Optical Feshbach Resonances and Coherent Photoassociation in a Strontium BEC

by

Mi Yan

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Approved, Thesis Committee:

Thomas C. Killian, Chair
Professor of Physics and Astronomy

Randall G. Hulet
Fayez Sarofim Professor of Physics and Astronomy

Matteo Pasquali
Professor of Chemical and Biomolecular Engineering and Chemistry

Houston, Texas

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ABSTRACT

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The divalent electronic structure of alkaline-earth metal atoms such as strontium gives rise to metastable, excited triplet levels and narrow intercombination transitions. These have been exploited for optical clocks, powerful laser cooling techniques that are critical for achieving quantum degeneracy in strontium, and proposals for quantum information architectures and studies of novel magnetism. Metastable excited electronic states also give rise to narrow photoassociation (PA) transitions that differ in many ways from traditional PA with broad, electric-dipole-allowed transitions. In this thesis, I will introduce the use of narrow-line PA near the $^{1}S_{0}-^{3}P_{1}$ intercombination transition for two experiments with $^{88}$Sr Bose-Einstein condensates.

The first experiment demonstrates the control of the collapse and expansion of an $^{88}$Sr Bose-Einstein condensate using an optical Feshbach resonance (OFR) near the $^{1}S_{0}-^{3}P_{1}$ intercombination transition at 689 nm. Significant changes in dynam-
ics are caused by modifications of scattering length by up to $\pm 10 a_{bg}$, where the background scattering length of $^{88}\text{Sr}$ is $a_{bg} = -2 a_0$ ($a_0 = 0.053\text{nm}$). Changes in scattering length are monitored through changes in the size of the condensate after a time-of-flight measurement. Because the background scattering length is close to zero, blue detuning of the OFR laser with respect to a photoassociative resonance leads to increased interaction energy and a faster condensate expansion, while red detuning triggers a collapse of the condensate. The results are modeled with the time-dependent non-linear Gross-Pitaevskii equation.

In the second experiment, we access a regime of coherent one-color photoassociation in which we observe atom-molecule Rabi oscillations and create condensates of excited-state molecules. We attain atom-molecule Rabi frequencies that are comparable to decoherence rates by driving photoassociation of atoms in an $^{88}\text{Sr}$ condensate to a weakly bound level of the metastable $^1S_0 + ^3P_1$ molecular potential, which has a long lifetime and a large Franck-Condon overlap integral with the ground scattering state. Transient shifts and broadenings of the excitation spectrum are clearly seen at short times, and they create an asymmetric excitation profile that only displays Rabi oscillations for blue detuning from resonance.
Acknowledgments

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List of Publications

[1] M. Yan, B. J. DeSalvo, Y. Huang, P. Naidon, T. C. Killian, 
*Rabi Oscillations between Atomic and Molecular Condensates Driven with Coherent One-Color Photoassociation*


*Numerical Modeling of Collisional Dynamics of Sr in an Optical Dipole Trap.*


*Degenerate Fermi Gas of $^{87}$Sr.*


*Bose-Einstein Condensation of $^{84}$Sr.*


*Bose-Einstein Condensation of $^{88}$Sr through Sympathetic Cooling with $^{87}$Sr.*


Two-photon Photoassociative Spectroscopy of Ultracold $^{88}$Sr.


*Repumping and Spectroscopy of laser-cooled Sr Atoms using the $(5s5p)^3P_2 - (5s4d)^3D_2$ Transition.*


*Inelastic and Elastic Collision Rates for Triplet States of Ultracold Strontium.*

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Chapter 1

Introduction and Background

1.1 Introduction

It has been nearly two decades since the first achievement of the quantum degenerate atomic gases in 1995 [1–3]. Most experiments form degenerate gases of alkali-metal atoms. Recently, alkaline-earth-(like) elements joined the quantum degenerate family [4–15], and they have attracted significant interest experimentally and theoretically. Table 1.1 lists all quantum degenerate gases produced with alkaline-earth-(like) elements.

The divalent electronic structure of alkaline-earth-(like) elements gives rise to extremely narrow dipole-forbidden intercombination transitions between singlet ground states and triplet states. There are various application of these transitions including the further laser cooling process which is crucial to the achievement of quantum degeneracy [4, 5, 8], the next generation of optical clocks [16], and quantum information architectures [17]. The absence of hyperfine structure in ground states of alkaline-earth-(like) elements makes them immune to external fields, which simplifies the experimental design and theoretical calculation [18, 19].

Our work focuses on strontium, which has four stable isotopes, three bosons and one fermion, allowing diverse choices of mixtures in experiments. For example, although the extremely small interatomic scattering length of $^{88}\text{Sr}$ of $-2 \ a_0$ [20] results
Table 1.1: Summary of quantum degenerate of alkaline-earth-(like) elements. Inter-atomic s-wave scattering lengths $a$ are in units of Bohr radii $a_0 = 0.053$nm.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Particle</th>
<th>Abundance</th>
<th>$a [a_0]$</th>
<th>Group</th>
<th>Year</th>
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<tr>
<td>$^{88}$Sr</td>
<td>Boson</td>
<td>82.58%</td>
<td>-2</td>
<td>Killian (USA) [4]</td>
<td>2010</td>
</tr>
<tr>
<td>$^{87}$Sr</td>
<td>Fermion</td>
<td>7.00%</td>
<td>96</td>
<td>Killian (USA) [5]</td>
<td>2010</td>
</tr>
<tr>
<td>$^{86}$Sr</td>
<td>Boson</td>
<td>9.86%</td>
<td>800</td>
<td>Schreck (Austria) [6]</td>
<td>2010</td>
</tr>
<tr>
<td>$^{84}$Sr</td>
<td>Boson</td>
<td>0.56%</td>
<td>123</td>
<td>Schreck (Austria) [7]</td>
<td>2009</td>
</tr>
<tr>
<td>$^{40}$Ca</td>
<td>Boson</td>
<td>96.94%</td>
<td>50 - 300</td>
<td>Sterr &amp; Riehle (Germany) [9]</td>
<td>2009</td>
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<tr>
<td>$^{176}$Yb</td>
<td>Boson</td>
<td>12.6%</td>
<td>-24</td>
<td>Takahashi (Japan) [10]</td>
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<td>31.6%</td>
<td>105</td>
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<td>16.2%</td>
<td>199</td>
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<td>2007</td>
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<td>-3</td>
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<tr>
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<td>3.1%</td>
<td>64</td>
<td>Takahashi (Japan) [14]</td>
<td>2007</td>
</tr>
<tr>
<td>$^{168}$Yb</td>
<td>Boson</td>
<td>0.135%</td>
<td>252</td>
<td>Takahashi (Japan) [15]</td>
<td>2011</td>
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in poor evaporative cooling efficiency in an optical dipole trap (ODT), $^{88}\text{Sr}$ can be sympathetically cooled with the aid of $^{87}\text{Sr}$ to achieve the Bose-Einstein condensate (BEC) due to the large and positive scattering length between $^{88}\text{Sr}$ and $^{87}\text{Sr}$ [4]. Furthermore, the isolated, large nuclear spin $I_{\text{nuclear}} = 9/2$ in $^{87}\text{Sr}$ resulting in a $\text{SU}(2I_{\text{nuclear}}+1)$ symmetry, opens a door to test a series of theoretical models, e.g. the Hubbard model [21–24] and its special-condition cases including the Heisenberg model [25, 26], the Kugel-Khomskii model [27], and the Kondo model [28, 29], and facilitates many experiments, like observing a chiral spin liquid [30], $\text{SU}(N)$ symmetry breaking states [31], and unconventional antiferromagnets [32].

In this thesis I will present two experiments with $^{88}\text{Sr}$ BEC related to photoassociation (PA) using the narrow intercombination transition $^1S_0^{-3}P_1$ with the line width of $\Gamma/2\pi = 7.5\text{kHz}$: the first one is controlling collapse and expansion of $^{88}\text{Sr}$ BECs with an optical Feshbach resonance (OFR); the second one is attaining atom-molecule Rabi oscillations and creating condensates of excited-state molecules.

1.2 Background

1.2.1 Photoassociation

PA [33] is a phenomenon in which two colliding atoms absorb a photon to create a bound, electronically excited molecule. Figure 1.1 shows the PA process of Sr: the bottom one is the $^1S_0^{-1}S_0$ ground state potential, the long-range part of which is described by the $R^{-6}$ van de Waals term, and the top one is the excited molecular $^1S_0^{-3}P_1$ potential whose interaction at large separation is given by the $R^{-3}$ dipole term. Two free ultracold Sr atoms in the ground state $^1S_0$ with the thermal collision
energy of $E_g$ approach each other, and absorb a photon with the energy of $h\nu$ to photoassociate to a molecular bound state of the excited potential around the Condon point $R_c$. The bound state $|\Psi_e\rangle$ has the binding energy of $E_b$, and the Condon point $R_c$ is the interatomic separation where PA occurs most likely which is characterized by the Franck-Condon overlap integral discussed later. The formation of the molecule is followed by spontaneous or stimulated decays, usually creating either two free atoms with high kinetic energy or ground state molecule, both of which usually are lost due to extra high energies. Detecting the loss of atoms is a typical method to perform PA.

According to the conventional treatment [33, 34], the strength of the PA transition is proportional to the Franck-Condon overlap integral between wave functions of ground-collisional and excited bound state. Since the wave functions oscillate fast at the short interatomic separation, the Franck-Condon overlap integral is dominated by the amplitude of the ground-state wave function near the Condon point $R_c$ [33]. It is noticed that in $^{88}\text{Sr}$ the Franck-Condon overlap between the second least bound state on the $^1S_0+^3P_1$ potential and the ground-collisional state is large, which is a crucial feature to the applicability of experiments discussed in this thesis.

PA has proven to be a very powerful tool in ultracold physics, such as determining absolute binding energies, extracting atomic collisions information, modifying collision strength, and making ultracold molecules [33].

### 1.2.2 Optical Feshbach Resonance

The ability to tune interactions in ultracold atomic gases makes these systems ideal for exploring many-body physics [35] and has enabled some of the most important recent
Figure 1.1: Schematic diagram of the PA process of Sr. By absorbing a photon from the 689 nm laser field, a pair of Sr atoms in the ground state $^1S_0$ are excited to the $|{^1S_0 + ^3P_1}\rangle$ excited molecular bound state $|\Psi_e\rangle$ with the binding energy of $E_b$. Accompanying spontaneous or stimulated decays lead to the loss of atoms. $R_c$ is the Condon point around which PA occurs. Refer to the text for the definition of other notations.
advances in atomic physics, such as investigation of the Bose-Einstein condensate -
Bardeen-Cooper-Schrieffer crossover regime [35] and creation of quantum degenerate
molecules [36, 37]. Magnetic Feshbach resonances (MFRs) [38] are the standard tool
for changing atomic interactions, and they have proven incredibly powerful.

In MFRs, there is an open channel describing two free ultracold atoms colliding
near a resonant closed channel that is a molecular bound state. The energy difference
between these two channels is $\Delta E$. If the open and closed channels have different
magnetic moments, $\Delta E$ can be controlled by external magnetic fields which leads
to the modification of the interaction between atoms. However, MFRs only allow
low-frequency spatial and temporal modulation due to the methods for generating
magnetic fields, and do not occur in samples with non-degenerate ground states, such
as alkaline-earth-metal atoms.

Alternatively, an optical Feshbach resonances (OFR), which modulates inter-
atomic interactions by coupling two colliding atoms to a bound molecular state of
an excited potential with photoassociation [39], can overcome these limitations, al-
though it has limitations of its own. In ultracold physics, the first OFR experiments
were performed using strong dipole-allowed transitions of alkali-metal atoms [40–42],
and they were always accompanied by huge loss of atoms, which makes it unpractical
as a tool for controlling the properties of ultracold gases. However, the intercombi-
nation transition in alkaline-earth-(like) atoms allows changing the interaction with
much smaller loss rate, which makes OFRs more promising. Optical Feshbach res-
onances may open new avenues of research in nonlinear matter waves [43–45] and
quantum fluids [46–48], and could be very valuable for experiments with fermionic
alkaline-earth atoms [5, 49] in lattices [23].
1.2.3 Coherent One-Color Photoassociation

Coherent conversion of atoms into molecules in quantum degenerate gases provides a path to create molecular condensates, and it is also of fundamental interest because of the complex dynamics of many-body systems with nonlinear couplings [50].

Magnetoassociation is a standard tool for studying coherent atom-molecule coupling [51–54]. Donley et al. reported the first coherent atom-molecule oscillations in a BEC by using a time-dependent magnetic field near a MFR. Subsequently, a series of experiments observed Rabi oscillations in $^{85}$Rb thermal bosons [53], bosonic-atom pairs in a Mott insulator state in an optical lattice [55], and a Bose-Fermi mixtures [56].

Alternatively, PA [33] is another mature technique for creating ultracold molecules, and the description of PA in a dressed picture (Sec. 3.1.2) reveals its similarity to magnetoassociation [38]. But so far, only two-color PA has been employed to form a ground-state molecule which is coupled to atomic colliding states by two laser fields through an intermediate level [57–63]. Ref. [64] theoretically studied the parameter regime for coherent one-color PA in alkali-metal and alkaline-earth systems. As far as electric-dipole allowed transitions in alkali-metal atoms are concerned, a short lifetime of the excited molecular state limits these experiments to the adiabatic regime, and the coherent regime is unaccessible. Furthermore, giant Rabi oscillations between atomic and molecular condensates have not been observed in experiments, although they have been studied since the earliest days of the study of quantum gases [64–75].

But this is not the case in an alkaline-earth system where a long-lived $^3P_1$ state exists. Ref. [64] mapped the parametric ranges of the accessible coherent regime for
strontium. Here we demonstrate coherent, one-color PA and Rabi oscillations between atomic and molecular $^{88}$Sr condensates. We also observe transient shifts and broadenings of the excitation spectra at short times and indications of universal dynamics on resonance [64], all of which have not been experimentally studied previously. We access the coherent regime by photoassociating atoms in an $^{88}$Sr condensate [4] to a weakly-bound level of the metastable $^1S_0+^3P_1$ molecular potential. This yields a long molecular lifetime and large Franck-Condon overlap integral between ground and excited states, which allows atom-molecule coupling to exceed loss and decoherence rates.

1.3 Thesis Outline

The structure of this thesis is as follows: In Chapter 2, I will introduce the experimental methods used in these two experiments. Since the procedure of achieving $^{88}$Sr BECs has been described in detail [76, 77], I will briefly introduce it and focus on the OFR and coherent PA procedures. In Chapter 3 I will present all aspects of the OFR experiments including the previous works about OFRs, the theoretical model, experimental details, characterization of the apparatus, and the experimental results, which are modeled well with the time-dependent nonlinear Gross-Pitaevskii equation. In Chapter 4, a comprehensive description of the coherent PA experiment will be detailed, and the structure of this chapter is similar to that of Chapter 3. In Chapter 5, for the future application of other isotopes, the PA resonances of $^{86}$Sr are carefully studied. Chapter 6 is the conclusion.
Chapter 2

Experimental Methods

This chapter describes experimental techniques used in experiments on an optical Feshbach resonance and coherent photoassociation in $^{88}\text{Sr}$, the most abundant isotope of strontium. As shown in Table 1.1 in Chapter 1, the $s$-wave scattering length of $^{88}\text{Sr}$ of $-2a_0$ [78] is so small that the evaporative cooling efficiency in an optical dipole trap is too poor to achieve a BEC by itself. Fortunately, $^{88}\text{Sr}$ can be sympathetically cooled by the fermionic isotope $^{87}\text{Sr}$ efficiently. The scattering length of non-polarized $^{87}\text{Sr}$ of $96a_0$ is large for efficient thermalization and evaporation, and the reasonable inter-isotope scattering length between $^{88}\text{Sr}$ and $^{87}\text{Sr}$ of $55a_0$, makes $^{87}\text{Sr}$ a good refrigerant to help cool down $^{88}\text{Sr}$ in an ODT. In this way, we achieved the first $^{88}\text{Sr}$ BEC in 2010 [4]. The procedure of creating $^{88}\text{Sr}$ BEC has been detailed in Refs. [4, 79]. In this chapter I will introduce the preparation of $^{88}\text{Sr}$ BEC briefly, and focus on the generation of the OFR/PA beams and corresponding procedures related to the two experiments. The comprehensive description of experimental procedures will be given in Chapter 3 and 4.

2.1 Preparation of BEC

Figure 2.1 is the schematic of all laser beams involved in our experiments. The timing of each laser will be introduced in Chapter 3 and 4. Figure 2.2 is the simplified partial
Figure 2.1: Schematic of all laser beams involved in experiments. Atoms are cooled and trapped in the 461 nm and 689 nm MOTs in turn, and then transferred to the ODT/dimple traps where the BEC is achieved via evaporative cooling. The OFR/PA beams are applied at the end of the evaporation procedure. The timing of OFR and PA beams are a little different which will be described in Sec. 2.2.
energy level diagram for strontium without the hyperfine structure of fermionic $^{87}$Sr. Sr atoms are decelerated by a Zeeman slower beam and then trapped in a magneto-optical trap (MOT) operating on the $^1S_0 - ^1P_1$ transition of 461 nm. The lifetime of this $^1P_1$ state is only 5 ns allowing fast cooling of atoms. It is noticed that the $^1P_1$ state can decay to $^3P_1$ and $^3P_2$ through $^1D_2$ with the branching ratio of $2 \times 10^{-5}$. Atoms at the $^3P_1$ state decay back to the ground state $^1S_0$ to rejoin the cooling cycle, whereas atoms in the dark state $^3P_2$ with the long lifetime of 9 minutes [80] is immune to the 461 nm laser and can be trapped by the quadrupole magnetic field of the MOT [81–84]. The collection of atoms in $^3P_2$ continues as the MOT cooling and trapping is performed. The ability of accumulating atoms in $^3P_2$ helps overcome the difficulty of achieving the BEC of $^{84}$Sr whose natural abundance is only 0.6% [8], and makes the trapping of mixtures of isotopes possible. After several hundred milliseconds loading of $^{88}$Sr to the quadrupole field, the frequency of the cooling laser is switched to collect $^{87}$Sr atoms. A typical time of 8 s is needed to load a sufficient number of $^{87}$Sr. Atoms in the $^3P_2$ state are repumped to the state $^3D_2$ by a 3.0 $\mu$m laser with the intensity of 3 W/cm$^2$ [78], followed by decaying back to the ground state $^1S_0$ via state $^3P_1$. In OFR experiments, we load $^{88}$Sr for several hundred milliseconds and then $^{87}$Sr for 20 seconds. Typically, $2.2 \times 10^8$ $^{87}$Sr and nearly the same number of $^{88}$Sr atoms are trapped in this 461 nm MOT with the temperature of a few mK. The loading time of $^{88}$Sr is fine adjusted everyday so that the same numbers of $^{88}$Sr and $^{88}$Sr atoms are prepared for evaporative cooling in the ODT. In later coherent PA experiments with the new dimple trap, the loading time of $^{87}$Sr is only 8 seconds, which shortens the experimental period significantly.

