the electric field, $|E_{sim}|$ has to be used. The result of the simulated spectrum, S_{sim} is shown in Fig. 7.10d.

The general features of Fig. 7.10d agree well with the experimental spectrum in Fig. 7.10a. That is, the change of the broad EIT resonance to a narrower resonance at 200 K and another reversal of the EIT resonance at 230 K. However, the resonance closer to the surface is too weak to be detected in the experiment, rendering the inner resonance line in Fig. 7.10d unseen in Fig. 7.10a.

To investigate further the reversal feature at 230 K in Fig. 7.10a, we shifted the

Figure 7.12: a) Temperature dependent spectrum with $\delta_c = 21$ MHz. b) Langmuir isobar curves for physisorbed layers on YSZ substrate (blue) and on YBCO (red) with adsorption energy of 703 meV and 783 meV respectively. c) Electric fields distribution v.s. temperature with the behaviour of surface dipole density on YSZ and on YBCO changing in linear proportion to the Langmuir isobar curves according to Eq. 7.12. d) Simulated spectrum with c) subject to a gaussian equation such that the EIT resonance at δ_c decays in a gaussian form.

resonance signal further from the diffraction lines close to the chip surface by decreasing the coupling beam detuning to 21 MHz. In addition, in this experiment, the atom chip was not saturated with physisorbed atoms on purpose. The acquired spectrum is shown in Fig. 7.12a. It is clearly seen with these parameters, the reversal feature is further away from the surface. With a non-saturated physisorbed layer, the resultant electric field is smaller and the cancellation of the electric field occurs at a lower temperature of 202 K, showing the coverage dependence of adsorption energy [152]. As before, we use a similar approach to simulate the temperature dependent spectrum. The temperature dependent coverage is plotted in Fig. 7.12b with adsorption energy of 703 meV (blue) and 783 meV (red) respectively for the adsorption on YSZ and on YBCO. With these two Langmuir isobar curves, the temperature dependent electric field is simulated and shown in Fig. 7.12c. This again is transformed using Eq. 7.13 to obtain the simulated temperature dependent spectrum as shown in Fig. 7.12d.

7.7.1 Discussion

Our work on physisorption overlaps with other researches in different fields. These include the studies of LIAD [183, 184, 185, 186], electrical conductivity of the glass cell wall exposed to alkali vapour [187, 188], interaction of atoms with cell coatings inside vapour cell [189, 190, 191, 192], etc. These studies of physisorption are extensive and one can borrow some insights in the context of their results. In this regards, the adsorption energies from these references are quoted and listed in Table 7.2. Their works and the relations to our studies are discussed in details in the following sections.

Relation between adsorption energy and heat of vaporization

The adsorption energy for caesium adsorbed on pyrex was measured to be 0.53 eV [189]. For caesium adsorbed on fused silica, it was found to be 0.66 eV [188]. Noticed by Hatakeyama, A., *et al.* [193], these measured values are close to the heat of vaporization of metallic bulk caesium (0.66 eV [194]). This is to be expected because adsorbates would show its bulk property and metallic nature with increasing coverage towards a monolayer. The physical insight for this direct link between adsorption energy and heat of vaporization can be applied in our case. For bulk rubidium, its heat of vaporization is 787 meV [195]. As mentioned earlier, our temperature dependent Rydberg spectroscopy results show the adsorption energies to be 790 meV for the saturated case and 703 meV for non-saturated case on YSZ. For YBCO, they're 880 meV and 783 meV respectively. Our fitted adsorption energies based on Eq. 6.16 are close to the heat of vaporization² of bulk rubidium showing a similar relationship as is found in the caesium case [193]. This finding also serves as a double confirmation as discussed in Sec. 7.6 that the sources of electric fields are indeed from rubidium atoms. In addition, since our fitted adsorption energies

²To be correct the heat of sublimation should be used for comparison rather than heat of vaporization as the adsorbate-substrate system is between a solid and a gas phase. However, the heat of fusion (energy required to change from solid to liquid phase) is only 23 meV for rubidium [195]. So, the heat of sublimation is mostly contributed by heat of vaporization. It is to be cautious that one should not take the value for the adsorption energy converted from heat of sublimation as absolute. It can be used as a rough estimate for the adsorption energy. This is because the parameter heat of sublimation is usually measured under the standard temperature and pressure (STP) conditions which may not apply to the physisorbed atoms which are not at room temperature.

are close to the heat of vaporization, this suggests at least a monolayer of rubidium is formed on our substrate.