There is a featured intercombination transition with $\Delta S = 1$ in Sr, where $S$ is
Figure 2.2: Simplified partial energy level diagram for Sr laser cooling and trapping related to transitions introduced in the text. The unique hyperfine structures of $^{87}$Sr are not shown. The laser cooling transitions with the wavelength of 461 nm (blue) and 689 nm (red) are labeled with the line widths and lifetime. The repumping transition with the wavelength of 3.0 $\mu$m is shown too. Arrows with solid line are laser induced transition, and those with dashed line are natural decay.
spin. Usually, the electric dipole (E1) operator does not connect $\Delta S = \pm 1$ transitions, and E1 transitions can occur only between states of the same $S$. It is true for small-mass atoms where the L-S coupling holds, and the spin-orbit coupling is small. However, for large-mass atoms such as strontium, the increasing spin-orbit coupling will mix singlet and triplet states of the same electronic term, e.g. $(5s5p)^1P_1$ and $(5s5p)^3P_1$ in strontium. In this case, transitions with $\Delta S = 1$ (intercombination lines) are weakly allowed, resulting in extremely narrow line widths. As we know, the Doppler cooling limit is proportional to the line width of the cooling transition, which dominates the temperature limit of Sr atoms in the 461 nm MOT. In strontium, the intercombination transition $^1S_0-^3P_1$ with the narrow line width of 7.5 kHz, which is three orders narrower than that of $^1S_0-^1P_1$ of 32 MHz, has the potential to cool atoms three orders colder than the $^1S_0-^1P_1$ transition can do.

Thus, after the 461 nm MOT laser is extinguished, the 689 nm lasers driving the $^1S_0-^3P_1$ transition in each isotope are turned on to form intercombination-line MOTs for $^{88}$Sr and $^{87}$Sr, respectively. The setting parameters of 689 nm lasers for both isotopes are documented in Refs. [4, 79]. About 30% of the atoms in the 461 nm MOTs are captured in the intercombination-line MOTs. After 200 ms of intercombination-line cooling, an optical dipole trap composed of two crossed 1064 nm laser beams is applied and overlapped with the intercombination-line MOTs for 100 ms. In this stage, atoms are further cooled and transferred to the ODT. Immediately after turning off the 689 nm beams, the ODT beams are ramped to about 8 W within 30 ms to capture as many atoms as possible. The ac Stark shift of the intercombination transition induced by the ODT laser with a power of 8 W is up to 200 kHz, and spatially dependent. Typically, there are both $3 \times 10^6$ $^{88}$Sr and $^{87}$Sr atoms with the temperature
of $3\, \mu K$ in the ODT prepared for the following evaporative cooling process.

As mentioned before, $^{88}\text{Sr}$ is sympathetically cooled by $^{87}\text{Sr}$ to achieve BECs. In OFR experiments performed earlier, a typical evaporation time of 10 s is needed. In the later coherent PA experiments, a superimposed dimple trap is added to improve the evaporation efficiency to shorten the evaporation time by half to about 5 s. The design and characterization of the new trap is detailed in Ref. [85], and I will introduce it briefly in Sec. 4.3. The number of $^{88}\text{Sr}$ atoms, the size of clouds, and the sample temperature are determined with time-of-flight absorption imaging using the $^{1}S_{0}^{1}P_{1}$ transition at 461 nm (Sec. 2.3). Figure 2.3 shows false color 2-dimensional renderings (left) and 1-dimensional slices through (right) the time-of-flight absorption images recorded after 16 ms or 22 ms of expansion for various points along the evaporation trajectory. At the end of the evaporation, we create condensates with $7000 - 9000$ $^{88}\text{Sr}$ atoms. The $1/\sqrt{e}$-density radius of the condensate is about 0.6 $\mu m$, and the peak density $n_0$ is on the order of $10^{15}$ cm$^{-3}$. It is noticed that the loading time of $^{88}\text{Sr}$ and $^{87}\text{Sr}$ to the quadrupole magnetic field is adjusted everyday to get a sufficiently high temperature of the thermal atoms of 100 nK, so that the bimodal structure can be distinguished clearly in the time-of-flight measurement.

### 2.2 Setup of OFR / PA Laser

The OFR and coherent PA experiments are both performed near the second least bound state supported by the $^{1}S_{0}^{+3}P_{1}$ molecular potential which has the binding energy of $h \times 24$ MHz [86]. Both the OFR laser and the PA laser are generated from the same setup (Fig. 2.4).
Figure 2.3: Appearance of Bose-Einstein condensation in absorption images (left) and areal density profiles (right). Data correspond to 16 ms (bottom) or 22 ms (top three) of free expansion after indicated evaporation times (t). Images on the left have the same time stamp as on the right. The areal density profiles are from a vertical cut through the center of the atom cloud, and temperatures are extracted from 2D Bose-Einstein distribution fits to the thermal pedestal. At 7.5 s, a bimodal distribution is evident, indicative of Bose-Einstein condensation. A pure condensate is shown at 9 s of evaporation. The Maxwell-Boltzmann distribution does not accurately describe low velocity atoms near degeneracy, and the Bose-Einstein distribution does not describe the condensate contribution. So in these respective regimes, a central region slightly larger than the condensate radius is excluded from fitting. For bimodal data, fugacity is constrained to 1 [4, 76]. Figure from Ref. [4].
Figure 2.4: Schematic drawing of 689 nm laser system. The 689 nm MOT beam and the OFR/PA beams are all generated here. A 689 nm external-cavity diode laser serves as a master laser and injection locks slave laser 1. A small part of the master laser goes to the high-finesse cavity. The output beam of the slave laser 1 is 90 MHz blue detuned with respect to the $^1S_0 - ^3P_1$ atomic transition, controlled by the saturated-absorption system. Beams travelling into the two acousto-optic modulators (AOMs) are both from the slave laser 1. The MOT beam is formed from the negative first order of AOM 1. The OFR/PA beams are around -24 MHz red detuned with respect to the atomic transition via the negative first order of AOM 2 driven by the RF signal of around 114 MHz, and transported to the atoms through a single-mode optical fiber.
The OFR/PA beams are derived from a 689nm master-slave diode laser system that has a linewidth of approximately 10 kHz. Short term stability is provided by locking the laser frequency to a moderate finesse \( F = 2000 \) optical cavity, and long term stability is assured through saturated absorption spectroscopy of the \( ^1S_0 - ^3P_1 \) atomic transition in a vapor cell. As shown in Fig. 2.4, the output beam of the slave laser 1 is 90 MHz blue detuned with respect to the \( ^1S_0 - ^3P_1 \) atomic transition, resulting in the same detuning of lasers into the two acousto-optic modulators (AOMs). The MOT beam is formed from the negative first order of AOM 1. The OFR/PA beams are around -24 MHz red detuned with respect to the atomic transition via the negative first order of AOM 2 driven by the RF signal of around 114 MHz, and transported to the atoms through a single-mode optical fiber.

The experimental parameters of OFR/PA beams are different. The waist of the OFR beam is 725 \( \mu m \) which is much larger than the condensate dimension, and the intensity of the beam is typically 0.057 W/cm\(^2\). The OFR laser is applied to the condensate 20 \( \mu s \) before extinguishing the ODT and left on for a variable time \( \tau \) on the order of ms during expansion of atom clouds (Refer to Sec. 3.5 for details). In one-color coherent PA experiments, the PA beam has a waist of 200 \( \mu m \) and the intensity used is up to 2.4 W/cm\(^2\). As introduced before, a dimple trap is added, and the relative timing between the PA beam and dimple beam is different from that in OFR experiments. The PA laser here is applied for a time of several \( \mu s \) while the dimple beam is still on (Refer to Sec. 4.3 for details). The PA beam and the dimple beam are turned off simultaneously to allow the expansion of the cloud for the time-of-flight measurement.
2.3 Imaging Techniques

A traditional time-of-flight absorption imaging technique is employed here to determine the number and temperature of atoms. After a drop time \( t = 10^{-35} \) ms, an atom cloud is imaged by a \( \lambda_I = 461 \) nm laser with the incident intensity \( I_0 \), which corresponds to the \( ^1S_0 - ^1P_1 \) transition. According to Beer’s law, the 2D distribution of the transmitted intensity \( I_t(x, y) \) recorded by a CCD camera is given by

\[
I_t(x, y) = I_0(x, y) \exp[-OD(x, y)],
\]

where the optical depth \( OD(x, y) \) is calculated by integrating the product of the sample density distribution \( n(x, y, z) \) and the absorption cross section \( \alpha(\delta_I) \) along \( z \) axis (the direction of the incident imaging beam):

\[
OD(x, y) = \alpha(\delta_I) \int n(x, y, z)dz.
\]

The absorption cross section \( \alpha(\delta_I) \) depends on \( \delta_I \), the laser detuning with respect to the atomic resonance:

\[
\alpha(\delta_I) = \frac{3\lambda_I^2}{2\pi} \frac{1}{1 + I/I_{\text{sat}} + (2\delta_I/\Gamma)^2},
\]

with the saturation intensity \( I_{\text{sat}} = 42 \) mW/cm\(^2\) for this transition, and the intensity of imaging beam \( I \).

The number of atoms is given by

\[
N = \iiint n(x, y, z)dxdydz = \iint OD(x, y)dxdy / \alpha(\delta_I),
\]

where the second equality is obtained by substituting \( \int n(x, y, z)dz = OD(x, y)/\alpha(\delta_I) \) given by Eq. 2.2.
In each cycle of measurement, firstly we obtain $I_t(x,y)$ by taking an absorption image with atoms, and then $I_0(x,y)$ is measured after the atom cloud falls outside the field of view of the camera. According to Eq. 2.1, we can get

$$OD(x, y) = \ln \left( \frac{I_0(x,y)}{I_t(x,y)} \right). \tag{2.5}$$

Substituting Eq. 2.5 into Eq. 2.4 yields

$$N = \frac{1}{\alpha(\delta_t)} \iint \ln \left( \frac{I_0(x,y)}{I_t(x,y)} \right) dxdy. \tag{2.6}$$

Alternatively, $N$ can also be extracted by fitting $OD(x, y)$ with a 2D gaussian function

$$OD(x, y) = \frac{\alpha(\delta_t)N}{2\pi\sigma_x\sigma_y} \exp \left[ -\frac{x^2}{2\sigma^2_x} - \frac{y^2}{2\sigma^2_y} \right], \tag{2.7}$$

where the fitting parameters $\sigma_x$ and $\sigma_y$ are the rms widths of the atom cloud along $x$ and $y$ axes, respectively.

The expansion of the cloud can be described by [87]

$$\sigma_i^2 = \sigma_{0i}^2 + \left( \frac{k_B T_i}{m} \right) t^2, \quad (i = x, y) \tag{2.8}$$

with the initial size of the cloud $\sigma_{0i}$, the Boltzmann constant $k_B$, the mass of the atom $m$ and the sample temperature $T_i$. So temperature $T_i$ can be obtained with given $\sigma_i$ and $t$. 
Chapter 3

Optical Feshbach Resonance

3.1 Introduction and Background

Feshbach resonances, named after Herman Feshbach, were first studied by Herman Feshbach in nuclear physics [88], and Ugo Fano in atomic physics [89] independently. When two colliding particles have collision energy nearly equal to the energy of a quasi-bound state, the scattering amplitude is enhanced, i.e. a Feshbach resonance occurs. In 1993, before the first achievement of the BEC, Tiesinga et al. [90] proposed the possibility of modifying the atomic interaction in ultracold physics using a Feshbach resonance. A couple of years later, Inouye et al. [91] observed the magnetic Feshbach resonance (MFR) in a BEC for the first time.

3.1.1 Magnetic Feshbach Resonance

Nowadays, MFRs [38], which are the standard tool for changing atomic interactions, have proven incredibly powerful in ultracold physics, such as investigation of the Bose-Einstein condensate-Bardeen-Cooper-Schrieffer crossover regime [35], and creation of quantum degenerate molecules [36, 37]. Figure 3.1 illustrates the basic picture of the magnetic Feshbach resonance: two free ultracold atoms with the energy $E_g$ collide in the open channel. The closed channel supports a molecular bound state. Since the magnetic moments of the open and closed channels are different, the energy difference
Figure 3.1: Schematic diagram of the magnetic Feshbach resonance between the scattering state of the open channel with the energy $E_g$ and the bound state of the closed channel. The energy difference $\Delta E$ can be adjusted by an external magnetic field. A MFR occurs when the energy difference $\Delta E \rightarrow 0$, which results in the modification of the interatomic interactions.
\( \Delta E \) between the scattering state of the open channel and the molecular state in the closed channel can be controlled by an external magnetic fields \( B \). MFR occurs when \( \Delta E \to 0 \), i.e. these two channels are coupled, which leads to the modification of the interaction between atoms. The \( B \)-dependence of the \( s \)-wave scattering length \( a \) is [38]

\[
a(B) = a_{bg} \left( 1 - \frac{\Delta}{B - B_0} \right),
\]

(3.1)

where \( a_{bg} \) is the background scattering length without magnetic fields, \( \Delta \) is the resonance width, and \( B_0 \) is the resonance value where \( a \) diverges.

However, MFRs are limited because the methods for creating magnetic fields make high-frequency spatial and temporal modulation difficult. Also, in atoms with non-degenerate ground states, such as alkaline-earth-metal atoms, magnetic Feshbach resonances do not exist.

### 3.1.2 Optical Feshbach Resonance

Some of the limitations of MFRs can be overcome by using an optical Feshbach resonance (OFR), which tunes interatomic interactions by coupling a colliding atom pair to a bound molecular level of an excited state potential with photoassociation [39]. Analogous to MFRs, optical Feshbach resonances can be described in the similar way in dressed state pictures. Taking Sr for instance, the dressed state picture corresponding to Fig. 1.1 is shown in Fig. 3.2. The ground state is composed of two \( ^1S_0 \) ground state atoms and \( n \) photons, and the excited state consists of state \( |\Psi_e\rangle \) (Fig. 1.1) and \( n - 1 \) photons. The energy difference \( \Delta E \) between these two states can be controlled by the detuning of the PA laser. Similarly, when \( \Delta E \to 0 \), an OFR is
induced producing a modification of interaction between the atoms.

Figure 3.2: Optical Feshbach resonance described by the dressed state picture corresponding to Fig. 1.1. The initial state represents a pair of atoms in the $^1S_0$ ground state with $n$ photons, and the closed channel state is the bare molecular bound state $|\Psi_e\rangle$ (Fig. 1.1) with $n-1$ photons. The energy difference $\Delta E$ can be adjusted by a laser field. An OFR occurs modifying interaction between the atoms when the energy difference $\Delta E \to 0$.

Tuning the interaction of ultracold atoms using an OFR was proposed by Fedichev et al. [39] initially. Later, Bohn et al. [92, 93] further studied the OFR theoretically. Julienne et al. [94, 95] pointed out that when the laser detuning from the molecular resonance $|\delta|$ is much larger than $\gamma_m$, the natural linewidth of the excited molecular level, the OFR-induced scattering length $a_{opt}$ is proportional to $|\langle \Psi_e|\varepsilon_r\rangle|^2 \gamma_m/\delta$, ac-
companied by the loss rate proportional to $|\langle \Psi_r | \varepsilon_r \rangle|^2 (\gamma_m / \delta)^2$, where $\varepsilon_r$ is the kinetic energy of the colliding atom pair. I will discuss this model in detail in Sec 3.2.1.

Early experiments on OFRs [40–42] used strong dipole-allowed transitions in alkali-metal atoms to alter the atomic collision properties. The first OFR was observed in an ultracold thermal sodium vapor [40], where one-color PA was performed, and the modification of the scattering wave function induced by the OFR was demonstrated via the change of the PA transition rate proportional to the photo-ionization signals. Theis and Thalhammer et al. [41, 42] directly measured the OFR-driven atomic losses in $^{87}$Rb BECs via the one-color PA [41] and two-color Raman transitions [42]. Bragg spectroscopy [96] was used to determine the change of the scattering length here. All these OFR experiments using strong dipole-allowed transitions in alkali-metal atoms were always accompanied by rapid atom losses, which made OFRs difficult to use.

Tuning of interactions in alkali-metal atoms, but with a smaller atom loss, was recently obtained with a magnetic Feshbach resonance using an ac Stark shift of the closed channel to modify the position of the resonance [97–99]. Recently, a multiple-laser optical method was proposed for wider modulation of the interaction strength near a magnetic Feshbach resonance [100]. Unfortunately, none of these hybrid variations are feasible for atoms lacking magnetic Feshbach resonances, such as alkaline-metal-(like) systems.

In alkaline-metal-(like) atoms, such as strontium and ytterbium, an OFR can be induced by a laser tuned near a weakly allowed intercombination transition featuring small $\gamma_m$, which can modify the interatomic interactions with significantly less induced losses than those in alkali-metal atoms, as predicted by Ciurylo et al. [94, 95]. Near the intercombination transitions, molecular states, even weakly bound molecular states,
are spaced by more than thousands of $\gamma_m$ (so that $\gamma_m/\delta \ll 1$) and still have large $|\langle \Psi_e|\varepsilon_r \rangle|$. In this case, the scattering length can be varied significantly with very small loss of atoms. In alkali atoms, it is difficult to satisfy the small $\gamma_m/\delta$ and large $|\langle \Psi_e|\varepsilon_r \rangle|$ simultaneous due to the large $\gamma_m$ near dipole-allowed transitions. Thus significant OFR-induced modification of the scattering length in alkali atoms is always accompanied by large loss rates.

Intercombination-transition OFRs have been observed in a series of experiments [101–103]. Similar to the first OFR experiment [40], Enomoto et al. [101] used an OFR to control the scattering wave function in a thermal gas of Yb, which was monitored by the change of photoassociation rate. Two years later, in the same group, submicron-spatial modulation of the interaction between atoms was demonstrated by monitoring modulated mean-field energy of atoms in a Yb BEC induced by an OFR-laser standing wave [102]. Small detunings from a molecular resonance were used in this work ($|\delta| < 10 \gamma_m$, where $\gamma_m$ is the natural decay rate of the excited molecular level), which led to short sample lifetimes on the order of microseconds. Afterwards, Blatt et al. [103] reported the modulation of thermalization and loss rates in a thermal gas of $^{88}$Sr. Longer exposure times and detunings $|\delta| < 50 \gamma_m$ were used [103], but at much lower atomic density than typically found in a degenerate sample.

There is great interest in intercombination-line OFRs at much larger detuning in quantum degenerate gases of divalent atoms [4, 5, 7, 8], with the goal of modifying the scattering length and still maintaining sample lifetimes on the order of dynamical timescales of quantum fluids [47, 48]. In our work, we tune a laser near the PA transition to the second least bound state on the $^1S_0+^3P_1$ molecular potential, which induces an OFR to control collapse and expansion of an $^{88}$Sr condensate during time-
of-flight measurements. $^{88}\text{Sr}$ has an $s$-wave background scattering length of $a_{bg}$ close to zero [20, 104], which allows convenient modification of the scattering length either positive or more negative, resulting in a faster expansion or a collapse of the condensate, respectively. Changes in scattering length are monitored through changes in the size of the condensate after a time-of-flight measurement. Large relative change in scattering length $a_{opt}/a_{bg} = \pm 10$ is demonstrated, with the loss-rate constant $K_{in} \sim 10^{-12}$ cm$^3$/s comparable to Ref. [99]. We explore $|\delta|$ as large as $500 \gamma_m$, and obtain sample lifetimes of milliseconds during application of the OFR beam.

It is worth emphasizing that the value of $a_{bg}$ is crucial in the following simulations. Our group determined $a_{bg}$ to be $(-1.4 \pm 0.6) a_0$ from the measurement of the binding energy of the least-bound vibrational state of the $^1S_0 + ^3P_1$ molecular potential using two-color PA [20]. Later, Stein et al. extracted the value $a_{bg} = (-2.0 \pm 0.3) a_0$ by Fourier-transform spectroscopy [104]. In the analysis of our OFR work, we will employ $a_{bg} = -2.0 a_0$ following the treatment in the first achievement of $^{88}\text{Sr}$ [4].

3.2 Theory

Descriptions of ultracold collision theory can be found in, for example, Refs. [33, 105, 106]. Assuming a spherical scattering potential $V(r)$, scattering atoms with angular momentum $l$ undergo an effective potential $V(r) + \hbar^2 l(l + 1)/(2 \mu R^2)$ with reduced mass $\mu = m/2$. In the ultracold region, all partial waves with $l > 0$ are suppressed by the centrifugal barrier, and only $s$-wave ($l = 0$) contributes to the collision procedure. In the limit of low energy ($k \rightarrow 0$), the $s$-wave scattering length $a$ characterizes the scattering process. For $a > 0$, it means that the scattering wave
in the scattering potential $V(r)$ is equivalent to scattering from a hard spherical core with the radius $a$. It is noticed that this analogy is valid only if $a$ is positive, indicating the interaction between atoms is repulsive. $a$ is also allowed to be negative, which means the interaction is attractive.

In this section, I will introduce the theoretical models describing the OFR.

3.2.1 Optical Feshbach Resonance

(a) Julienne’s Model

The isolated resonance model [94, 95] was developed by Julienne et al., where a complex scattering length $a_{opt}(\delta, I) = -ib(\delta, I)$ was introduced. An OFR laser of wavelength $\lambda$ and intensity $I$ is detuned by $\delta$ from a PA transition to an excited molecular state $|\Psi_e\rangle$. The real part $a_{opt}$ denotes the modification of the atomic scattering length according to $a = a_{bg} + a_{opt}$ and the imaginary part $b$ is used to derive the two-body inelastic collisional losses described by the loss rate constant $K_{in}$, where

$$a_{opt} = \frac{\ell_{opt} \gamma_m \delta}{\delta^2 + \left(\frac{\eta \gamma_m}{2}\right)^2};$$

$$K_{in} = \frac{2\pi \hbar}{\mu} \frac{\ell_{opt} \eta \gamma_m^2}{\delta^2 + \left(\frac{\eta \gamma_m + \Gamma_{stim}}{4}\right)^2}. \quad (3.2)$$

$K_{in}$ is defined such that it contributes to the evolution of density $n$ as $\dot{n} = -K_{in} n^2$ for a BEC. The optical length $\ell_{opt}$, which characterizes the strength of the OFR, is defined as

$$\ell_{opt} = \frac{\lambda^3 |\langle \Psi_e | \varepsilon_r \rangle|^2 I}{16\pi c \epsilon_r}, \quad (3.3)$$

where $c$ is the speed of light, $I$ is the intensity of the OFR beam, and $k_r$ is the wavenumber for colliding atoms. $|\langle \Psi_e | \varepsilon_r \rangle|^2$ is the Franck-Condon factor per unit en-
ergy for the free-bound PA transition. Following the Wigner threshold law, $|\langle \Psi_e | \epsilon_r \rangle|^2$ is proportional to $k_r$ in the ultracold regime [93], so that $k_r$ in the denominator of Eq. 3.3 is canceled out, and $\ell_{\text{opt}}$ is independent of the collision energy. $\gamma_m = 2\pi \times 15 \text{kHz}$ is the natural linewidth of the excited molecular level in Sr, and $\Gamma_{\text{stim}} = 2k_r\ell_{\text{opt}}\gamma_m$ is the laser-stimulated linewidth. The parameter $\eta > 1$ accounts for enhanced molecular losses, as observed in previous OFR experiments [41, 103]. Julienne [103] treated $\eta$ as a free phenomenological parameter, and a further theoretical interpretation is needed.

However, the isolated-resonance-model expressions (Eq. 3.2) break down at large detunings from photoassociative resonance. Alternatively, coupled channels calculations was introduced by Julienne et al. [103], where the induced scattering length $a_{\text{opt}}$ crosses zero between PA resonances. At small detunings, coupled channels calculations still can reproduce the results from isolated-resonance calculations well [103].

(b) Model Accounting for the Extra Background Loss $K_b$

In our experiments, the largest detuning $|\delta|$ is around $667\gamma_m$ for this photoassociative transition, which is still much less than the spacing between excited molecular states. Thus we find the isolated-resonance-model expressions (Eq. 3.2) still useful for describing our measurements with the modification. The total loss rate constant is given by $K_{\text{total}} = K_{\text{in}} + K_b$, taking into account a background atom-loss term $K_b$.

$K_b$ can be understood in the following picture. Outside approximately $100\gamma_m$ from photoassociative resonance, the two-body loss is expected to make a transition to a broad background value that varies as $1/\delta_a^2$, where $\delta_a$ is $2\pi$ times the detuning from the atomic resonance [103]. A rigorous theoretical description for loss in this regime
is lacking, but the underlying mechanism is collisions involving a ground state atom and an atom excited in the wings of the atomic line, which is often described with the classical Gallagher-Pritchard model [107–109]. Figure 3.3 illustrates the Gallagher-Pritchard model for collisions between two Sr atoms. Two colliding atoms approaching each other absorb a photon at large interatomic separation and are excited to the attractive excited-state potential. Without decaying, atoms are accelerated along the excited-state potential curve to smaller separation, where a photon with smaller energy than the laser photon is emitted. According to the conservation of energy, the excess energy is converted to the kinetic energy so that atoms cannot be trapped resulting in the loss of atoms. The background loss is described phenomenologically in our regime as $K_b = K_0 \Gamma_{\text{mol}}/(2\delta_a)^2$.

In a coupled channels description, the background loss rate is sensitive to a cutoff atom-atom distance inside of which radiative loss is turned on, which is introduced as an ad hoc parameter. Our measurements could provide some experimental input to determine this cutoff distance.

3.2.2 BEC Dynamics

(a) Gross-Pitaevskii Equation

More than half century ago, Bogoliubov [110] formulated the basic idea of the mean-field theories for a dilute boson gas. The mean-field theories have proven quite effective in quantitatively describing the static and dynamic properties of ultracold atom systems. Starting from this theory, Gross [111] and Pitaevskii [112] derived the
Figure 3.3: Gallagher-Pritchard model for ground-excited-state collisions. A pair of Sr atoms in the $^1S_0$ ground state are excited to the $|^1S_0 + ^3P_1\rangle$ molecular potential by absorbing a laser photon at large interatomic separation. They are accelerated toward each other to a smaller separation. Then both atoms may spontaneously emit a photon red detuned to the laser photon. The resulting extra kinetic energy of atoms could lead to the loss of atoms in the trap.
familiar Gross-Pitaevskii (GP) equation for condensates independently:

\[ i\hbar \frac{\partial}{\partial t} \Phi(r, t) = \left[ -\frac{\hbar^2 \nabla^2}{2m} + V_{\text{ext}}(r) + g|\Phi(r, t)|^2 \right] \Phi(r, t). \quad (3.4) \]

Here \( \Phi(r, t) \) is the time- and position-dependent condensate wave function, \( V_{\text{ext}}(r) \) is the external trap potential, and \( g = 4\pi \hbar^2 a/m \) is the coupling constant related to the \( s \)-wave scattering length \( a \).

Following the treatment of Ref. [113], the Gross-Pitaevskii equation can be obtained via a variational procedure:

\[ i\hbar \frac{\partial}{\partial t} \Phi = \frac{\delta E}{\delta \Phi^*}, \quad (3.5) \]

where the energy \( E \) is defined as

\[ E[\Phi] = \int d\mathbf{r} \left[ \frac{\hbar^2}{2m} |\nabla \Phi|^2 + V_{\text{ext}}(r)|\Phi|^2 + \frac{g}{2} |\Phi|^4 \right]. \quad (3.6) \]

The first term on the right hand is the quantum kinetic energy of the condensate arising from the uncertainty principle, the second term is the harmonic-oscillator energy, and the third term is the mean-field interaction energy.

(b) Variational Calculation

In the attractive interaction case \( (a < 0) \), if the quantum kinetic energy is too small to compensate the attractive energy \([113-115]\), condensates collapse. A variational calculation was proposed to study the dynamics of condensate collapse \([113, 116]\).

To get the ground state, the condensate wave function can be rewritten as \( \Phi(r, t) = \phi(r)e^{-i\mu t/\hbar} \), with the chemical potential \( \mu \) and real \( \phi \) which obeys the normalization rule \( \int d\mathbf{r} \phi^2 = N_0 \) where \( N_0 \) is the total number of condensate atoms. Assuming a
spherical trap $V_{\text{ext}}(r) = m\bar{\omega}^2 r^2/2$ with the geometric average of the angular frequencies $\bar{\omega}$, a variational trial wave function of $\phi(r)$ can be written as

$$\phi(r) = \left(\frac{N_0}{w^3 a_{\text{ho}}^3 \pi^{3/2}}\right)^{1/2} \exp\left[-\frac{r^2}{2w^2 a_{\text{ho}}^2}\right], \quad (3.7)$$

where $w$ is a dimensionless variational parameter relevant to the size of the condensate cloud, and

$$a_{\text{ho}} = \left[\frac{\hbar}{m \bar{\omega}}\right]^{1/2} \quad (3.8)$$

is the harmonic oscillator length, which is an important length scale characterizing the size of the condensate cloud of noninteracting bosons.

Substituting Eq. 3.7 into Eq. 3.6 yields

$$E = \int d\mathbf{r} \left\{ \frac{\hbar^2}{2m} \left[ \frac{N_0 r^2 e^{-r^2/(w^2 a_{\text{ho}}^2)}}{w^7 a_{\text{ho}}^4 \pi^{3/2}} \right] + \frac{m\bar{\omega}^2 r^2}{2} \left[ \frac{N_0 e^{-r^2/(w^2 a_{\text{ho}}^2)}}{w^3 a_{\text{ho}}^3 \pi^{3/2}} \right] - 2\pi \hbar^2 |a| N_0^2 \frac{e^{-2r^2/(w^2 a_{\text{ho}}^2)}}{m w^6 a_{\text{ho}}^6 \pi^3} \right\}$$

$$= \frac{3N_0 \hbar^2}{4mw^2 a_{\text{ho}}^2} + \frac{3N_0 m\bar{\omega}^2 w^2 a_{\text{ho}}^2}{4} - \frac{N_0^2 \hbar^2 |a|}{(2\pi)^{1/2} m w^2 a_{\text{ho}}^3}. \quad (3.9)$$

Replacing $a_{\text{ho}}$ with Eq. 3.8, and we can obtain the energy per particle in units of $\hbar \bar{\omega}$

$$E(w) = \frac{E(w)}{N_0 \hbar \bar{\omega}} = \frac{3}{4} (w^{-2} + w^2) - \xi (2\pi)^{-1/2} w^{-3}, \quad (3.10)$$

with the dimensionless parameter $\xi = N_0 |a|/a_{\text{ho}}$.

Following the illustration of Fig.1 in Ref. [116], Figure 3.4 shows the scaled energy per particle $\bar{E}$ as the function of $w$ (Eq. 3.10) for four different values of $\xi$ [116]. There are local minima $w_{\text{min}}$ in the $\xi = 0.30$ (pink solid) and $\xi = 0.50$ curves (blue dashed), whose $\xi$ are both smaller than the critical value $\xi_c = 0.67$ (red dotted). Stable condensates can exist in these cases at $w = w_{\text{min}}$. If $\xi$ is larger than $\xi_c$, there is no local minimum, e.g. the $\xi = 0.90$ curve (green dot-dashed), which results in collapse.
Figure 3.4: Energy per atom with attractive interactions in an ODT, in terms of $\hbar \omega$, versus the dimensionless parameter $w$ related to the size of the BEC cloud. Curves for four different values of $\xi$ are plotted. The $\xi_{cr} = 0.67$ (red dotted) curve corresponds to the critical condition of BEC collapse. There are minima in curves for $\xi < \xi_{cr}$ (pink solid and blue dashed) where stable BECs exist, and the minimum disappears for $\xi > \xi_{cr}$ (green dot-dashed) where BECs collapse.
of the condensate. It is noticed that the $\xi_{cr} = 0.67$ is close to the value of $\xi_{cr} = 0.58$ given by directly solving the Gross-Pitaevskii equation [117].

To extract the BEC size $\sigma_0$ when $\xi < \xi_{cr}$, firstly we determine $w_{\text{min}}$ by calculating

$$\frac{d\tilde{E}(w)}{dw}\bigg|_{w=w_{\text{min}}} = 0, \quad \text{and} \quad \frac{d^2\tilde{E}(w)}{dw^2}\bigg|_{w=w_{\text{min}}} > 0. \quad (3.11)$$

Now the density distribution of the BEC in the ODT is given by

$$n_{\text{BEC}} = |\phi(r)|^2 = \frac{N_0}{w_{\text{min}}^3 a_{\text{ho}}^3 \pi^{3/2}} \exp\left[-\frac{r^2}{2w_{\text{min}}^2 a_{\text{ho}}^2}\right], \quad (3.12)$$

where the expression of $\phi(r)$ in Eq. 3.7 is used.

In our works we defined the density of the BEC as

$$n_{\text{BEC}} = n_0 \exp\left[-\frac{r^2}{2\sigma_0^2}\right]. \quad (3.13)$$

By comparing Eq. 3.13 with Eq. 3.12, we can determine the BEC size

$$\sigma_0 = w_{\text{min}} a_{\text{ho}}/\sqrt{2}, \quad (3.14)$$

and the peak density of the BEC

$$n_0 = \frac{N_0}{(w_{\text{min}}^3 a_{\text{ho}}^3 \pi^{3/2})}. \quad (3.15)$$

The detail of the variational calculation of BEC properties in the ODT of OFR experiments, and the study of condensates collapse without the trap will be discussed in Sec. 3.3.4.

(c) Energy Conservation

The OFR laser, with a beam waist of 725 $\mu$m, is applied to the condensate 20 $\mu$s before extinguishing the ODT and left on for a variable time $\tau$ during expansion.
The exposure time in the ODT is short enough that the initial density distribution of the condensate reflects the ODT potential and the background scattering length, while the expansion dynamics is sensitive to the interaction energy determined by 

\[ a = a_{bg} + a_{opt}. \]

To obtain a qualitative understanding of the data, one can calculate the total energy immediately after the trap is extinguished by using the condensate energy functional \([113, 118]\) assuming a gaussian density for the BEC in the ODT with the initial size \(\sigma_0\) (Eq. 3.14). The total energy in the absence of the trap can be obtained from Eq. 3.9 by removing the harmonic-oscillator-energy term. When atom losses are negligible, this energy can be equated to the total kinetic energy when the condensate has expanded to a low density to give,

\[
N_0 \frac{3}{2} m \sigma_v^2 = N_0 \frac{3}{8} \frac{\hbar^2}{m \sigma_v^2} + N_0 \frac{g}{2(4\pi)^{3/2} \sigma_0^3}. \tag{3.16}
\]

The first and second terms on the right-hand side are the kinetic energy and interaction energy in the trap before release, respectively. \(\sigma_v\) is the rms velocity, which can be related to the BEC size after a long expansion time \(t\) through \(\sigma = \sigma_v t\). This construction assumes that all interaction energy has been converted into the kinetic energy with no significant atom loss during expansion, and it provides a useful qualitative description of the data. An OFR laser blue detuned near the -24 MHz PA line \([86]\) increases \(a\), leading to more interaction energy and larger expansion velocity and BEC size. Red detuning produces the opposite behavior. When the total energy becomes negative, this simple explanation breaks down, and one observes condensate collapse and significant losses of condensate atoms.
(d) Final Equations

As mentioned before, the OFR laser is turned on $20 \mu s$ before extinguishing the ODT. A calculation based on the time-dependent Gross-Pitaevskii equation is performed by Balasubramanian Ramachandran from Dr. Pu’s group to predict the number of atoms in the BEC and the size of BEC clouds after 35 ms free expansion after extinguishing the ODT. Sec. 3.2.1 describes how the scattering length can be modified by $a_{\text{opt}}$ as well as corresponding 2-body losses characterized by $K_{\text{total}}$. Now the time-dependent Gross-Pitaevskii equation will be modified as

$$i\hbar \frac{\partial}{\partial t} \Phi(r, t) = \left[ -\frac{\hbar^2 \nabla^2}{2m} + V_{\text{ext}}(r) + g|\Phi(r, t)|^2 - i\frac{\hbar}{2}K_{\text{total}}|\Phi(r, t)|^2 \right] \Phi(r, t). \quad (3.17)$$

It is noted that the coupling constant $g = 4\pi\hbar^2a/m$ has the same definition but $a = a_{\text{bg}} + a_{\text{opt}}$ includes the contribution from $a_{\text{opt}}$ arising from the OFR laser. In this calculation, the interaction between thermal atoms and atoms in the BEC are neglected, assuming that $T = 0$. The calculation results will be compared with experimental data in Sec. 3.6.

3.3 Experimental Setup

3.3.1 Description of the Optical Dipole Trap

The design and setup of the optical dipole trap (ODT) has been explained in detail in Refs. [76, 79]. $^{88}\text{Sr}$ and $^{87}\text{Sr}$ cooled and trapped in the 689 nm intercombination MOT with a sample temperature of several $\mu$K are transferred to the ODT with an efficiency of about 10%. The ODT is composed of two crossed beams which are generated from a multimode fiber laser (IPG Photonics) working at 1064 nm with the
output power of up to 21 W. The first order deflection of an AOM at 110 MHz serves as the input beam of the ODT, and the AOM controls the turn on/off and the power modification of the input beam. The input beam traverses the vacuum chamber and then is recycled through the chamber serving as the crossed beam, whose power is 90% of that of the input beam. Both the input and crossed beams are tilted by 10.5° from the horizontal, and the waists of both beams of around 90 μm overlap at the center of the vacuum chamber, where the atom samples are located. The power control of the ODT beams via the AOM allows the evaporative cooling to achieve BECs of 88Sr through sympathetic cooling with 87Sr. Typically, at the end of the evaporation, about 8000 88Sr atoms in the BEC are obtained.

Trap frequencies are crucial parameters of the ODT, which not only can aid us in determining the waists of the ODT beams, but also provide the value of the geometric average of the angular trap frequencies $\bar{\omega}$ for the study of BEC dynamics. The uncertainties of the trap frequencies is one of the primary uncertainty sources in our experiments.