Coverage dependence of adsorption energy

In addition, our results also show the adsorption energies of rubidium atoms on both YSZ and YBCO are lower in the non-saturated case compared to the saturated case. This finding agrees with what has been found by Torralbo-Campo, L., *et al.* [196]. Adsorption energy for rubidium atoms on pyrex is measured to be 0.5 eV [196]. For a longer rubidium vapour exposure time, the adsorption energy is measured to be 0.7 eV [196]. There were also attempts to reduce the adsorption energy of alkali atoms on insulating substrate by coating organic films [190, 191, 192] to prevent the formation of a metallic monolayer [190, 81]. All these studies point to the fact that a lower adsorption energy is always associated to a lower density for the adsorbates [197], consistent with Langmuir equation.

Minimize physisorption

The adsorbates electric field from chemisorption are proven to be detrimental in a QED experiment [70] based on the electric field coupling between a Rydberg atom and a resonator [63]. One can minimize this chemisorption by careful selection of the materials. This can be accomplished by choosing a substrate with closer work function to the ionization energy of rubidium atoms [67] or by using an insulating substrate [66]. However, even with these suggested schemes, our studies show that at cryogenic temperature, physisorption can still dominate and give rise to an electric field that shifts the energy levels of the Rydberg states.

In a typical optical pumping or LIAD experiment, the Langmuir equation used is a modified one [198] compared to Eq. 6.16. As such, the sticking time of atoms on the substrate, τ_s is related to the adsorption energy, ΔE_{ads} as given by

$$\tau_s = \tau_0 \exp \left(\frac{\Delta E_{ads}}{kT} \right) \quad (7.14)$$

where τ_0 is the sticking time at the high temperature limit and is $\approx 10^{-12}$ s [199]³. Eq. 7.14 suggests a lower adsorption energy leads to shorter sticking time which makes the collision with the wall elastic. In the end, this implies lower surface density and thus, an increase of the relaxation times of atomic polarization [201, 202]. As a consequence, in these types of experiments, the goal is to select an appropriate choice of substrate or coating to minimize the adsorption energy which is similar to our situation.

Since the Van der Waals interaction strength is proportional to the dielectric constant, Stephens, M., *et al.* [189] work on materials that have low dielectric constant to minimize adsorption energy. Alkali-resistant glass has a dielectric constant of ≈ 6 while pyrex has

³In the field of surface chemistry, usually it is the heat of adsorption that is used and measured as shown in [200]. However, the atomic physics groups listed in Table 7.2 used adsorption energy. Difference between these two energy terms are discussed in Sec. 6.4.1.

a dielectric constant of ≈ 4.4 . Thus, Stephens, M., *et al.* [189] choose pyrex over alkali-resistant glass as they expect a lower adsorption energy for pyrex. Our YSZ substrate has a relatively high dielectric constant (> 25 [203, 204, 205]) which is similar to the dielectric constant of YBCO [206]. Since the dielectric constants are high, these may contribute to a strong physisorbed dipole moment and thus higher adsorption energy.

There were also studies to reduce the adsorption energy of alkali atoms by coating the insulating substrate of a vapour cell with an organic films. The methyl group of the organic materials polarize its neighbour such that a smooth surface is formed [189, 207]. This makes the organic film less polarizable for an incoming atom to form an image induced dipole, resulting in low dielectric constant [189, 207]. Adsorption energies as low as 0.1 eV were found by coating with silane [190] or paraffin [191]. These coatings inhibit the formation of a metallic monolayer [190, 81]. However, a quick calculation with the adsorption energy of 0.1 eV using Langmuir equation in Eq. 6.16 shows that physisorption will occur at about 24 K for a vacuum pressure of 3×10^{-11} mbar (typical pressure for a liquid helium cryostat [47]). By choosing a substrate with a lower dielectric constant or an organic coating, the adsorption energy can be lowered. However, in most cases, it is not low enough to prevent physisorption from occurring in a typical superconducting atom chip experiment that operates at liquid helium temperature [208].