The parametric resonance technique [119] is used to measure trap frequencies of the ODT. The trap depth is modulated periodically by adding a sinusoidal modulation with the frequency of $f_m$ to an AOM controlling the power of the ODT beams. The power of the modulation is up to 10% of the fixed power of the beam. The modulation is applied for hundreds of milliseconds to several seconds to get about 50% atom losses at the end of the modulation. In the harmonic-potential approximation, which is the case of our ODT, there is a characteristic frequency $f_i$ (i = x, y, z) in each principle axis. When the modulation frequency is in resonance with the characteristic
Figure 3.5: Trap frequencies measurements for ODTs with powers of the input beam of 1.4 W, 1.5 W, and 1.7 W, respectively. The trap frequencies ($f_x, f_y, f_z$) are determined to be 61 Hz, 70 Hz, 49 Hz for the 1.4 W trap, 68 Hz, 75 Hz, 55 Hz, for the 1.5 W trap, and 75 Hz, 83 Hz, 56 Hz for the 1.7 W trap. The Y direction is vertical, and X and Z are horizontal. From the data our trap model gives the best-fit waists: 97.5 $\mu$m (horizontal) and 105 $\mu$m (vertical) for the input beam, and 82.5 $\mu$m (horizontal) and 105 $\mu$m (vertical) for the crossed beam. Resonances corresponding $n = 1$ ($||$) and $n = 2$ ($\parallel$) in Eq. 3.18 are observed. Dashed lines are a guides showing the shift of the same resonances with the power of the ODT beams.
frequencies, i.e. satisfying
\[ f_{in} = \frac{2f_i}{n}, \quad (n = 1, 2, ...), \tag{3.18} \]

atoms will receive more energy to escape from the trap resulting in the loss of atoms. Atom loss spectra can be obtained by scanning the modulation frequency \( f_m \). Figure 3.5 shows the scaled number of atoms as a function of the modulation frequencies for three shallow traps we are interested in. All resonances in three axes corresponding to \( n = 2 \) are observed, and some \( n = 1 \) resonances are observed. We develop a model [85, 120] to estimate values of \( f_i \) for given powers and waists of the ODT beams. Firstly, we obtain an appropriate expression for the total potential consisting contributions from the ODT beams and the gravitational potential, and this expression is a function of the powers and waists of the ODT beams. A 3D-harmonic-oscillator approximation is assumed in the vicinity of the minimum of the potential, and the trap frequencies \( f_i \) can be extracted by fitting the small region near the trap minimum using a 3D harmonic expression. By comparing to the measurement values of frequencies, the best-fit waists of ODT beams are determined to be 97.5 \( \mu \)m (horizontal) and 105 \( \mu \)m (vertical) for the input beam, and 82.5 \( \mu \)m (horizontal) and 105 \( \mu \)m (vertical) for the crossed beam. These values are in agreement with the measured waists of the ODT beams of about 90 \( \mu \)m. In the final trap where OFR experiments performed, the final power of the input beam is 1.1 W. The model gives the geometric mean of the trap oscillation frequency \( \bar{\omega} = 2\pi \times (f_x f_y f_z)^{1/3} = 2\pi \times (60 \pm 5) \) Hz.

It is worth mentioning that the ODT is off when applying OFR beams, so it is not necessary to take into account ac Stark shift of the transition due to the ODT beam.
3.3.2 Improvement of Sample Lifetimes in the Optical Dipole Trap

An ultra-high vacuum system is needed in the experiments. The pressure in the chamber is on the order of $10^{-11}$ torr after running the titanium sublimation pump. As introduced before, a BEC of $^{88}$Sr is achieved via sympathetic cooling with $^{87}$Sr. The stability of the number of $^{88}$Sr atoms in the BEC depends to a great extent on the stability of the number of $^{87}$Sr atoms. Initially, it was observed that the number of $^{87}$Sr at the end of the evaporation procedure fluctuated dramatically, which was induced by the relative short lifetime of atoms in the ODT. Thus it results in the instability of the number of $^{88}$Sr atoms in the BEC. The lifetime of atoms in the ODT is primarily limited by the background pressure in the vacuum chamber. To get a stable number of atoms, it is necessary to reduce the background pressure further to increase the lifetime of atoms in the ODT.

The solution is liquid nitrogen. On the top of the vacuum chamber there is a reservoir with two vertical ports: one for filling liquid nitrogen and the other for venting. When the reservoir is filled with liquid nitrogen whose temperature is about 77 K, the pumping speed of the titanium sublimation pump increases dramatically and we get significant cryopumping of most gases, which leads to improved vacuum conditions.

Figure 3.6 shows the measurements of the lifetime of $^{87}$Sr in an ODT before and after adding liquid nitrogen, respectively. The lifetime increases significantly by about 50% from 21.6 s to 32.8 s. The number of $^{88}$Sr atoms in the BEC becomes more stable. It is noticed that liquid nitrogen evaporates very fast, and needs to be refilled every two hours during experiments.
Figure 3.6: Lifetime of $^{87}$Sr atoms in the ODT before and after adding liquid nitrogen. The number of atoms versus the holding time $t$ in the ODT are recorded. Data are fitted using the exponential function $N = N' \exp[t/\tau_0]$ with the initial number $N'$ and the lifetime $\tau_0$. 

$$\tau_0 = 21.6 \pm 0.8 \text{ s}$$

$$\tau_0 = 32.8 \pm 1.2 \text{ s}$$
3.3.3 Optical Dipole Trap Formed by the OFR Beam

The OFR laser is only 24 MHz red detuned from the 689 nm atomic resonance. Similar to the principle of the 1064 nm optical dipole trap, the red detuned OFR laser can also form an attractive optical dipole trap, which may make the trap stronger and deeper leading to the change of the trap frequencies. Thus it is necessary to estimate the influence from the attractive trap formed by the OFR beam.

Since the OFR laser is quite close to the 689 nm transition and very far away from any other transition, we can consider only the 689 nm transition contributing to the ac Stark shift. The dipole potential is given by [121]

\[ U_{\text{dip}} = \frac{3\pi c^2}{2\omega_0^2} \frac{\Gamma}{\delta_a} I, \]  

(3.19)

where \( c \) is the speed of light, \( \omega_0 = 2\pi c/\lambda \) with \( \lambda = 689 \text{ nm} \), \( \Gamma = 2\pi \times 7.5 \text{ kHz} \), the red detuning of the OFR beam with respect to the atomic resonance \( \delta_a/(2\pi) = 24 \text{ MHz} \), and the intensity of the OFR beam \( I = 0.057 \text{ W/cm}^2 \). Thus the potential formed by the OFR beam is estimated to be \( U_{\text{dip}}/k_B = 200 \text{nK} \). Moreover, the waist of the OFR beam of 725 \( \mu \text{m} \) is much larger than that of ODT beams, which makes the potential formed by the OFR beam extremely shallow so that it can be neglected here.

3.3.4 Variational Calculation of BEC Properties

In Sec. 3.2.2, a variational calculation of the BEC properties in a trap have been discussed. Now I will focus on the related study in OFR experiments.
(a) Collapse of BECs in the trap

In the OFR experiments, the dimensionless parameter $\xi = N_0|a_{bg}|/a_{ho}$ is calculated to be 0.53 with the number of $^{88}$Sr BECs $N_0 = 7000$, the background scattering length of $^{88}$Sr $a_{bg} = -2a_0$, and $a_{ho} = 1.38\,\mu$m for the geometric mean of trap frequencies of the ODT $\bar{\omega} = 2\pi \times 60\,\text{Hz}$. Since $\xi < \xi_{cr} = 0.67$, the condensate is stable. According to Eq. 3.14, the BEC size $\sigma_0$ is determined to be $0.8\,\mu$m with $w_{\min} = 0.83$ corresponding to the minimum of the $\xi = 0.53$ curve (blue solid) in Fig. 3.7. The peak density of the condensate is $n_0 \simeq 1 \times 10^{15}\,\text{cm}^{-3}$.

With the fixed scattering length $a_{bg} < 0$, collapse of condensates is triggered when the BEC number $N_0$ exceeds a critical number $N_{cr} = \xi_{cr}a_{ho}/|a_{bg}|$. The GP equation predicts $N_{cr}$ to be $7300 - 8000$ with $\xi_{cr} = 0.58$, and the variational calculation (Var Cal) gives rise to $N_{cr} = 8400 - 9200$ with $\xi_{cr} = 0.67$. The uncertainty in $N_{cr}$ is from uncertainties of trap frequencies $\bar{\omega} = 2\pi \times (60 \pm 5)\,\text{Hz}$. Figure 3.8 is the histogram of the number of $^{88}$Sr BEC including 56 independent measurements. The red solid vertical line corresponds to $N_{cr} = 8000$, the upper limit of $N_{cr}$ given by the GP equation, and the blue dashed vertical line at $N_{cr} = 9200$ is the upper limit of $N_{cr}$ estimated by the variational calculation. As expected, all data are below $N_{cr} = 9200$, and 95% data are less than $N_{cr} = 8000$.

(b) Collapse of BECs without the trap

Now we will consider the collapse of condensates with the modified negative scattering length in absence of a trap, which is the case in the experiments when the OFR beam is applied. Without the term due to the ODT, the scaled energy per particle in units
Figure 3.7: Energy per atom with attractive interactions in the ODT (solid curve) or without the ODT (dotted and dotted-dashed curves), in terms of $\hbar\bar{\omega}$, versus the dimensionless parameter $w$. The solid curve corresponds to $a = a_{bg} = -2a_0$ ($\xi = 0.53$) in the ODT which is the condition in OFR experiments, and $w_{\text{min}}$ (red point and gray dashed line), indicates the local minimum of the curve. The dotted and dot-dashed curves show cases when $a = -2a_0$ and $a = -5a_0$ without the ODT, respectively, as well as $w_{\text{max}}$ where the maximum of each curve (square and triangle) is located. In the absence of the ODT, collapse is triggered as long as $w_{\text{max}} > w_{\text{min}}$, e.g. the $a = -5a_0$ case (dot-dashed curve).
Figure 3.8: Histogram of the number of $^{88}\text{Sr}$ condensate atoms before applying the OFR beam. There are 56 repeated shots recorded. The red solid and blue dashed lines correspond to the upper limit of the critical number $N_{cr}$ given by the Gross-Pitaevskii equation ($\xi_{cr} = 0.58$), and the variational calculation ($\xi_{cr} = 0.67$), respectively.
of $\hbar \omega$ can be obtained by removing the trap-potential term $3w^2/4$ from Eq. 3.10:

$$\tilde{E}(w) = \frac{3}{4}w^{-2} - \xi(2\pi)^{-1/2}w^{-3}, \quad (3.20)$$

To figure out the critical condition of collapse in this case, we let

$$\frac{d\tilde{E}(w)}{dw} \bigg|_{w=w_{\text{max}}} = 0, \quad (3.21)$$

which yields

$$w_{\text{max}} = \sqrt{\frac{2}{\pi}} \frac{N_0|a|}{a_{ho}}. \quad (3.22)$$

Figure 3.7 shows the curves of energy per atoms with $a < 0$ in the ODT and without the ODT, respectively. The solid curve is the experiment condition in the ODT without applying the OFR beam ($a = a_{bg} = -2a_0$), and the BEC size can be calculated from the position of the minimum $w_{\text{min}}$. After turning off the ODT, the energy curve jumps to the dashed one for which the location of the maximum is $w_{\text{max}} < w_{\text{min}}$. Therefore, the condensate expands with time of flight. However, if the scattering length is modified to more negative by an OFR beam, i.e. $a = -5a_0$ (dot-dashed curve), so that

$$w_{\text{max}} > w_{\text{min}}, \quad (3.23)$$

where $w_{\text{min}}$ is still calculated for $a = a_{bg} = -2a_0$, it results in the collapse of the condensate.

Considering $a$ is negative, combining Eqs. 3.22 and 3.23 yields the condition of BEC collapse in the absence of the ODT

$$a < -\sqrt{\frac{\pi}{2}} \frac{w_{\text{min}}a_{ho}}{N_0}, \quad (3.24)$$

where $a$ is the value with the OFR beam on. We will study the collapse in $^{88}\text{Sr}$ BEC experiments in Sec. 3.6.
3.3.5 Momentum Distribution of BECs

In the Thomas-Fermi (TF) approximation, where the interaction is repulsive \((a > 0)\) and \(N_0a/a_\text{ho} \gg 1\), the quantum pressure kinetic energy term \(-\hbar^2\nabla^2/2m\) in the GP equation (Eq. 3.4) can be neglected. The density of the condensate in a harmonic potential has an anti-parabolic profile \([113]\)

\[
n(x, y, z) = n_0 \left(1 - \frac{x^2}{R_x^2} - \frac{y^2}{R_y^2} - \frac{z^2}{R_z^2}\right)^{3/2} \theta \left(1 - \frac{x^2}{R_x^2} - \frac{y^2}{R_y^2} - \frac{z^2}{R_z^2}\right),
\]

with the Heaviside function \(\theta\) and the radii of the condensate cloud \(R_x, R_y,\) and \(R_z\).

The TF approximation is valid in the case of an \(^{84}\text{Sr}\) condensate with the scattering length \(a = 122.7a_0\) \([8, 76]\).

However, for \(^{88}\text{Sr}\) with a weak attractive interaction \((a < 0)\) and \(N_0|a|/a_\text{ho} < 1\), quantum pressure plays a significant role and cannot be ignored. Thus the TF approximation is invalid in this case. Following the treatment in Ref. \([113]\), a gaussian density distribution is used, as discussed in Sec. 3.2.2. The condensate density in an isotropic harmonic trap is given by Eq. 3.12.

Taking the Fourier transform of \(\phi(r)\), the momentum distribution of the condensate also has a gaussian distribution. In time-of-flight (TOF) measurements, absorption images after expansion measure the areal density, which is fit with a bimodal function including a Bose distribution for the thermal atoms and a narrow gaussian density distribution for the BEC

\[
n_{\text{TOF}}(r) = \frac{N_0}{2\pi\sigma^2} \exp \left[-\frac{r^2}{2\sigma^2}\right].
\]

Here the number of atoms in the condensate and the size of condensate after expansion are both determined from the fitting. The resolution of the camera system will be discussed in Sec. 3.4.
3.3.6 Single-atom Light Scattering

The atom loss due to the single-atom light scattering is not negligible here, and we assume that every scattering event results in the loss of one atom. The scattering rate $\Gamma_{sc}$ is given by [122]

$$\Gamma_{sc} = \frac{s_0 \Gamma/2}{1 + s_0 + (2\delta_a/\Gamma)^2},$$

(3.27)

where the dimensionless on-resonance saturation parameter $s_0 = I/I_s = 1.86 \times 10^4$ with the saturation intensity $I_s = 3 \mu W/cm^2$ for the 689 nm intercombination transition and intensity of the OFR beam $I = 0.057 W/cm^2$.

The scattering rate calculation according to Eq. 3.27 gives, for example, $\Gamma_{sc} = 11.7, 12.7$, and $17.0 s^{-1}$ for lasers blue detuned by $\delta = 2\pi \times 1, 2$, and $5 MHz$ from the -24 MHz PA resonance, respectively, whose detuning from the atomic resonance is $\delta_a = 2\pi \times 23, 22$, and $19 MHz$.

3.4 Characterization of the Imaging System

3.4.1 Calibration of Pixel Sizes

The pixel sizes of the camera systems is calibrated via time-of-flight measurements. Figure 3.9 shows the relative vertical position of atom clouds with a series of drop times, which are fitted by the freely falling formula. The pixel sizes are determined to be $9.9 \mu m/pixel$. 
Figure 3.9: Calibration of pixel sizes of imaging systems in OFR experiments via time-of-flight measurements. Pixel sizes are determined to be 9.9 µm/pixel by fitting data using the freely falling formula.
3.4.2 Characterization of the Resolution of Imaging System

Firstly, a very tiny $^{88}\text{Sr}$ thermal-atom cloud is prepared and dropped for the time $t$. We assume a gaussian density profile of an atom cloud

$$n(r) = n'_0 \exp\left[-\frac{r^2}{2\sigma^2}\right] \tag{3.28}$$

where $n'_0$ is the peak density of an atom cloud after expansion, and the width after expansion $\sigma$ can be described by [87]

$$\sigma^2 = \sigma_0^2 + (vt)^2, \tag{3.29}$$

with the original size of the atom cloud before expansion $\sigma_0$, and the speed of thermal atoms $v$.

Now the resolution of the imaging system $\sigma_{\text{res}}$ will be taken into account. The point spread function (PSF) of the imaging system is given by

$$L(r) = \frac{1}{2\pi\sigma_{\text{res}}^2} \exp\left[-\frac{r^2}{2\sigma_{\text{res}}^2}\right], \tag{3.30}$$

which is also gaussian.

Thus, the observed density profile can be received by convolution

$$n_f(r) = n(r) \otimes L(r) = \frac{n'_0\sigma^2}{\sigma^2 + \sigma_{\text{res}}^2} \exp\left[-\frac{r^2}{2(\sigma^2 + \sigma_{\text{res}}^2)}\right] = n_0 f \exp\left[-\frac{r^2}{2\sigma_{\text{obs}}^2}\right] \tag{3.31}$$

with the observed size of the atom cloud after expansion

$$\sigma_{\text{obs}}^2 = \sigma^2 + \sigma_{\text{res}}^2. \tag{3.32}$$

Substituting Eq. 3.29 into Eq. 3.32 yields

$$\sigma_{\text{obs}}^2 = \sigma_0^2 + (vt)^2 + \sigma_{\text{res}}^2. \tag{3.33}$$
When the drop time \( t \) is long enough (e.g. 1-4 ms) so that \( vt \gg \sigma_0, \sigma_{\text{res}} \), we have \( \sigma_{\text{obs}} \simeq vt \). The temperature of thermal atoms \( T \) can be extracted according to

\[
\frac{1}{2} k_B T = \frac{1}{2} m v^2 \simeq \frac{1}{2} m \left( \frac{\sigma_{\text{obs}}}{t} \right)^2.
\] (3.34)

With the parabolic-trap approximation, the original size of the thermal could before expansion \( \sigma_0 \) can be determined from

\[
\frac{1}{2} k_B T = \frac{1}{2} m (\bar{\omega} \sigma_0)^2,
\] (3.35)

where angular trap frequencies \( \omega = 2\pi \times f \) that can be predicted by the trap model at the final power of the input ODT beam before the vacuum chamber of 9.5 W (Table 3.1).

Combining Eqs. 3.34 and 3.35 yields

\[
\sigma_0 = \frac{\sigma_{\text{obs}}}{\bar{\omega} t}.
\] (3.36)

Figure 3.10: Observed sizes of atom clouds \( \sigma_{\text{obs}} \) in horizontal (Left) and vertical (Right) axes after a series of long drop times. \( \sigma_{\text{obs}}/t \) is extracted by linear fit of data.
Figure 3.10 shows the time-of-flight data with a series of drop times, from which $\sigma_{\text{obs}}/t$ is determined to be 8.9 $\mu$m/ms and 10.1 $\mu$m/ms in the vertical (Y) and horizontal (X) axes, respectively. Thus, the original size of the atom cloud $\sigma_0$ can be calculated according to Eq. 3.36 (Table 3.1). Fig. 3.11 shows an example of the data with an expansion time of 2 ms, which can be well fitted using a gaussian distribution.