Recently, Thiele, T., *et al.* [209] successfully minimized the stray electric field from adsorption and demonstrated coherence measurement using Rydberg states of helium atoms on a cryogenic surface. It was done by maintaining the sample above the adsorption temperature of the adsorbing species (H_2O , N_2 , O_2 , and CO_2) initially while steadily lowering the temperature of the cooling stage [209]. Thus, the adsorbates were formed on the cooling stage and not on the sample [209]. After the residual gases were below a certain partial pressure, the temperature of the sample was lowered gradually until it finally achieved 3 K [209]. The adsorbates electric fields were successfully minimized. Still, there was a small electric field from the environment which was cancelled with an applied external electric field [209]. The final electric field was decreased below 100 mV/cm over a region of 6 mm [209]. In addition, their choice of using helium atoms does not give rise to any measurable electric field as compared to our observation for rubidium atoms as discussed in Sec. 7.6. This atom candidate was chosen on purpose such that it minimizes the adsorption [210].

Dipole moments for physisorbed atoms

Our studies not only show that physisorption occurs at cryogenic temperature, but the dipoles formed from physisorbed adatoms (rubidium) are directed towards the substrate. As discussed in Sec. 6.3, the direction of the dipole moments formed differs for light and heavy atoms [168, 169]. For physisorbed hydrogen, the simulated dipole moment is oriented inward at equilibrium position of the van der Waals potential [169]. However, the dipole moment of Xe atoms physisorbed on Pd(100) is found to be directed outward, which is verified experimentally by the decrease of the work function of the substrate with increasing coverage [170]. So far, the theoretical and experiment works by Kamins, T. I. [187] match closely to our observation. This model is based on image-force and predicts that caesium adatoms on an insulating substrate have a dipole moment of 3 D and is pointing toward the substrate [187]. The simulated value for this dipole moment

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7.7. Temperature dependent spectrum

is also close to what has been measured by Obrecht, J. M., *et al.* [67]. The rubidium atoms adsorbed on fused silica are measured to have dipole moment of 3.2 D [67].

Authors	Adsorbates on substrate	Adsorption energy (eV)
Gozzini, S., <i>et al.</i> [183]	Na on pyrex glass	0.71 ± 0.02
Bonch-Bruevi, A. M., <i>et al.</i> [184, 185]	Na on sapphire	0.75 ± 0.25
Werij, H. G. C, <i>et al.</i> [186]	Na on gehlenite glass	≈ 1
Kamins, T. I. [187]	Cs on glass	≈ 0.7
Bouchiat, M. A., <i>et al.</i> [188]	Cs on glass (fused silica)	0.66 ± 0.05
Stephens, M., <i>et al.</i> [189]	Cs on dry film (octadecyltrichlorosilane (OTS) on Pyrex)	0.40 ± 0.03
	Cs on pyrex	0.53 ± 0.03
	Cs on sapphire	0.43 ± 0.1
Xu, J. H., <i>et al.</i> [190]	Na on silane-coated cell	0.1
Bouchiat, M. A., <i>et al.</i> [191]	Rb on paraffin-coated cell	0.1
Yi, Y. W., <i>et al.</i> [192]	Rb on OTS	0.065 ± 0.002
Torralbo-Campo, <i>et al.</i> [196]	Rb on pyrex	0.5
	Rb on pyrex (longer evolution of rubidium pressure)	0.7
Atutov, S. N., <i>et al.</i> [211]	Rb on (poly)dimethylsiloxane (PDMS)	0.7 ± 0.1

Table 7.2: List of the adsorption energies obtained from various sources for physisorption of alkali atoms on insulating substrate.