![Figure 3.11: 2D absorption image of a thermal atom cloud after 2 ms expansion time of (Left) and areal density profile (Right) fitted using a Gaussian distribution.](image)

Then we measure the observed size of thermal cloud with the extremely short expansion time of 1 $\mu$s $\sigma'_{\text{obs}}(X) = 10.6 \mu$m and $\sigma'_{\text{obs}}(Y) = 6.7 \mu$m (Fig. 3.12), so that $vt \ll \sigma_0, \sigma_{\text{res}}$. Therefore, Eq. 3.33 can be rewritten as

$$\sigma'^2_{\text{obs}} = \sigma^2_0 + \sigma^2_{\text{res}}, \quad (3.37)$$

from which the resolution of the imaging system can be determined by

$$\sigma_{\text{res}} = \sqrt{\sigma'^2_{\text{obs}} - \sigma^2_0}. \quad (3.38)$$

Table 3.1 lists measured parameters for calculation of the resolution, which is determined to be 6.3(5) $\mu$m and 4.6(2) $\mu$m in the horizontal and vertical directions, respectively.
Figure 3.12 : 2D absorption image of a thermal atom cloud after extremely short expansion time of 100 µs (Left) and areal density profile (Right) fitted using a gaussian distribution.

<table>
<thead>
<tr>
<th>Axes</th>
<th>$f$ [Hz]</th>
<th>$\sigma_0$ [$\mu$m]</th>
<th>$\sigma'_\text{obs}$ [$\mu$m]</th>
<th>$\sigma_{\text{res}}$ [$\mu$m]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Horizontal (X)</td>
<td>190(10)</td>
<td>8.5(4)</td>
<td>10.6</td>
<td>6.3(5)</td>
</tr>
<tr>
<td>Vertical (Y)</td>
<td>290(10)</td>
<td>4.9(2)</td>
<td>6.7</td>
<td>4.6(2)</td>
</tr>
</tbody>
</table>
3.5 OFR Experimental Sequence

The preparation of the $^{88}$Sr condensate has been briefly introduced in Chapter 2. Figure 3.13(a) shows the full timing of OFR experiments. The condensates are trapped in a shallow ODT with the power of the input beam of 1.1 W. Then the ODT is turned off and the condensates fall and expand freely. The OFR laser is applied to the condensate 20 $\mu$s before extinguishing the ODT and left on for a variable time $\tau$ on the order of ms during expansion of atom clouds [Fig. 3.13(b)]. The waist of the OFR beam is 725 $\mu$m which is much larger than the condensate dimension, and the intensity of the beam is 0.057 W/cm$^2$. After a 35 ms time of flight, the atom cloud is monitored with the absorption imaging using the $^1S_0-^1P_1$ transition.

The exposure time in the ODT is short enough that the initial density distribution of the condensate reflects the ODT potential and the background scattering length, while the expansion dynamics is sensitive to the interaction energy determined by $a = a_{bg} + a_{opt}$.

Figure 3.14 shows 1D slices through absorption images of atoms after a 35 ms time-of-flight with and without application of the OFR laser. Absorption images measure the areal density, which is fit with a bimodal function including a Bose distribution for the thermal atoms and a narrow gaussian density distribution for the BEC to determine the number of atoms in the BEC $N_0$ and BEC size $\sigma$. (Quoted sizes reflect correction for imaging system resolution as discussed in Sec. 3.4.2.) The condensate size after a long time of flight is a good probe of interactions because of the sensitivity to the initial interaction energy. Since $a_{bg}$ is close to zero, the blue-detuned OFR laser with respect to a PA resonance leads to increased interaction energy and a faster
Figure 3.13: Full and partial experimental timing for OFR experiments. (a) Full experimental timing for OFR experiments and (b) Zoom in on the partial timing in the ellipse in (a) where the relative starting time 0 corresponds to 18.5 s in (a). The OFR beam is applied immediately after extinguishing the ODT and lasts for time of $\tau$. 

(a)

(b)
Figure 3.14: Line profiles through absorption images showing OFR-induced variation of BEC expansion. Data correspond to no OFR laser and an OFR laser blue and red detuned by 0.5 MHz with respect to the -24 MHz PA line [86] applied for $\tau = 1.2$ ms. Expansion times are 35 ms. Fits are a Bose distribution for the thermal atoms (-) and a Gaussian density distribution for the BEC. The blue-detuned OFR laser with respect to a PA resonance leads to increased interaction energy and a faster condensate expansion (right column), whereas red detuning triggers a collapse of the condensate (left column).
condensate expansion, whereas red detuning triggers a collapse of the condensate.

3.6 Results

3.6.1 Timescale of Full Conversion of Interaction Energy to Kinetic Energy

As introduced in Sec. 3.2.2, we can qualitatively understand the TOF data via the conversion of the interaction energy relevant to the scattering length $a$ to kinetic energy extracted from the TOF data. Here we will estimate the timescale of the full conversion of interaction energy to kinetic energy with a hydrodynamic description of the condensate dynamics [113].

The acceleration of atoms during expansion arises from the interaction pressure

$$P = gn(r)^2/2, \quad (3.39)$$

and a characteristic acceleration $\ddot{a}$ can be approximated from

$$mn(r)\ddot{a} \approx -\nabla P \approx -n(r)\nabla [gn(r)], \quad (3.40)$$

which yields

$$\ddot{a} = -\nabla [gn(r)]/m \sim gn_0/m\sigma_0. \quad (3.41)$$

In the large $N_0a/a_{ho}$ limit where $a$ has been modified by the OFR laser to positive, one can neglect the kinetic-energy term in Eq. 3.16 to find the characteristic final velocity given by the conservation of energy,

$$v_f \sim \sigma_v \sim \sqrt{gn_0/m}. \quad (3.42)$$
This implies a conversion timescale

$$\tau = \frac{v_f}{a} \sim \sigma_0 \sqrt{\frac{m}{g n_0}},$$

(3.43)
of 1 ms for $a_{\text{opt}}$ of $10 a_0$, which roughly matches observations. Losses from single-atom light scattering preclude leaving the OFR beam on during the entire expansion time, and knowledge of the time required for close to full conversion is helpful for interpreting the results of experiments in which we apply the OFR laser for a fixed interaction time and vary the detuning, which will be discussed below.

### 3.6.2 Variation of BEC Size and Numbers versus Exposure Time

In Fig. 3.15, we study the variation of the BEC size and number with the exposure time, $\tau$, for several blue detunings of the OFR laser. To quantitatively analyze the variation of size and atom number versus interaction time and extract OFR parameters, it is necessary to treat dynamics and atom loss with the time-dependent non-linear Gross-Pitaevskii equation, including the effects of $a_{\text{opt}}$, $K_{\text{total}}$, and single atom light scattering, and neglecting effects of thermal atoms. The fit parameters are $\ell_{\text{opt}}/I$, $\eta$, and $K_0$. The rate of atomic light scattering $\Gamma_{\text{sc}}$ varies from 12 to $17 \, \text{s}^{-1}$ (Sec. 3.3.6), and is included in the simulation. Balasubramanian Ramachandran from Dr. Pu’s group did the simulation and created the lookup tables of BEC numbers and sizes for three different blue detunings of 1, 2, and 5 MHz, and these lookup tables are evaluated at a series of exposure times $\tau$ and fit parameters $\ell_{\text{opt}}/I$, $\eta$, and $K_0$. I use these lookup tables to find interpolating functions for the parameter dependence of BEC numbers and sizes. Using the interpolating functions, the evolution curves of the BEC number and size can be easily found for the given fit parameters.
Figure 3.15: The BEC size and number versus exposure time. (a) BEC size after 35 ms of expansion versus the exposure time of the OFR laser with the intensity of 0.057 W/cm² and three different detunings from the -24 MHz PA line. (b) Number of condensate atoms versus exposure time. Curves calculated by the Gross-Pitaevskii equation correspond to a combined fit of the data, yielding $\eta = 19.5$, $\ell_{opt}/I = 2.2 \times 10^4 a_0/(W/cm^2)$, and $K_0 = 5.8 \times 10^{-7} cm^3/s$. Error bars represent the standard deviation of the mean from multiple measurements.
The fit parameters can be determined by fitting the calculated evolution curves to experimental data (Fig. 3.15). The data at largest detuning from photoassociative resonance strongly determines the background loss because loss from the OFR is small there. The fit optical length is $\ell_{\text{opt}}/I = (2.2 \pm 1.0) \times 10^4 \, a_0/(\text{W/cm}^2)$, and the fit parameter $K_0 = (5.8 \pm 1.3) \times 10^{-7} \, \text{cm}^3/\text{s}$. Loss from the OFR is described by $\ell_{\text{opt}}$ and $\eta = 19.5^{+8}_{-3}$, and there is strong anti-correlation between $\ell_{\text{opt}}$ and $\eta$. The uncertainty is dominated by systematic uncertainty in the trap oscillation frequency and imaging resolution. These results are in good agreement with the measured value $\ell_{\text{opt}}/I = 1.58 \times 10^4 \, a_0/(\text{W/cm}^2)$ and disagree slightly with $\ell_{\text{opt}}/I = 8.3 \times 10^3 \, a_0/(\text{W/cm}^2)$ calculated directly from knowledge of the molecular potentials [103].

Experiments with a thermal strontium gas [103] found larger losses associated with an OFR than described by theory, which was described by $\eta = 2.7$. These measurements probed the vicinity of the photoassociative transition ($|\delta| < 50 \, \gamma_m$). The additional loss is not well understood theoretically [103]. We see a similar resonance width in a BEC when we significantly reduce the laser intensity and interaction time and take a photoassociative loss spectrum of this vicinity of the PA transition. Our use of the OFR probes the distant wings ($50 \, \gamma_m < \delta < 667 \, \gamma_m$), and a fit of the loss using the single resonance model requires an even larger value of $\eta$. We could interpret the varying $\eta$ values as meaning that the full spectrum of photoassociative loss, including the far wings, is not well described by a Lorentzian. A further theory describing $\eta$ is needed.

Figure 3.16 shows the numbers and temperatures of thermal atoms versus exposure time corresponding to the same experimental conditions in Fig. 3.15. There is no significant change of the numbers and temperature of thermal atoms during the
Figure 3.16: The number and temperature of thermal atoms versus exposure time. (a) Number of thermal atoms, and (b) Temperature of thermal atoms versus exposure time, which corresponds to the same experimental conditions in Fig. 3.15.
application of the OFR beam.

3.6.3 BEC Sizes and Number versus Detuning

The dependence of the BEC size and number on detuning from the -24 MHz PA line is shown in Fig. 3.17 for a fixed intensity and interaction time $\tau = 4$ ms. With the fit parameters from Fig. 3.15, the simulation curves were calculated by Balasubramanian Ramachandhran using the GP equation. Simulation curves describe the data well over this range. Note that the number of atoms initially increases with blue detuning from PA resonance as the loss from the OFR ($K_{in}$) decreases. The number then slowly decreases because the background loss ($K_b$) increases approaching atomic resonance.

The BEC size data predicted by Eq. 3.16, which neglects atom loss and assumes that the OFR laser is applied long enough to fully convert interaction energy into kinetic, is also shown in Fig. 3.17(a). The difference between this curve and the data highlights that atom loss is significant during the conversion process at smaller detunings, and the Gross-Pitaevskii equation simulation is required to describe the data. A typical total scattering length [Fig. 3.17(a) inset] is $a = 20a_0$ for $\delta = 2\pi \times 1$ MHz $\simeq 67\gamma_m$.

For red detuning, the OFR laser makes the scattering length more negative and triggers a collapse of the condensate, which is evident as large loss in the plot of condensate number remaining after expansion [Fig. 3.17(b)]. The dramatic asymmetry of loss with respect to detuning from resonance shows that the loss must reflect condensate dynamics [115,123,124], not photoassociative loss directly caused by the OFR laser. The Gross-Pitaevskii equation provides a good description of the BEC number data for red detuning in spite of the fact that the collapse dynamics may contain beyond-mean-field effects [125] not taken into account in the Gross-Pitaevskii
Figure 3.17: The BEC size (a) and number (b) versus the detuning with respect to the -24 MHz PA resonance for an intensity of 0.057 W/cm². The OFR beam is applied for 4.0 ms, and the data are recorded after 35 ms of expansion. The insets give the total scattering length $a$ and the loss rate constants.
formalism.

A variational calculation of the condensate energy functional as a function of condensate size [113,116,118] for the parameters of Fig. 3.17 predicts that the condensate expands initially after the trap is extinguished if $a > -3.8 \pm 0.2 \alpha_0$. For more negative $a$ ($-10 \pm 3 \text{MHz} < \delta/2\pi < 0 \text{MHz}$), there is no repulsive energy barrier on the effective potential for the system and collapse results. Numerical simulation of the Gross-Pitaevskii equation supports this interpretation. Simulations show that collapse can be very non-uniform, as predicted in [123], with significant density increase only near the condensate center for $a$ only moderately more negative than the threshold. Similar to what previous works observed [115], in our work, there are some remnant atoms in condensates after collapse, even at small red detunings from the PA resonance corresponding to large attractive interactions.

Furthermore, similar to Fig. 3.17 where the exposure is 4 ms, Fig. 3.18 shows results with the exposure time of 1.2 ms. The fitting curves are given by GP equations, which reproduce the data at small detuning, but deviate from the data of BEC numbers a little bit at larger detuning [Fig. 3.18(b)] since there is no loss channel $K_b$ included in the fitting.
Figure 3.18: The BEC size (a) and number (b) versus the detuning with respect to the -24 MHz PA resonance for an intensity of 0.057 W/cm². The OFR beam is applied for 1.2 ms, and the data are recorded after 35 ms of expansion. The insets give the total scattering length and the loss rate constant $K_{in}$. 
Chapter 4

Rabi Oscillations between Atomic and Molecular Condensates Driven with Coherent One-Color Photoassociation

4.1 Introduction and Background

Coherent coupling between the atomic and molecular condensates has been demonstrated experimentally via MFRs [53, 55, 56, 126] and two-color PA [33, 59, 61, 63] for more than ten years. It provides a method to create molecular condensates, which overcomes the difficulty of directly cooling molecules due to the complex molecular energy levels. However, coherent one-color PA has never been observed due to the extremely short lifetime of the excited molecular states in alkali-metal atoms, which are used in most ultracold experiments. Recently, alkaline-earth atoms have attracted more attention both theoretically and experimentally. They feature a very narrow intercombination transition to a triplet state $^3P_1$, where excited molecular states with a long lifetime exist. So it provides an opportunity to achieve coherent one-color PA. Moreover, although Rabi oscillations between atomic and molecular condensates have been demonstrated in MFR experiments [53, 55, 56, 126], there are no Rabi oscillations reported in PA experiments, even in coherent two-color PA.

In this chapter, I will present the first coherent one-color PA driven near an intercombination transition [4] in an $^{88}$Sr condensate where the induced Rabi oscillations
between strontium atomic and molecular condensates are achieved (Fig. 4.1). Besides the long lifetime of the molecular state, this work also benefits from the large Franck-Condon overlap integral between ground and excited states, which makes atom-molecule coupling larger than the decoherence rate. At the same time, transient shifts and broadenings of the excitation spectra at short times predicted by Naidon et al. [64] are observed in this work. The many-body model [64] can describe the data well.

Figure 4.1: Rabi Oscillations between atomic and molecular condensates driven with coherent one-color photoassociation.
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Figure 4.1: Rabi Oscillations between atomic and molecular condensates driven with coherent one-color photoassociation.
4.1.1 Realization via Magnetoassociation

As introduced in Sec. 3.1.1, the s-wave scattering length of ultracold atoms can be modified by external magnetic field via MFRs, which facilitates the experimental achievement of atom-molecule coherence. Rabi oscillations have been seen with MFRs in BECs [126], thermal bosons [53], bosonic-atom pairs in a Mott insulator state in an optical lattice [55], and Bose-Fermi mixtures [56].

The first study of coherent atom-molecule conversion [126] used a Ramsey-pulse sequence with a MFR in a $^{85}$Rb Bose-Einstein condensate (BEC) to create superpositions of atomic and molecular fields and detect atom-molecule oscillations. After applying a time-dependent magnetic field near a Feshbach resonance, absorption images are recorded via a time-of-flight measurement. There are three components: detectable cold remnant condensates, a relatively hot atomic burst consisting of correlated pairs, and the undetected Feshbach molecules. The total number of observed components displays a clear oscillation with the time, which proves the oscillation of the number of the third component, the Feshbach molecules. This oscillatory behavior arises from the superposition state of separated atoms and Feshbach molecules. The authors also studied the magnetic-field dependence of the oscillation frequency, which can be well reproduced by a coupled-channel model well. The analysis can help determine the binding energy of the molecular bound state with very high precision [57,127]. However, the Rabi oscillations were observed only within a very limited region of the magnetic field, and the atom-molecule conversion efficiency is low, on the order of a few percent.

Three years after the publication of Ref. [126], the same group led by Wieman
reported the Rabi oscillations in $^{85}$Rb thermal bosons with magnetoassociation [53]. Instead of ramping the magnetic field crossing the magnetic Feshbach resonance, a novel and efficient technique of applying a sinusoidally oscillating magnetic field was devised, which does not require crossing the Feshbach resonance. Thus the technique overcomes problems of heating and enhanced collision loss induced by the resonance, which also improves the conversion efficiency significantly. Similar to the principle of dissociating molecules using radio-frequency photons, the modulation frequency is tuned close to the binding energy of the molecular state, and photons from the oscillating magnetic field stimulate a pair of atoms to decay to the molecular state by emitting a extremely low-energy photon. An oscillation in the conversion efficiency from thermal atoms to molecules as the function of the coupling time was observed. This technique can also be employed in atom-molecule conversion of $^{85}$Rb BECs, and the conversion fraction is up to 55%.

Later, Syassen et al. made use of MFRs to achieve Rabi oscillations in bosonic-atom pairs in a Mott insulator state [55]. Similar to the work of Ref. [126], magnetic-field sweeps crossing a Feshbach resonance are applied to a $^{87}$Rb atomic Mott insulator in an optical lattice. Rabi oscillations between atomic and molecular states with up to 29 cycles were observed. It is emphasized that a deep optical lattice guarantees the discrete entrance-channel states, and the coupling between one entrance-channel state and the molecular state is allowed for sufficiently weak coupling. Moreover, the inelastic-collision loss of molecules can be suppressed in optical lattices. Thus the conversion efficiency is large, resulting in Rabi oscillations with large amplitude. Thalhammer et al. [60] reported a very high conversion efficiency of 95% in an optical lattice and the lifetime of molecules can reach up to 700 ms.
Recently, Rabi oscillations in a Bose-Fermi mixture of $^{87}\text{Rb}$ and $^{40}\text{K}$ atoms were achieved via the magnetoassociation [56], where the magnetic-field dependence of frequencies of the oscillations has been studied.

In a word, magnetoassociation is now a common tool for creating quantum degenerate molecular gases and exploring the unique properties of universal Feshbach molecules [51–54].

### 4.1.2 Realization via Two-Color Photoassociation

PA [33] is another well-established technique for forming ultracold molecules. PA described in a dressed picture (Fig. 3.2) is fundamentally analogous to magnetoassociation [38]. Two-color PA, in which two light fields couple atomic scattering states to a ground molecular state through an intermediate, excited molecular level, is often used to create molecules in quantum degenerate gases through stimulated Raman transitions [58–60, 115], creation of coherent atom-molecule superpositions in a dark state [61], or stimulated Raman adiabatic passage (STIRAP) [62, 63].