Chapter 8

Conclusion and Outlook

8.1 Conclusion

Part I

To conclude Part I, we have simulated and experimentally produced magnetic potentials for ultracold atoms using magnetic fields generated by the vortices in a type-II superconductor. The vortices can be introduced by applying a magnetic field B_{ext} in the range of $B_{c1} < B_{ext} < B_{c2}$ or by applying a transport current. Employing the hysteresis characteristic of the type-II superconductor, we have shown the various types of complex trapping geometries based on the simulations using critical state model. In the experiments, we have successfully realized and characterized some of these complex potentials. This includes a single trap (superconducting strip and square structures) and double traps (superconducting strip structure) above our superconductors. We even achieve the self-sustaining trap (superconducting strip and square structures). This trap is formed solely by the persistent currents of the superconductor without the use of any homogeneous bias fields. We not only show that the use of type-II superconductors presents a rich pool of trapping geometries, but also its reconfigurability. We show in our simulations that a single trap formed above a superconducting disk or ring can be transformed to a ring potential by tuning the strength of the applied perpendicular homogeneous bias field. In our experiment, we have demonstrated the reconfigurability on a superconducting square. We have transformed the magnetic potential from a single trap to a four traps, thus showing the four-way beam splitter like analogy in this experiment.

For atoms trapped by the magnetic potentials generated by the vortices, we have shown that these atoms can be used as a sensitive probe of the vortex pattern. We have measured the trap to surface distance of the atomic cloud and show that the vortices in our type-II superconductor do not relax and are stable for as long as 2 hours. Motivated by the vortex stability in our experiments, we continue to investigate further and show the hysteresis behaviour of the type-II superconductor by plotting the trap to surface distance with respect to the magnetized field. This hysteresis plot agrees well with our simulation. In addition, we also show that an applied transport current will alter and redistribute the existing vortex pattern. The resulted change is manifested in the measured change of the trap position of our atomic cloud. By bringing the atomic cloud closer to our

superconducting square, we use the density spatial profile of the atomic cloud to image the magnetic potentials formed by the vortices.

Part II

To conclude Part II, we have successfully measured the electric field above the superconducting square structure using Rydberg spectroscopy at room temperature, cryogenic temperature (83 K) and its dependency on temperature. The electric field is dominantly produced by chemisorbed atoms adsorbed on the 1 mm superconducting square at room temperature. The electric field decays over a length of 1 mm, consistent with the size of our square structure. At cryogenic temperature, physisorption overcomes chemisorption and gives rise to a much stronger electric field compared to room temperature. Physisorbed atoms do not only adsorb on our superconducting square but also on the much larger area of our YSZ substrate. This gives rise to a smaller electric field gradient, a length scale characteristic of the size of our atom chip (20 mm). Using Rydberg spectroscopy, we have also studied the temperature dependency of the electric field from room to cryogenic temperature. The Rydberg spectrum obtained are then modelled and simulated based on the Langmuir equation. Due to the reversal EIT features of the measured Rydberg spectrum, we discovered that the electric dipole moment for the physisorbed rubidium atoms are opposite as compared to the chemisorbed atoms. Finally, the simulated Rydberg spectrum agrees qualitatively with the experimental Rydberg spectrum.

8.2 Outlook

Part I

The realization of the various complex trapping potentials shows the advantages of flexibility and versatility in using superconductors to trap ultracold atoms. Thus, one can design various magnetic potentials using the same microstructures on the atom chip. These advantages can also be used for a quantum simulator experiment. One such example is a magnetic lattice potential formed by the vortices pinned to an array of defects fabricated in a type-II superconductor [212]. Tunnelling rate for the atoms among the lattice sites can be tuned such that it mimics a Hubbard Hamiltonian system [212]. The formation of a ring potential using a superconducting disk or ring structure can also be employed to simulate an atomtronics system such as superconducting quantum interference device (SQUID) [112].

The trapped atoms can also be used to probe the various properties of a type-II superconductor. For example, the vortex noise due to flux creep can be studied by measuring the coherence and spin-flip lifetime of the trapped atoms. The induced current in the superconductors due to the magnetic dipole moment of the trapped BEC can also be investigated by measuring the change of the trapping frequency [213]. Ultimately, one can also use the atoms to probe the property of a single flux pinned to a superconductor [214]. If the atoms are brought close enough to the surface, imaging of the spatial distribution of a single vortex is possible [114].