The MFR is restricted to create molecules in the high rovibrational levels and is only applicable in a specific number of systems, whereas a stimulated two-photon Raman transition can form deeply bound vibrational levels that MFR cannot access, so it is a more general technique. In a simple three-level system, e.g. $\Lambda$-type scheme (Fig. 4.2), a pair of atoms populating in the state $|1\rangle$ initially, are excited to the state $|2\rangle$ by absorbing a photon from the laser field 1, and dumped to the final state $|3\rangle$ by emitting a photon to the laser field 2. It is noticed that the state $|2\rangle$ has spontaneous decay channels other than the the state $|3\rangle$, which results in a low transfer efficiency.

In Ref. [58], a stimulated Raman transition was used to couple the free and bound
Figure 4.2: Schematic of A-type-three-level excitation for a stimulated two-photon Raman transition or STIRAP. Atoms in the initial state $|1\rangle$ are transferred to the final state $|3\rangle$ with the aid of an intermediate state $|2\rangle$. The state $|2\rangle$ has spontaneous decay channels other than the state $|3\rangle$.

states of an $^{87}$Rb BEC, which forms molecules in a well-defined state. A extremely narrow resonance is displayed in the spectra of the Raman transition rate, which allows the determination of the binding energy of the molecular state with highly-improved accuracy [57]. The interactions between molecular condensate was also measured in this work. As mentioned in Sec. 4.1.1, an optical lattice can enhance the atom-molecule conversion efficiency via MFRs. In the same way, optical lattices was combined with the Raman transition to improve the efficiency of creating $^{87}$Rb$_2$ molecules in a selected molecular state from a Mott insulator state [59]. The number of the molecules is monitored through the measurement of the atom-loss spectra. The internal vibrational and the external center of mass motion quantum states of the molecules can be controlled simultaneously here.
To achieve a complete and uniform conversion efficiency fast, the STIRAP scheme was proposed [62]. Revisiting the Λ-type scheme (Fig. 4.2), the laser 2 couples the states $|3\rangle$ and $|2\rangle$, the two initially empty states, to form a coherent-superposition state. Subsequently, this state is coupled to the initially-occupied state $|1\rangle$ through the laser 1, and atoms are adiabatically transferred to the final molecular state $|3\rangle$ directly, avoiding the transient stage of staying at the radiatively decaying state $|2\rangle$. The cancelation of the $|1\rangle \rightarrow |2\rangle$ transition is due to the quantum destructive interference of the two excitation transitions, similar to the principle of the Electromagnetically Induced Transparency.

The dark superposition of the atomic and molecular states is the prerequisite of the STIRAP. For the first time, the dark superposition state coupling the atomic $^{87}$Rb BEC state ($|1\rangle$) and the ground molecular $^{87}$Rb$_2$ BEC state ($|3\rangle$) was observed in Ref. [61]. The photoassociative loss induced by the transition from the atomic BEC state ($|1\rangle$) to the intermediate excited molecular state ($|2\rangle$) is drastically suppressed, which indicates the formation of the dark state. The number of formed molecules in the molecular ground state is limited to 100 because of the laser induced loss. The laser-intensity and detuning dependence of the spectra were also measured and described by a simple three-level model well. Most experiments using STIRAP are performed in alkali-atom systems. Recently, STIRAP is also employed in alkaline-earth systems, where MFRs do not exist. For example, $4 \times 10^4$ molecules were created in the ground molecular state by STIRAP near the 689 nm intercombination transition of $^{84}$Sr [63]. Additionally, combining MFR and STIRAP, molecules in very deep rovibrational states, even the rovibronic ground state (the lowest state of all rovibrational states) can be formed from initial very weakly bound molecules [128, 129].
Although two-color PA has proven successful in creating molecular condensates [59, 68, 70-72], no work has reported the observation of Rabi oscillations between atomic and molecular condensates driven by two-color PA. The reason is that molecules formed by PA can be excited to another molecular state by absorbing photons from laser fields, and spontaneously decay to other states, resulting in the loss. Also, excited vibrational levels are unstable to collisional relaxation, leading to the loss of atoms. Usually, the loss is so large that the atom-molecule coupling is smaller than the decoherence rate, which prevents from displaying Rabi oscillations [59]. Very recently, Fu et al. [130] observed spin-orbit-coupling-induced Rabi oscillations between Feshbach molecules and a fully polarized Fermi gas using the two-photon Raman process.

4.1.3 One-Color Photoassociation

Rabi oscillations between atomic and molecular condensates have been of intense theoretical interest and experimental effort for over a decade in the quantum optics and ultracold atom communities since the first production of molecules in a BEC, but they have never been observed with photoassociation. Coherence seems even less accessible in one-color PA than two-color PA because of the short lifetime of excited molecular states. Coherent wave-function dynamics have been observed with broadband, femtosecond PA excitation in a thermal Rb gas [131], but the coherence between the molecule and atom fields was not discussed.

As pointed out in Ref. [64], the spontaneous emission of the excited molecular state in alkali systems is large, and the lifetime on the order of 10 ns is too short to allow the achievement of coherent coupling between atomic and molecular condensates.
Figure 4.3(a) shows the parameter regime for one-color PA using an electric-dipole allowed transition of sodium condensates, where the coherent regime is vanishingly small.

To access the coherent regime, it is required that the coherent coupling $w\sqrt{n}$ is comparable to or even larger than the spontaneous emission of the excited molecular state $\hbar\gamma_m$:

$$w\sqrt{n} \geq \hbar\gamma_m$$

(4.1)

Here $n$ is the condensate density, and $w$ is the transition matrix element between the relative-motion wave function for two atoms in the molecular and atomic-scattering channels. The matrix element

$$w \propto \sqrt{f_{FC}D},$$

(4.2)

where $I$ is the laser intensity, $f_{FC}$ is the Franck-Condon factor per unit energy for the PA transition, and $D$ is the transition dipole moment. In other words, the Rabi frequency

$$\Omega_R = \frac{w\sqrt{n}}{\hbar} \geq \gamma_m.$$  

(4.3)

Considering that the spontaneous decay rate $\gamma_m \propto D^2$ and substituting Eq. 4.2, we can get the ratio of Rabi frequency to the spontaneous decay rate

$$\epsilon = \frac{\Omega_R}{\gamma_m} = \frac{w\sqrt{n}/\hbar}{\gamma_m} \propto \frac{\sqrt{f_{FC}D}\sqrt{n}}{D^2} = \frac{\sqrt{f_{FC}n}}{D}. $$

(4.4)

To obtain a large $\epsilon$, a transition with a small transition dipole moment $D$ will be a good candidate. It is exactly the case of the intercombination transition in alkaline-earth systems. As introduced before, the angular-momentum coupling results in a weakly allowed transition with small effective transition dipole moment. For example,
Figure 4.3: Parameter regimes for one-color PA for a condensate of (a) Na, (b) Yb, and (c) Sr. The dipole transition ($\gamma_m = 2\pi \times 18 \text{ MHz}$) is used in the Na condensate, and the intercombination transition is used in Yb ($\gamma_m = 2\pi \times 364 \text{ MHz}$) and Sr ($\gamma_m = 2\pi \times 15 \text{ kHz}$). Adopted from Ref. [64].
the lifetime of the excited molecular state on the $|^{1}S_0 + ^3P_1\rangle$ molecular potential of $^{88}$Sr is 21 $\mu$s, which is three orders longer than that in alkali gases. Taking into account the $n$ and $f_{FC}$ dependence of $\epsilon$ (Eq. 4.4), $^{88}$Sr is a ideal system to achieve one-color coherent PA since it has the feature of an extremely high peak density of the condensate arising from small negative scattering length, and the large Franck-Condon overlap integral between ground and the excited second least bound molecular states supported by the $|^{1}S_0 + ^3P_1\rangle$ molecular potential. It is worth emphasizing that simply increasing the laser intensity and atom-molecule Rabi frequency, which is proportional to $\sqrt{I}$, does not necessarily offer a solution because transfer of population to non-condensate atomic states, which is proportional to $I$, becomes dominant at high laser coupling [64, 70-75]. (This limitation is analogous to spontaneous Raman scattering in two-color PA.) Figure 4.3(b) and 4.3(c) show the parameter regime for the intercombination transitions of ytterbium and strontium, respectively. The coherent regime, particularly in strontium, could be reached in experiments.

In this chapter, we demonstrate coherent, one-color PA and Rabi oscillations between atomic and molecular condensates. We also observe transient shifts and broadenings of the excitation spectra at short times and indications of universal dynamics on resonance [64], all of which have not been experimentally studied previously. We access the coherent regime by photoassociating atoms in an $^{88}$Sr condensate [4] to a weakly bound level of the metastable $|^{1}S_0 + ^3P_1\rangle$ molecular potential (Fig. 4.4). This yields a long molecular lifetime and a large Franck-Condon overlap integral between ground and excited states, which allows atom-molecule coupling to exceed loss and decoherence rates.
4.2 Theory of Coherent One-Color Photoassociation

Over the last ten years, a wealth of theoretical papers have discussed one-color PA of condensates [64–75]. Starting from the mean-field approximation, two coupled GP equations are firstly introduced to describe the mixed system of atomic and molecular condensates [64, 75]. It can distinguish two regimes of PA, the adiabatic and coherent regimes.

The adiabatic regime is defined where the excited molecular state is an intermediate state when two condensate atoms collide. A single GP equation is derived accompanied by a complex scattering length, whose real part indicates the change of the s-wave scattering and imaginary part represents the loss of atoms. This is exactly the description of the OFR discussed in Chapter 3. For a large molecular decay rate or detuning, one obtains the familiar expressions for PA loss in the adiabatic regime,
which is incoherent and described by a rate equation for the evolution of density $n$, 
\[ \dot{n} = -K_{\text{in}} n^2 \] for a BEC. The time-independent loss rate constant $K_{\text{in}}$ is
\[ K_{\text{in}} = \frac{4\pi\hbar}{M} \frac{\ell_{\text{opt}} \gamma_m^2}{(\delta - \delta')^2 + (\gamma_m + \Gamma_{\text{stim}})^2/4}, \]  
where $\delta$ is the laser detuning from the PA resonance, and $\delta'$ is the ac Stark shift of the PA transition due to the PA laser. Up to now, all one-color PA experiments were performed in this regime.

In the coherent regime, a molecular condensate is created in the excited molecular state, which has the properties of a one-body BEC. There is a coherent conversion between atomic and molecular condensates. As discussed before, it is difficult to access due to the fast spontaneous decay rate of dipole transition in alkali atoms, but it is not the case in the intercombination transition in the alkaline-earth system, especially in Sr. It is noticed that in the mean-field treatment, the pair correlation is excluded in adiabatic and coherent regimes.

However, the mean-field approximation breaks down when the correlated pair of condensates cannot be ignored, which is called "rogue dissociation" [72]. Not only does a molecular condensate return back to atomic condensates, but also to noncondensate modes, which are in the form of correlated atomic pairs. This process becomes more dominant at higher laser intensity, and limits the populations of molecules. In this case, the many-body theory, which includes a term for the dynamic correlation in the condensate, is developed to deal with this situation [64, 75]. In our work, we focus on the coherent regime with partial "rogue dissociation".

On short time scales compared to decoherence and for $\Omega_R > \sqrt{(\delta - \delta')^2 + (\gamma_m/2)^2}$, PA is coherent and transient spectral shifts and broadenings resulting from the turn-
on of the laser at \( t = 0 \) become important, which can be predicted well by the many-body model. The most striking prediction for this regime is coherent oscillations between atomic and molecular condensates, which obviously cannot be described by a rate equation.

With the first-order cumulant approximation, the many-body theory describes photoassociation in a BEC through [64]

\[
i \dot{\Psi} = \frac{\Psi^*}{\hbar} \left( g\Psi^2 + \int \frac{d^3k}{(2\pi)^3} gC_{\vec{k}}^{\text{dyn}} + w\Psi_m \right),
\]

\[
i \dot{\Psi}_m = \left( \delta' - \delta - i \frac{\gamma_m}{2} \right)\Psi_m + \frac{w}{\hbar} \Psi^2 + \int \frac{d^3k}{(2\pi)^3} wC_{\vec{k}}^{\text{dyn}},
\]

\[
i \dot{C}_{\vec{k}}^{\text{dyn}} = \frac{E_k}{\hbar} C_{\vec{k}}^{\text{dyn}} - i \dot{C}_{\vec{k}}^{\text{ad}},
\]

\[
(4.6)
\]

where \( \Psi \) is the atomic condensate wave function, \( \Psi_m \) is the excited-state molecular condensate wave function, and \( C_{\vec{k}}^{\text{dyn}} \) and \( C_{\vec{k}}^{\text{ad}} \) are amplitudes for initially unpopulated scattering channels (indexed by wave vector \( \vec{k} \)) reflecting the dynamic and adiabatic response to the turn-on of the PA laser, respectively. \( g = 4\pi\hbar^2 a/m \) describes the mean-field atom-atom interaction, where \( E_k = \hbar^2 k^2/m \) with the colliding-atom wave number \( k \). The ac Stark shift of the PA transition due to the PA laser \( \delta' \) arises from coupling of the excited molecular state to atomic scattering states besides the condensate and to bound states of the ground molecular potential [93]. We have neglected elastic and inelastic collisions involving molecules, which do not qualitatively change the results. It is noticed that when eliminating the dynamic-correlation term, i.e. setting \( C_{\vec{k}}^{\text{dyn}} = 0 \), Eqs. 4.6 will reduce to mean-field GP equations. In this case, transient phenomena cannot be predicted by the GP equations.
4.3 Experimental Setup

4.3.1 Description of the Optical Dipole Trap

The setup of the optical dipole trap in coherent PA experiments, which is described and characterized in detail in Ref. [85], is different from the old ODT setup in the OFR experiments consisting of two crossed 1064 nm laser beams with the waists of about 100 µm. In the new design, firstly, a large-volume pancake-shaped ODT is formed by two 1064 nm beams with a vertical waist of 60 µm and a horizontal waist of 300 µm, which serve as a loading trap. The two beams are generated from the same laser source as the old ODT beams. The advantage of this configuration is that larger number of atoms can be transferred from the 689 nm MOT, and the reproducibility for experiments is improved. The number of atoms collected in the ODT is increased by a factor of 3. Subsequently, the dimple trap, which is composed of two crossed beams with much tighter waists of 66.5 µm in the same plane serving as the loading trap, is turned on to overlap with the loading trap. The power of the dimple-trap beams arise linearly from zero to the maximum of about 8 W within 1 second [Fig. 4.5(a)]. At the same time, the power of the loading-trap beams decrease exponentially and is turned off as soon as the dimple-trap beams reach the maximum. The tight waists of the dimple beams guarantee the higher density of atom samples resulting in the higher efficiency of evaporation. Typically, in this dimple trap, $^{88}$Sr BEC can be created after 5 s evaporation, only half of the evaporation time needed in the old ODT trap.

The PA beam is applied to $^{88}$Sr condensates in the dimple trap with the power of 0.59 W and 0.50 W for the input and cross dimple beams, respectively [Fig. 4.5(b)]. The trap frequencies of the final cylindrical dimple trap are experimentally determined
Figure 4.5: Full and partial experimental timing for coherent PA experiments. (a) Full experimental timing for coherent PA experiments and (b) Zoom in on the partial timing in the ellipse in (a) where the relative starting time 0 corresponds to 14.5s in (a). The dimple beam is added to overlap with the ODT beam for 100ms to load atoms to the dimple trap. The evaporative cooling is performed in the dimple trap. The PA beam is turned on $\tau$ ms before extinguishing of the dimple beam, and turned off at the same time of extinguishing of the dimple beam.
to be $116 \pm 10$ Hz (axial) and $89 \pm 5$ Hz (radial) [85].

### 4.3.2 Optical Dipole Trap Formed by PAS beam

The 689 nm PA laser has a waist of $200 \mu$m on the atoms. Following the treatment in Eq. 3.19, for the highest intensity used here ($2.4 \text{ W/cm}^2$), the potential depth formed by the PA laser is about $10 \mu$K. Considering the trap frequencies of the ODT are on the order of 100 Hz, the $10 \mu$s timescale of the PA beam is too short to drive any dynamics resulting from the change in the ODT, such as sound waves or collective oscillations. Thus, resulting mechanical effects due to the PA laser can be neglected here.

### 4.3.3 AC Stark Shift of the Transition due to the Dimple Beam

In Ref. [20], the parameter of the ac Stark shift due to the ODT is experimentally determined to be $160 \text{ kHz}/(100 \text{ kW/cm}^2)$. As mentioned before, in the final dimple trap, the best-guess waists of the dimple beams are $66.5 \mu$m, and the powers of the input and cross dimple beams are 0.59 and 0.50 W, respectively. Thus the peak intensity is calculated to be $1.6 \times 10^4 \text{ W/cm}^2$. The peak ac Stark shift of the PA transition due to the ODT is $1.6 \times 10^4 \text{ W/cm}^2 \times 160 \text{ kHz}/(100 \text{ kW/cm}^2) = 26$ kHz. The ac Stark shift is constant for all PA measurements, but spatially dependent.

### 4.3.4 Intensity Calibration of PA Beams

In this section I will describe how to convert amplitudes of oscilloscope traces recorded via a monitor photodiode to the power of the PA beams.

In the experiments, a Thorlabs InGaAs Amplified detector (item number: PDA10CF)
with a 50Ω terminator is used to monitor powers of the PA beams, and traces are recorded by an oscilloscope. A window mounted in the optical path of the PA beam before the vacuum chamber reflects about 10% power of PA laser (Fig. 4.6). The reflected beam with a power of about 0.1 mW is focused down by a lens and falls on the detector. A typical trace is displayed in black in Fig. 4.7, which shows a large negative offset when the beam is off, and a slow drift in amplitude with time. Electrical specifications of the InGaAs detector are listed in Table 4.1. The dark offset of 20 mV with a 50 Ω load is comparable to the measured output voltage in Fig. 4.7 resulting in an observed large negative offset. The wavelength of the PA laser of 689 nm is beyond the working wavelength range of 700-1800 nm.

<table>
<thead>
<tr>
<th>Detector</th>
<th>Item #</th>
<th>Range [nm]</th>
<th>Rise Time [ns]</th>
<th>50Ω Dark Offset [mV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>InGaAs</td>
<td>PDA10CF</td>
<td>700 - 1800</td>
<td>2</td>
<td>20</td>
</tr>
<tr>
<td>Silicon</td>
<td>DET36A</td>
<td>350 - 1100</td>
<td>14</td>
<td>0.35</td>
</tr>
</tbody>
</table>

This problem was not noticed until experiments were completed. In order to extract accurate power measurements from the InGaAs detector measurements, another detector, the Thorlabs Silicon detector (item number DET36A), is mounted in the optical path of PA beams between the vacuum chamber and the reflection window (shown in dotted lines in Fig. 4.6). The Silicon detector gets the full power of PA beams on the order of 1 mW. Measurements are performed using both detectors with a 50 Ω terminator simultaneously. Table 4.1 shows that the Silicon detector has a
Figure 4.6: Schematic drawing of coherent PAS experiments. The detector and lens shown in dotted lines are only present after experiments (see text). It is noticed that the isolator is made of three components: an input vertical polarizer, a Faraday rotator, and an output polarizer tilted by 45° from the horizontal.
much smaller dark offset of 0.35 mV and its working range covers 689 nm, the wavelength of the PA laser. A typical trace using the Silicon detector is displayed in red in Fig. 4.7, which shows little offset when the beam is off and no drift with time. Thus, it is clear that the large negative offset and drift in monitor data (black one) is just an artifact of the monitor InGaAs detector. Furthermore, the rise times of both detectors are an order smaller than the turn-on time of signals which indicates the observed slope of the turn-off and turn-on are real.

Now we will focus on the calibration of the detectors. The PA beams are turned on for a long time, and the amplitudes of both traces $V_{\text{DET36A}}$ and $V_{\text{PDA10CF}}$ are recorded in the oscilloscope. The power of the PA beam $P_{\text{PA}}$ is also measured before the Silicon detector using a calibrated power meter (Thorlabs item number S121C). It is noted that the drift of the InGaAs detector has reached its steady state value at
Figure 4.8: Conversion from amplitudes of traces recorded by the monitor PDA10CF to those by DET36A.

A long time. A series of traces pairs with different $P_{PA}$ are recorded. There is no sign of saturation of the detectors with large powers. Data are plotted in Fig. 4.8 and 4.9, and fitting data using the linear function gives

$$V_{DET36A} = 0.92375 \times V_{PDA10CF} + 2.5061; \quad (4.7)$$

$$P_{PA} = 0.0503 \times V_{DET36A}; \quad (4.8)$$

where $V_{DET36A}$ and $V_{PDA10CF}$ are in units of mV, and $P_{PA}$ is in units of mW.

Combining Eqs. 4.7 and 4.8 gives the calibration of the power of PA beams with respect to the amplitude of monitor traces $V_{PDA10CF}$:

$$P_{PA} = 0.0465 \times V_{PDA10CF} + 0.1261. \quad (4.9)$$

As mentioned before, the drift of the InGaAs detector reaches its saturation value at a long time, which can be described by an exponential function $V_{PDA10CF}(t) =$
Figure 4.9: Power calibration of PA beams with respect to $V_{DET36A}$.

Figure 4.10: One trace recorded using the InGaAs detector with the exposure time of about $8 \mu s$. The red solid curve fits the first $2 \mu s$ data using the exponential function, and the green dashed curve is the extrapolated one using the same function.
Figure 4.11: One trace recorded using the InGaAs detector with the exposure time of about 2 µs.

\[ V_{\text{PDA10CF}}(t) = \exp[a + b/(t + c)] \]

where \( t \) is the time in µs, and \( a \), \( b \), and \( c \) are fitting parameters. Figure 4.10 shows a monitor trace with a long time of about 8 µs. It shows that the exponential function fitting the short time (2 µs) data (red solid curve) can predict the saturation value well at a long time (green dashed curve). Therefore, the exponential function is a good model to estimate saturation values of monitor traces with a short time. In Fig. 4.11, the exponential function is used to fit the trace with the short time of about 2 µs. We can get the saturation value \( V_{\text{PDA10CF}}(t \to \infty) = \exp[a] \). Powers of PA beams \( P_{\text{PA}} \) can be obtained by substituting \( V_{\text{PDA10CF}}(t \to \infty) \) into Eq. 4.9.

### 4.3.5 Characterization of the Offset of Time Pulses

In this section I will describe how to characterize the extra pulse width of photoassociation beams in the coherent PA experiments.

From traces of PA beams recorded by the oscilloscope, we notice that the actual
Figure 4.12: Trace of PA beam where the set value of the pulse width is 1.50 µs.
pulse widths of PA beams are always longer than the input values of the pulse widths in Labview interface. Figure 4.12 is a typical trace where the set value of the pulse width is 1.50 $\mu$s, while the actual pulse width is 1.788 $\mu$s. Here the actual pulse width is defined using the full width at half maximum.

We record similar traces at a series of set pulse widths, and extract the actual pulse width in each case. Figure 4.13 shows the results. Fitting data using the linear function whose slope is fixed at 1 gives the extra pulse widths of 0.288 $\mu$s.

Figure 4.13: Actual pulse widths vs set pulse widths. Fitting linear curve (red curve) to data (solid circle) gives the extra width of pulses of 0.288 $\mu$s.
4.4 Variational Calculation of BEC Properties

Here we describe the cylindrical dimple trap used in the coherent PA experiment, and calculate parameters of BECs in the trap.

4.4.1 Non-Spherically Symmetric Trap

The trap potential of the cylindrical dimple trap has the form of

\[ V(\rho, z) = \frac{m}{2}(\omega_\rho^2 \rho^2 + \omega_z^2 z^2), \]  

(4.10)

with the axial trap frequency \(\omega_z = 2\pi \times (116 \pm 10) \text{ Hz}\), and the radial trap frequencies \(\omega_\rho = 2\pi \times (89 \pm 5) \text{ Hz}\). The condensate is close to an oblate spheroid, which yields an oblate spheroid density distribution.

Following the treatment in Ref. [116, 132], the trial wave function is given by

\[ \psi(\rho, z) = \left( \frac{N_0}{\pi^{3/2}\ell_z \ell_\rho} \right)^{1/2} \exp\left( -\frac{\rho^2}{2\ell_\rho^2} - \frac{z^2}{2\ell_z^2} \right), \]  

(4.11)

where \(N_0\) is the BEC number, and \(\ell_\rho\) and \(\ell_z\) are determined by minimizing the condensate energy

\[ E(\ell_\rho, \ell_z) = \frac{N_0 h^2}{4m} \left( \frac{2}{\ell_\rho^2} + \frac{1}{\ell_z^2} + \frac{2\ell_\rho^2}{\ell_\rho^4} + \frac{2\ell_z^2}{\ell_z^4} - \frac{4}{\sqrt{2\pi}} \frac{N_0 |a|}{\ell_\rho \ell_z} \right), \]  

(4.12)

with \(\ell_{0i} = (\hbar/m\omega_i)^{1/2}\).

Now we will focus on the calculation of the critical BEC number \(N_c\) where the minimum of the condensate energy \(E\) starts to disappear. According to the derivation of \(\nabla E = 0\) in Ref. [116, 132], given a value of \(\beta = (2/\pi)^{1/2} N_0 |a| \ell_{0z}\), one can find the minimum in \(E\) only if equation

\[ Q(q_z) \equiv q_z^4 + \beta \frac{\ell_{0z}^2}{\ell_\rho} \frac{q_z^{3/2}}{\sqrt{q_z - \beta}} - 1 = 0 \]  

(4.13)
has a positive real solution, where

\[ q_z = \frac{\ell_z}{\ell_{0i}}; \quad (4.14) \]

\[ q_\rho = \frac{\ell_z}{\ell_{0\rho}}; \quad (4.15) \]

\[ q_\rho = (1 - \beta/q_z)^{1/4}. \quad (4.16) \]

Numerical calculation by Mathematica\textsuperscript{TM} gives the value of critical BEC number \( N_{cr} = 14300/|a| \) with the uncertainty of 3.6% arisen from uncertainties of trap frequencies of the ODT. Here the scattering length \( a \) is in units of \( a_0 \). Figure 4.14 plots the scattering-length dependence of the critical number of BECs. The critical number of condensate atoms for collapse arising from attractive interactions ranges from 7000 to 10000, which is in agreement with the observation of the number of condensates displayed in Sec. 4.4.2.

The density distribution of a BEC is given by

\[ n(\rho, z) = |\psi(\rho, z)|^2 = n_0 \exp \left( -\frac{\rho^2}{\ell_\rho^2} - \frac{z^2}{\ell_z^2} \right), \quad (4.17) \]

with the peak density

\[ n_0 = N_0/\left( \pi^{3/2} \ell_z \ell_\rho^2 \right). \quad (4.18) \]

It is noticed that in our program we defined the density distribution as

\[ n(\rho, z) = n_0 \exp \left( -\frac{\rho^2}{2\sigma_\rho^2} - \frac{z^2}{2\sigma_z^2} \right), \quad (4.19) \]

with BEC sizes defined in our paper

\[ \sigma_z = \ell_z/\sqrt{2}; \quad (4.20) \]

\[ \sigma_\rho = \ell_\rho/\sqrt{2}. \quad (4.21) \]
Figure 4.14: The critical number of BEC $N_{cr}$ depends on the scattering length $a$. 
Figure 4.15 : Histogram of the number of $^{88}\text{Sr}$ condensate atoms in the dimple trap before applying the PA beam. There are 53 repeated shoots recorded.
4.4.2 Observations of Critical Number for Collapse

Figure 4.15 is the histogram of 53 independent measurements of the number of $^{88}\text{Sr}$ atoms in the BEC. A similar experiment was performed by Sackett et al. [114]. The number is within a range of 7000 to 10000, in agreement with the above estimation from the variational calculation. The number of atoms in condensates constrains the scattering length $a > (-1.60 \pm 0.05) a_0$.

For $a = -1.6 a_0$ and $N = 8800$ atoms, and only including uncertainty from trap geometry, the variation calculation gives rise to the BEC sizes $\sigma_z = (0.55 \pm 0.04) \mu m$ and $\sigma_\rho = (0.59 \pm 0.04) \mu m$, and the peak density $n_0 = (3.0 \pm 0.7) \times 10^{15} \text{ cm}^{-3}$. This very high density is important for attaining large Rabi frequencies.

4.4.3 Momentum Distribution of BEC

Here we will derive the distribution in the momentum space. By assuming spherical distribution of $n(r)$ since $l_\rho \simeq l_z = l_r$, the wave function Eq. 4.11 can be rewritten as

$$
\psi(\vec{r}) = \psi(r) = \left( \frac{N_0}{\pi^{3/2} \ell_r^3} \right)^{1/2} \exp\left( -\frac{r^2}{2\ell_r^2} \right) \tag{4.22}
$$

where $r$ is the radial coordinate, and $\ell_r \sim 0.8 \mu m$ is the size of BEC.

The Fourier transform gives the distribution in the momentum space:

$$
\psi(\vec{p}) = (2\pi \hbar)^{-3/2} \int d\vec{r} \psi(\vec{r}) e^{i\vec{p} \cdot \vec{r}/\hbar} \tag{4.23}
$$

The distribution in the momentum space is also spherical and is calculated by substituting Eq. 4.22 into Eq. 4.23:

$$
\psi(p) = \left( \frac{N_0 \ell_r^3}{\pi^{3/2} \hbar^3} \right)^{1/2} \exp\left( -\frac{\ell_r^2 p^2}{2\hbar^2} \right) \tag{4.24}
$$
So the distribution of momentum is given by

\[
n(p) = |\psi(p)|^2 = \frac{N_0 \ell_r^3}{\pi^{3/2} \hbar^3} \exp\left[-\frac{p^2}{2[\hbar/(\sqrt{2}\ell_r)]^2}\right],
\]

(4.25)

which is a Gaussian distribution with an rms width of \(\Delta p = \hbar/(\sqrt{2}\ell_r)\). The wavenumber \(k_r\) is on the order of \(\Delta p/h = 1/(\sqrt{2}\ell_r) = 8.8 \times 10^5 \text{ m}^{-1}\).

### 4.4.4 Probability Density Function of BEC Densities

(a) Evolution of the number of atoms in the BEC

We start from Eqs. 4.6 by ignoring \(C_{\text{dyn}}^{\text{dyn}}\) terms, which are the mean-field GP equations as introduced before:

\[
i\dot{\Psi} = \frac{\Psi^*}{\hbar} \left( g\Psi^2 + w\Psi_m \right),
\]

(4.26)

\[
i\dot{\Psi}_m = \left( \delta' - \delta - i\frac{\gamma_m}{2} \right) \Psi_m + \frac{w}{\hbar} \Psi^2.
\]

(4.27)

By assuming spherical distribution of \(n(r)\), the initial BEC density can be written as

\[
n(r) = n_0 e^{-r^2/(2\ell^2)}.
\]

(4.28)

Given values of the optical length \(\ell_{\text{opt}}\) and the detuning with respect to PA resonance \(\delta\), the evolution of the BEC density \(\tilde{n}(t, n(r))\) is a function of the time \(t\).

Now we will take into account the contribution of each density component. Firstly, a lookup table of \(\tilde{n}(t, n)\) with respect to a series of discrete values of \(t\) and \(n\) is built in Mathematica\textsuperscript{TM}. Using the interpolating function, the BEC density evolution \(\tilde{n}(t, n)\) can be found easily for given \(t\) and \(n\) (Fig. 4.16). Figure 4.17 shows a plot of
Figure 4.16: The evolution of the BEC density $\tilde{n}(t, n)$ with $n_0 = 3.1 \times 10^{21}\text{cm}^{-3}$ and $N_0 = 8700$.

Figure 4.17: The scaled evolution of the BEC density $\tilde{n}(t, n)/n$ with $n_0 = 3.1 \times 10^{21}\text{cm}^{-3}$ and $N_0 = 8700$. 
the evolution of the scaled BEC density $\tilde{n}(t,n)/n$ which describes the evolution of the density (number) of atoms with the initial local density $n$ in terms of percentage.

Since $n(r)$ is position dependent, we can get the evolution of the total number of atoms in the BEC $N(t)$ by integrating $\tilde{n}(t,n(r))$ over the whole real space $V$

$$N(t) = \int_V \tilde{n}(t,n(r))dV. \quad (4.29)$$

It is noted that $\tilde{n}(t = 0, n(r)) = n(r)$, so the initial number of atoms in the BEC is given by

$$N_0 = N(t = 0) = \int_V \tilde{n}(t = 0, n(r))dV = \int_V n(r)dV. \quad (4.30)$$

(b) Scaled probability density function of BEC density

Substituting Eq. 4.28 into Eq. 4.30 gives

$$N_0 = \int_0^{+\infty} 4\pi r^2 n(r)dr = 4\pi n_0 \int_0^{+\infty} r^2 e^{-r^2/(2l^2)} dr. \quad (4.31)$$

From Eq. 4.28, we can have

$$r^2 = 2l^2 \ln \left( \frac{n_0}{n} \right); \quad (4.32)$$

$$r = \sqrt{2l} \left[ \ln \left( \frac{n_0}{n} \right) \right]^{1/2}. \quad (4.33)$$

Differentiating Eq. 4.33 generates

$$dr = -\frac{\sqrt{2}l}{2} \left[ \ln \left( \frac{n_0}{n} \right) \right]^{-1/2} \frac{dn}{n}. \quad (4.34)$$

Substituting Eqs. 4.32 and 4.34 into Eq. 4.31

$$N_0 = 4\sqrt{2}\pi l^3 \int_0^{n_0} \left[ \ln \left( \frac{n_0}{n} \right) \right]^{1/2} dn. \quad (4.35)$$
Therefore, the scaled probability density function of the initial BEC density $n$ is defined as

$$f(n) = 4\sqrt{2}\pi l_r^3 \left[ \ln \left( \frac{n_0}{n} \right) \right]^{1/2}, \quad (4.36)$$

so that

$$N_0 = \int_0^{n_0} f(n) dn. \quad (4.37)$$

Figure 4.18: The scaled probability density function of the initial BEC density $f(n)$.

Figure 4.18 plots of $f(n)/(4\sqrt{2}\pi l_r^3)$ as the function of scaled density $n/n_0$.

In the density treatment, the evolution of the total number of atoms in the BEC (Eq. 4.29) can be rewritten as

$$N(t) = \int_0^{n_0} \tilde{f}(t, n) dn, \quad (4.38)$$
where \( \tilde{f}(t, n) = f(n)\tilde{n}(t, n)/n \) is the evolution of the scaled probability density function \( f(n) \). It is noticed that when \( t = 0 \), there is

\[
N(t = 0) = \int_{0}^{n_0} \tilde{f}(t = 0, n)dn = \int_{0}^{n_0} f(n)\tilde{n}(t = 0, n)dn = \int_{0}^{n_0} f(n)dn = N_0.
\]

(4.39)

Figure 4.19 shows the 3D plot and top-view plot of \( \tilde{f}(n)/(4\sqrt{2}\pi l_r^3) \). As expected, the Rabi frequency increases significantly with condensate density.

Here I use this model to qualitatively explain the influence of the inhomogeneous density on the data. Figure 4.20 shows the data of the evolution of BEC numbers with three different detunings, which will be discussed in detail in Sec. 4.5. I also plot the simulation results with the contribution of each density component. All simulation curves, particularly the blue one, can qualitatively reproduce the data well, which indicates that the loss of contrast of Rabi oscillations can be ascribed to the variation in Rabi frequencies due to the inhomogeneous density. It is noticed that although an artificial detuning (in square brackets) is used for better fit, this model can qualitatively demonstrate that the inhomogeneous density results in the reduction of contrast of Rabi oscillations. A more careful description of the data given by the many-body theory (Eqs. 4.6) will be discussed in Sec. 4.5.

(c) Characteristic average density

Condensate density inhomogeneity is treated with a local density approximation, where the characteristic average density is introduced and derived here.

The local loss equation for PAS

\[
\dot{n}(r) = -\beta|n(r)|^2,
\]

(4.40)
Figure 4.19: 3D plot (top) and top-view plot (bottom) of $\tilde{f}(n)/(4\sqrt{2} \pi l^3)$. 
Figure 4.20: Qualitatively explanation of the reduce of contrast of Rabi oscillations. The same data of scaled BEC number versus time as Fig. 4.24 which will be discussed in Sec. 4.5. Data explores zero detuning and different blue detunings. Legend gives the detuning with respect to the low-intensity PA resonance position, the detuning from the long-time resonance position including ac Stark shift (in parentheses), and the artificial detuning (in square brackets) used in simulation for better fit, followed by the intensity of PA beams. Three simulation curves are calculated with the contribution of each density component.
where the density distribution \( n(r) = n_0f(r) \) with the peak density \( n_0 \).

But in experiments we measure the number of atoms in BEC

\[
N = \int n(r)d^3r = n_0\int f(r)d^3r = n_0V_{\text{eff}},
\]

(4.41)

where \( V_{\text{eff}} = \int f(r)d^3r \) is the first effective volume [133].

\[
\dot{N} = \int \dot{n}(r)d^3r = -\beta \int [n(r)]^2d^3r = -\beta n_0^2\int [f(r)]^2d^3r = -\beta n_0^2V_{2\text{eff}},
\]

(4.42)

where \( V_{2\text{eff}} = \int [f(r)]^2d^3r \) is the second effective volume.

Combining Eqs 4.41 and 4.42 gives

\[
\frac{\dot{N}}{N} = -\beta \frac{n_0V_{2\text{eff}}}{V_{\text{eff}}} = -\beta n_{\text{eff}},
\]

(4.43)

where the single characteristic average density

\[
n_{\text{eff}} = n_0V_{2\text{eff}}/V_{\text{eff}}.
\]

(4.44)

Given \( f(r) = e^{-r^2/(2l^2)} \) (Eq. 4.28), \( n_{\text{eff}} \) is calculated to be \( n_0/(2\sqrt{2}) \).