Superconductors offer an opportunity to entangle an atomic quantum system and a solid state quantum system. One such hybrid system is a BEC trapped above a superconductor entangled to the flux qubit in a SQUID [215]. For a superposition of the flux qubit, the position of the trapped atoms is in superposition of state depending on the direction of the persistent current in the flux qubit [215].

Our studies show that it is not only possible to trap the atoms using superconductors, but also show an opportunity to form a hybrid quantum system. The advantages of each subsystem is maximized and utilized for quantum information processing. Neutral atoms have long coherence time while the superconducting circuits have fast qubit operation time. Thus, fast qubit operation is performed through a superconducting circuit while the memory is stored in the long coherence qubit state (hyperfine ground state [216], Rydberg state [63, 217, 218]) of the neutral atom.

Part II

Our studies show that the physisorbed rubidium atom has a dipole moment of opposite sign compared to a chemisorbed rubidium atom. This provides a possibility of cancelling out the electric field from chemisorbed atoms and minimize the overall stray electric field. The successful minimization of the adsorbate electric field on the cryogenic atom chip will pave the way towards a coherent control of the Rydberg state near a superconductor as demonstrated by Thiele, T. *et al.* [209]. In addition, the physisorbed atoms are formed by the van der Waals potential and may serve as a potential candidate for the studies of atom surface interactions at close distances.

Appendices

Appendix A: Circuit diagram for current control of dispenser

The schematic and picture of the circuit design used to control the current through the dispenser is shown in Fig. 1 and 2 respectively. Computer analogue voltage is varied at about 1.90 V for ± 0.10 V. An isolator is used to isolate the computer from the circuit. The MOSFET used is an N-MOS (IXYS - IXFN230N10). V_{S+} is set to 6 V.

Figure 1: Schematic of the circuit diagram in controlling the current going through the dispenser.

Figure 2: Picture of the circuit diagram in controlling the current going through the dispenser. Power supply used in providing the current at V_{S+} is not shown. The MOSFET and $1\ \Omega$ resistor are attached to the heat sink to provide heat dissipation.

Appendix B: Abbreviations for optical and electronic elements

Optical and electronic elements	Abbreviations
Acousto-optic modulator	AOM
Anamorphic prism	AP
Attenuator	ATTN
Beam block	BB
Cylindrical lens	CL
Dichroic mirror	DM
External cavity diode laser	ECDL
Fibre adaptor	FA
Fibre port	FP
Frequency divider	FD
Half-wave plate	$\lambda/2$
Interference filter	IF
Laser diode driver	LDD
Lithium triborate	LBO
Lens	L
Lock-in amplifier	Lock-In
Optical isolator	OI
Mirror	M
Mixer	MI
Piezoelectric transducer	PZT
Photodiode	PD
Polarizing beam splitter	PBS
Proportional-integral controller	PI
Quarter-wave plate	$\lambda/4$
Reference signal	Ref
Rubidium vapour cell	Rb VC
Second-harmonic generation	SHG
Tapered amplifier	TA
Walk-off polarizer	WOP

Table 1: List of the abbreviations used for some of the optical elements drawn in this thesis.

Appendix C: Characteristics of the magnetic coils

All our coils (MOT, transfer and bias coils) are made to have 50 windings¹. The measured current (using current metre) and hall probe voltage v.s. the computer analogue voltage for the MOT, transfer and bias coils are tabulated in Table 3, 4, 5, 6 and 7 respectively. These data are then plotted in Fig. 3 and fitted with a linear curve. From Fig. 3a, the current to the MOT coils is saturated at about 114 A. The reason for this saturation is due to limitation of the power supply's voltage which can only provide up to a maximum voltage of 20 V even though its maximum current is 165 A. The slight jigger at 114 A is due to temperature change in the circuit as such a high current produces some heats. From Fig. 3c, 3d and 3e, the currents to the bias coils along x , y and z -direction are also saturated. In contrast to the reason for current saturation of the MOT coils, these bias coils are current limited. The saturated currents measured are consistent with the maximum currents that can be provided by the power supplies which are at 35 A, 76 A and 110 A respectively. Finally, Table 8 lists the relevant parameters for the magnetic coils.

Magnetic coils	Brand - part number	Maximum voltage (V)	Maximum current (A)
MOT coils	TDK Lambda - GEN20-165	20	165
Transfer coils	TDK Lambda - GEN30-110	30	110
Bias coils (B_{bias}^x)	Agilent - 6674A	60	35
Bias coils (B_{bias}^y)	TDK Lambda - GEN20-76	20	76
Bias coils (B_{bias}^z)	TDK Lambda - GEN30-110	30	110

Table 2: Table showing the brand and part number for the power supplies used in supplying current for the relevant magnetic coils. Shown also are the limit for the maximum voltage and current.

¹The jargons used in our experiment for x , y and z -direction as shown in Fig. 2.20b are B-D, A-C, and E-F direction. These jargons are used in labelling the equipment and computer programme.

Computer analogue voltage (V)	Measured current (± 0.2 A)	Hall probe voltage (± 0.01 V)
1.00	14.3	0.16
2.00	29.6	0.328
3.00	44.6	0.492
4.00	59.7	0.656
5.00	74.9	0.832
6.00	90.2	0.992
6.50	97.8	1.08
6.75	101.6	1.13
7.00	105.4	1.17
7.25	109.2	1.21
7.50	112.9	1.25
7.50	112.9	1.24
7.75	114.4	1.25
8.00	114.0	1.25
8.00	113.6	1.25
8.00	116.6	1.28

Table 3: MOT coils measured current (using current metre) and hall probe voltage v.s. computer analogue voltage (Footnote 1).

Computer analogue voltage (V)	Measured current (± 0.2 A)	Hall probe voltage (± 0.01 V)
1.00	8.6	0.424
2.00	17.4	0.848
3.00	26.2	1.27
4.00	35.0	1.70
5.00	43.9	2.14
6.00	52.7	2.58
7.00	61.5	3.02
8.00	70.4	3.44
9.00	79.2	3.88
10.00	88.1	4.28

Table 4: Transfer coils measured current (using current metre) and hall probe voltage v.s. computer analogue voltage (Footnote 1).

Computer analogue voltage (V)	Measured current (± 0.2 A)	Hall probe voltage (± 0.01 V)
1.00	5.4	0.408
2.00	11.0	0.824
3.00	16.5	1.23
4.00	22.0	1.66
5.00	27.5	2.06
6.00	33.1	2.48
6.25	34.5	2.60
6.50	35.8	2.70
6.75	37.2	2.80
7.00	36.3	2.72
8.00	36.3	2.72
9.00	36.3	2.72

Table 5: Bias coils (B_{bias}^x) measured current (using current metre) and hall probe voltage v.s. computer analogue voltage (Footnote 1).

Computer analogue voltage (V)	Measured current (± 0.2 A)	Hall probe voltage (± 0.01 V)
1.00	9.9	0.736
2.00	19.9	1.46
3.00	29.8	2.20
4.00	39.9	2.92
5.00	49.8	3.72
6.00	60.0	4.48
7.00	70.1	5.24
7.25	72.4	5.40
7.50	74.9	5.60
7.75	76.1	5.68
8.00	76.2	5.68
9.00	76.1	5.68

Table 6: Bias coils (B_{bias}^y) measured current (using current metre) and hall probe voltage v.s. computer analogue voltage (Footnote 1).

Computer analogue voltage (V)	Measured current (± 0.2 A)	Hall probe voltage (± 0.01 V)
1.00	14.0	0.648
2.00	27.9	1.28
3.00	41.8	1.92
4.00	55.8	2.56
5.00	69.8	3.20
6.00	83.8	3.84
6.50	90.6	4.16
7.00	97.8	4.48
7.25	101.2	4.64
7.50	104.3	4.80
8.00	104.4	4.80
9.00	104.4	4.80
10.00	104.4	4.80

Table 7: Bias coils (B_{bias}^z) measured current (using current metre) and hall probe voltage v.s. computer analogue voltage (Footnote 1).

Magnetic Trap (Anti-helmholtz coils)	Conversion factor	Distance to centre	Diameter
units	G/cm/A	mm	mm
MOT	0.628	74	107
Transfer (Major axis)	1.11	46.5	143
(Minor axis)	2.43		76
Homogeneous bias field (Helmholtz coils)	Conversion factor	Distance to centre	Diameter
units	G/A	mm	mm
x -direction	1.27	194	119
y -direction	1.22	179	147
z -direction	0.281	272	108

Table 8: Table showing the relevant parameters for our MOT, transfer and bias coils.

Figure 3: Plot of the current v.s. computer analogue voltage for a) MOT coils, b) transfer coils, c) bias coils (B_{bias}^x), d) bias coils (B_{bias}^y) and e) bias coils (B_{bias}^z). The magnitude of the current is controlled by the LabVIEW analogue card with the analogue voltage varied between 0-10 V.

Appendix D: Current supply using car batteries

For the circuit diagram shown in Fig. 2.12, to find the relationship between the hall probe voltage and the current, a calibration is performed first with a known current. The result is plotted in Fig. 4. The 12 V car batteries can be connected in series. The measured voltage difference across the batteries are written down in Table 9. Included also are the measured hall probe voltage which are directly related to the current driven to the system. By using the fitting parameters in Fig. 4, the current can be known. The batteries voltage will drop over time and require some recharges.

Figure 4: Calibration is performed to find the relationship between the hall probe voltage and the current for the circuit diagram sketched in Fig. 2.12.

Number of batteries connected in series	Batteries voltage (V)	Hall probe voltage (V)
1	11.7	0.64
2	24.2	1.28
3	36.4	1.80
4	48.6	2.36
5	60.3	2.56
6	71.5	2.68

Table 9: Measured hall probe voltage for a certain number of 12 V car batteries connected in series.

Appendix E: Temperature and resistance of the YBCO

We approximate the linearity between resistance and temperature of our YBCO by using the result by Wu, M. K. *et al.* [75] as a reference. The linear region begins at an approximate value of $T_1 = 95$ K [75]. This occurs at a measured resistance of $R_1 = 30 \Omega$ for our YBCO. At room temperature $T_2 = 294.15$ K, our YBCO has a resistance of $R_2 = 110.6 \Omega$. Thus, the temperature T_{subs} of our substrate is related to the resistance of our YBCO R as

$$T_{subs} = \frac{T_2 - T_1}{R_2 - R_1}R + T_1 - \frac{T_2 - T_1}{R_2 - R_1}R_1 \quad (1)$$

which is calculated to be

$$T_{subs} = 2.47R + 20.9 \text{ (K)} \quad (2)$$

Table 10 shows the result for this linear correspondence. The resistance is measured using multimeter, thus it depends on the cable length used. The uncertainty could vary for $\pm 2 \Omega$.

R	T_{sub}	R	T_{sub}	R	T_{sub}	R	T_{sub}	R	T_{sub}
30	95.0	47	137.0	64	179.0	81	221.0	98	263.0
31	97.47	48	139.5	65	181.5	82	223.5	99	265.5
32	99.94	49	141.9	66	184.0	83	226.0	100	268.0
33	102.4	50	144.4	67	186.4	84	228.4	101	270.4
34	104.9	51	146.9	68	188.9	85	230.9	102	272.9
35	107.4	52	149.4	69	191.4	86	233.4	103	275.4
36	109.8	53	151.8	70	193.8	87	235.8	104	277.8
37	112.3	54	154.3	71	196.3	88	238.3	105	280.3
38	114.8	55	156.8	72	198.8	89	240.8	106	282.8
39	117.2	56	159.2	73	201.2	90	243.3	107	285.3
40	119.7	57	161.7	74	203.7	91	245.7	108	287.7
41	122.2	58	164.2	75	206.2	92	248.2	109	290.2
42	124.7	59	166.7	76	208.7	93	250.7	110	292.7
43	127.1	60	169.1	77	211.1	94	253.1		
44	129.6	61	171.6	78	213.6	95	255.6		
45	132.1	62	174.1	79	216.1	96	258.1		
46	134.5	63	176.5	80	218.5	97	260.5		

Table 10: Table showing the correspondence between the temperature of our substrate and the resistance of our YBCO.

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