4.5 Results

4.5.1 AC Stark Shift of the PAS Transition for Long Excitation Times

In order to interpret the PA excitation spectra, it is important to understand the long-time and transient behavior of the ac Stark shift of the transition due to the PA laser. The ac Stark shift arises from the interaction between the laser electric field and the induced dipole moment. The Stark shift will saturate with a long enough time to completely establish the induced dipole moment.
Figure 4.21: ac Stark shift due to PA light for long exposure times compared to the time scale for transient effects. Exposure times and intensities of the PA laser are given in the legend. A fit to the line centers (inset) yields $(-275 \pm 30) \text{ kHz/(W/cm}^2\text{)}$. Zero detuning is the line center at low intensity.
Figure 4.21 shows the shift for long excitation times, which is large compared to the linewidth for our highest intensities. Spectra are fit with Gaussians. The inset lists measured $\delta'$ at different laser intensities, from which the linear fitting gives rise to the measured value of $\delta'/(2\pi I) = (-275 \pm 30) \text{kHz/(W/cm}^2)$. It is in agreement with the value $\delta'/(2\pi I) = (-251 \pm 67) \text{kHz/(W/cm}^2)$ measured in a thermal gas in Ref. [103]. Our stated uncertainty is statistical. There is additional systematic uncertainty arising from the transient effects on the spectra discussed below, but these effects are minimized by working at a relatively long exposure time (> 3 $\mu$s).

4.5.2 PAS Spectra of BEC Number for Different Excitation Times

(a) Transient Shifts and Broadenings

Against the benchmark of the shift at long exposure times, we examine the transient behavior at shorter times. A dynamic shift at early times was predicted in Ref. [64], and to our knowledge, we present the first experimental observation of this phenomenon. Figure 4.22 shows a sample of high intensity PA spectra, along with an inset showing the line center as a function of exposure time. The ac Stark shift is small initially and increases on a few-microsecond time scale.

An expression for the transient behavior of the shift was derived in Ref. [64] under the sudden approximation that the amplitude in the molecular state reaches equilibrium on a much faster time scale than the observations. However, this is not valid in our experiments because the molecular-state occupation is changing during the course of the experiment. A phenomenological fit of the trend to $\Delta \nu(t) = \Delta \nu_0 [1 - e^{-t/t_{\text{exp}}}]$ yielding $t_{\text{exp}} = 1.24 \mu$s is shown in Fig. 4.22 (inset).
Figure 4.22: Spectra of the number of atoms remaining in BEC for 2.4 W/cm\(^2\) excitation intensity and various excitation times. Zero frequency detuning is the PA resonance position at vanishing PA laser intensity but including the ac Stark shift due to the trapping laser. Lines are predictions of Eqs. 4.6 for \(\ell_{opt}/I = 5000 \, a_0/(W/cm^2)\), showing the transient shift and oscillatory behavior for blue detuning. The inset shows the center frequency versus time, found by fitting Gaussians to the spectra, and the solid line is an exponential fit to the trend. Pascal Naidon performed the simulation here.
This transient behavior has an intuitive description. The ac Stark shift arises from the interaction between the laser electric field and the induced dipole moment reflecting coherences between molecular and continuum states. After the laser is turned on, time is required to transfer population amplitude along the internuclear axis and create coherences, and the increasing ac Stark shift over the first few microseconds in our experiment tracks this evolution. If Eqs. 4.6 are solved numerically with $C^\text{dyn}_k = 0$ set artificially, no transient shift is observed, but the full equations predict the shift well (Fig. 4.22).

Weak, coherent excitation of a two-level system is described by a sinc$^2$ spectrum with FWHM $\sim 1.2/t$ Hz from the energy uncertainty due to the short time of the laser pulse $t$ [106]. In Fig. 4.22, calculated widths for 1.3 $\mu$s, 2.3 $\mu$s, and 3.3 $\mu$s are 0.92 MHz, 0.52 MHz, and 0.36 MHz, respectively, which match the widths of the spectra of 1.08 MHz, 0.47 MHz, and 0.49 MHz, respectively. Interaction-time broadening is the dominant contribution. In Sec. 4.4.3, the wavenumber $k_r$ is estimated to be on the order of $\Delta p/\hbar = 1/(\sqrt{2}\ell_r) = 8.8 \times 10^5$ m$^{-1}$. For 2.4 W/cm$^2$, the stimulated line width is $\Gamma_{\text{stim}} = 2k_r\ell_{\text{opt}}\Gamma_{\text{mol}} = 2\pi \times 17$ kHz by taking $\ell_{\text{opt}} = 2.4$ W/cm$^2 \times 5000 a_0/(W/cm^2) = 12000 a_0$.

(b) Oscillations

Another feature expected from an analogy with the coherent excitation of a simple two-level atomic system is oscillatory behavior, which is observed in experiment and simulation near zero detuning in Fig. 4.22 (around 600 kHz blue detuning from the resonance position at long times, including ac Stark shift). In the case of coherent PA, oscillations are between atomic and molecular populations. In contrast with the
atomic analogy, however, we observe distinct asymmetries and detect no oscillations at red detuning. This asymmetry is well produced in calculations of the spectra based on Eqs. 4.6, and it can be understood in the dressed atom picture (Fig. 4.23). For blue detuning ($\delta - \delta' > 0$) (Fig. 4.4), the molecular state is below the continuum and stable enough to contribute to oscillations (Fig. 4.23(a)). For red detuning ($\delta - \delta' < 0$), the molecular state is embedded in the continuum and unstable against decay into noncondensate pairs, where no Rabi oscillations are observed (Fig. 4.23(b)). Similar behavior is expected for MFRs [38].

4.5.3 BEC Number vs Time During Coherent PAS

The oscillatory phenomenon is one of the most interesting aspects of our observations, as it directly manifests coherence between atomic and molecular states and has been the subject of significant theoretical attention [64, 65, 67–75]. So, we focus on the variation of condensate population versus time for different detunings in Fig. 4.24, where experimental parameters are summarized in Table 4.2. As predicted in Ref. [64], at the very beginning, PA converts the initial atomic condensates to the excited molecular state, and the loss rate coefficient increases from zero, which results in the initial quadratic variation of the number of atomic condensates with time, with loss independent of detuning. This phenomena is shown in Fig. 4.24 at the extremely short time, which is a signature of the coherence of the excitation. With increasing time, the number at larger detuning reaches a minimum and oscillations set in, which peels away from lines with smaller detunings.

Clear oscillations are visible for the largest blue detuning, which corresponds to the region of oscillatory behavior in Fig. 4.22. A solution of Eqs. 4.6, which includes
Figure 4.23: Coherent PA described by the dressed state picture. (a) PA described by the dressed state picture corresponding to Fig. 4.4, the blue-detuned PA, where $\Delta E < 0$. The dressed molecular state $|n_b = -2, (n-1)h\nu\rangle$ is below the continuum and stable enough to contribute to oscillations. (b) PA described by the dressed state picture corresponding to the red-detuned PA, where $\Delta E > 0$. The dressed molecular state $|n_b = -2, (n-1)h\nu\rangle$ is embedded in the continuum and unstable against decay into noncondensate pairs. Thus no Rabi oscillations are displayed here.
Figure 4.24: Number of BEC atoms versus time. Experimental parameters are summarized in Table 4.2. The number is scaled to the number with no PA (∇:8800, ○:8800, □:6700) and time is scaled to the inverse Rabi frequency for average density, $\bar{n} = n_0/\sqrt{8}$, $2\pi/\bar{\Omega}_R = (∇:7.1, ○:7.1, □:10.5) \mu s$. The legend states the detuning with respect to PA resonance at low intensity and (in parentheses) the detuning with respect to the long-time resonance position including ac Stark shift, followed by the intensity. Rabi oscillations are clearest for large blue detuning from the ac-Stark-shifted resonance (∇). Solid lines are predictions of Eqs. 4.6 performed by Pascal Naidon for $\ell_{\text{opt}}/I = 5000a_0/(W/cm^2)$ and peak density at the upper limit of the measurement uncertainty. The universal prediction (dash-dotted line) is given by Eq. (9) in Ref. [64]. The inset shows the calculated population of atoms in the excited-state molecular BEC and ground-state atoms in noncondensate modes.
### Table 4.2: Experimental Parameters used in Fig. 4.24

<table>
<thead>
<tr>
<th>Data</th>
<th>BEC num</th>
<th>$\sigma_\rho$ [µm]</th>
<th>$\sigma_z$ [µm]</th>
<th>$n_0$ [m$^{-3}$]</th>
<th>$n_{\text{eff}}$ [m$^{-3}$]</th>
<th>$I$ [W/cm$^2$]</th>
<th>$\delta$ [MHz]</th>
<th>$\delta - \delta'$ [MHz]</th>
<th>$\Omega_{\text{peak}}/2\pi$ [MHz]</th>
<th>$\Omega_R/2\pi$ [MHz]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Blue</td>
<td>8800</td>
<td>0.571</td>
<td>0.537</td>
<td>$3.2 \times 10^{21}$</td>
<td>$1.1 \times 10^{21}$</td>
<td>2.06±0.12</td>
<td>0.07±0.04</td>
<td>0.64±0.04</td>
<td>0.24</td>
<td>0.14</td>
</tr>
<tr>
<td>Red</td>
<td>8800</td>
<td>0.571</td>
<td>0.537</td>
<td>$3.2 \times 10^{21}$</td>
<td>$1.1 \times 10^{21}$</td>
<td>2.06±0.12</td>
<td>-0.43±0.04</td>
<td>0.14±0.04</td>
<td>0.240</td>
<td>0.14</td>
</tr>
<tr>
<td>Green</td>
<td>6700</td>
<td>0.678</td>
<td>0.617</td>
<td>$1.5 \times 10^{21}$</td>
<td>$0.5 \times 10^{21}$</td>
<td>1.90±0.18</td>
<td>-0.55±0.05</td>
<td>-0.03±0.05</td>
<td>0.16</td>
<td>0.095</td>
</tr>
</tbody>
</table>
Figure 4.25: The same data as Fig. 4.24 and the results predicted by Eqs. 4.6 with setting $C_{\text{dyn}}^k = 0$ artificially. Pascal Naidon performed the simulation.
coupling to the continuum and associated transient effects, fits the data well. Again, if we numerically solve Eqs. 4.6 by setting $C_{k}^{\text{dyn}} = 0$ artificially, although the results still display Rabi oscillations, but the Rabi frequencies disagree with the data (Fig. 4.25).

Density inhomogeneity is treated with a local density approximation (Sec. 4.4.4). The variation in Rabi frequency resulting from the inhomogeneous density in the condensate accounts for nearly all of the reduction in contrast to the oscillations (Fig. 4.19). We expect smaller contributions from natural decay of the molecular state [86, 134], laser linewidth, and atom-molecule and molecule-molecule collisions.

Furthermore, as illustrated in Fig. 4.19, there is a trend that data at smaller detuning (○) oscillates slower than larger one (▽), and there is no visible oscillation near the resonance (□). From an analogy with single-particle-two-level systems, one would expect the effective Rabi frequency $\Omega_{\text{eff}} = \sqrt{\Omega_{R}^{2} + (\delta - \delta')^{2}}$. In other words, increasing the value of detuning $|\delta - \delta'|$ increases the effective Rabi frequency, and decrease the amplitude of the oscillation, which exactly agrees with the trend of the three sets of data. As to the near-resonance data, another factor contributing to the lack of visibility of Rabi oscillations is a fundamental difference between coherent atom-molecule conversion and Rabi oscillations in single-particle systems. As discussed in Ref. [69], in coherent photoassociation there is no oscillation of population exactly on resonance, and the relative population in the molecular and atomic states are described with elliptical functions instead of the more familiar sinusoidal. This arises from the basic structure of the atom-molecule equations for a many-body or two-body system and appears even if the coupling to the continuum is neglected.

The best fit in Fig. 4.24 requires a density at the upper limit of experimental uncertainty and $\ell_{\text{opt}} / I = (5000 \pm 1000) a_{0}/(W/cm^{2})$. This is significantly less than
the $\ell_{\text{opt}}$ values calculated directly from knowledge of the molecular potentials [103] and extracted from collisional effects of the optical Feshbach resonance in a BEC [134] and a thermal gas [103], which is a point for further investigation.

Rabi oscillation corresponds to the return of population to the atomic condensate, which is only possible if a coherent many-body molecular population exists. We interpret this as the presence of a molecular condensate [64, 65]. The simulation in Fig. 4.24 (inset) suggests the presence of as many as 2000 atoms in the excited molecular electronic state remaining as an excited-state molecular condensate after the laser is turned off. The lifetime of this molecular condensate is sensitive to unknown molecule-molecule and atom-molecule inelastic collision rates as well as natural and laser-induced decay. This represents the first demonstration of Rabi oscillations with photoassociation and the first creation of a condensate of molecules in an excited electronic state.

Figure 4.24 (inset) also shows a significant population in atomic, noncondensate modes, which is in the form of correlated atomic pairs. This process becomes more dominant at higher laser intensity, and it ultimately limits the population of molecules [70, 73]. As introduced in Ref. [64], there are three regimes during the evolution of the loss rate coefficient. The decay rate in the second regime (Eq. (9) in Ref. [64]), where the molecular condensates are driven back to the atom-pair continuum, is only determined by the mass and density of the atoms and has a universal behavior, which is heralded by a large atomic, noncondensate population here. The experimentally observed decay near resonance in Fig. 4.24 closely matches the universal prediction (dash-dotted line).
4.6 Comparing Coherent One-Color PAS to Magnetic Feshbach Creation of Molecules

The formation of a molecular condensate through coherent photoassociation is analogous to formation using magnetic Feshbach resonances [51]. It would be very interesting to explore this connection further, such as investigating the nature of broad versus narrow optical Feshbach resonances and the related molecular binding energy [38].

4.6.1 Dressed Atom Picture

As introduced in previous sections, the one-color PA can be described by the dressed-atom picture, so that it is more convenient to compare to MFR.

Figure 4.23 is the dressed-state picture of the coherent PA. The dressed state of excited molecular state supported by the $^1S_0+^3P_1$ potential with $n-1$ photons has the energy close to that of initial collision state $^1S_0+^1S_0$ with $n$ photons. The energy difference is modified by the detuning of the PA laser with respect to the PA resonance, so that the accompanying coupling between these two states varies, which is analogous to the adjustment via the strength of the magnetic field in MFRs.

4.6.2 Broad versus Narrow Resonance

The complex scattering length $\tilde{a}$ in a Feshbach resonance is given by [38]

$$\tilde{a} = a - ib = a_{bg} + \frac{a_{bg}\Gamma_0}{-E_0 + i(\gamma/2)},$$

(4.45)

where the resonance position $E_0$ is tuned by the strength $B$ of a magnetic field in MFRs or detuning $\delta$ of a laser field in OFRs, and $\Gamma_0$ is the resonance strength. The decay rate $\gamma$ includes all decay channels, which is zero or so small as to be negligible.
in MFRs, but cannot be ignored in OFRs since they are always accompanied by decay of an excited molecular state. There is a crucial difference in resonance width between MFRs and OFRs. The resonance width of OFRs $\Gamma_0(I)$ is proportional to the laser intensity $I$, which is controllable experimentally. Thus in OFR experiments, it is possible to change the resonance width arbitrarily by changing $I$. However, there is no analogous adjustability in MFRs. The resonance width of MFRs $\Gamma_0/\delta \mu$ with the magnetic moment difference of the two coupled states $\delta \mu$, is fixed and governed by the interatomic forces between the two atoms. In a word, OFRs provide an additional freedom of controlling the coupling strength.

![Graph](image)

Figure 4.26: Total scattering length $a$ versus detuning with respect to the long-time resonance with $\ell_{\text{opt}}/I = 5 \times 10^3 a_0/(\text{W/cm}^2)$ and $I = 2.4 \text{ W/cm}^2$. The inset zooms in the 200-1000 kHz blue-detuned region.

Moreover, it is worth stressing that unlike MFRs where the scattering length
diverges in the vicinity of the resonance, the scattering length in OFRs is finite (Fig. 4.26) for any detuning and has the maximum and minimum of \( a_{\text{bg}}(1 \pm \Gamma_0/\gamma) \).

### 4.6.3 Molecular Binding Energy

In a MFR, the atom-molecule oscillation frequency corresponds to the binding energy of the molecular state [56, 126]. In a PA, the GP model does a poor job in describing the molecular binding energy, since it is still a kind of bare molecular energy. We think it is plausible that the oscillation frequency calculated from Eqs. 4.6 matches the molecular binding energy in the dressed atom picture, but further simulation is needed to prove it. Further measurements may also allow experimental check of predictions of non-linear, non-sinusoidal population oscillations at small detuning for coherent PA [67–69].

### 4.6.4 Strongly Interacting Bose Gas

Viewing the PA transition as an optical Feshbach resonance (OFR) highlights several extreme features of the system. At detunings from resonance on the order of \( \gamma_m \), the scattering length due to the OFR for the highest intensity used here is extremely large \([a \sim \ell_{\text{opt}} = 12000 a_0 = 640 \text{nm} \, (\text{Fig. 4.26})]\). This is much larger than the range of the molecular potential, given by the van der Waals length \( R_{\text{vdW}} \equiv (MC_6/16\hbar^2)^{1/4} = 75 a_0 \), where \( C_6 \) is the van der Waals dispersion coefficient for the ground-state potential, and much larger than the typical interparticle spacing \( n^{-1/3} \approx 100 \text{nm} \). This implies the system is strongly interacting and should be described as a highly correlated many-body state, rather than a gas of atoms and molecules [52]. For the detuning at which Rabi oscillations are observed in this study, \( a \approx 500 a_0 \, [\text{Fig. 4.26(inset)}] \) and
$na^3 \ll 1$ (Fig. 4.27).

![Graph of $na^3$ versus detuning with respect to the long-time resonance with $\ell_{opt}/I = 5 \times 10^3 a_0/(W/cm^2)$ and $I = 2.4 W/cm^2$.](image)

Figure 4.27: $na^3$ versus detuning with respect to the long-time resonance with $\ell_{opt}/I = 5 \times 10^3 a_0/(W/cm^2)$ and $I = 2.4 W/cm^2$. 
Chapter 5

Analysis of intercombination transition photoassociation spectroscopy of $^{86}$Sr

Photoassociation spectroscopy (PAS) is a common technique to determine the bound-state energy levels of diatomic molecular potentials [33]. In this process, two colliding atoms absorb a photon to create a bound, electronically excited molecule. This tool has been applied to alkali-metal and alkaline-earth atoms. A series of PAS with Sr have been performed near broad $^1S_0^1P_1$ transition [135–137]. Later, Zelevinsky et al. [86] and Stellmer et al. [63] reported $^1S_0^3P_1$ intercombination-transition PAS with $^{88}$Sr and $^{84}$Sr in an optical lattice, respectively. In this chapter, I will report the determination of positions of the four least-bounded states of the $^1S_0^1P_1$ excited molecular potential of the third boson isotope of strontium, $^{86}$Sr, which affords an opportunity to test mass scaling of the excited molecular potentials [138]. It can guide future experiments for $^{86}$Sr.

5.1 Apparatus and Experimental Procedure

Figure 5.1 shows the schematic drawing of the experimental setup. To perform photoassociation, we prepare ultracold $^{86}$Sr atoms in an optical dipole trap via laser cooling and trapping techniques similar to those used for other Sr isotopes [82, 133]. The ODT is formed by the intersection of two mutually perpendicular beams focused to waists ($e^{-2}$ intensity radii) of 100 µm. Both beams are generated from a 1064-nm,
linearly-polarized, multilongitudinal-mode fiber laser. A period of forced evaporation to a trap depth of $3.6 \, \mu K$ yields $3 \times 10^5$ $^{86}$Sr atoms at a temperature of 400nK and peak density of $10^{13} \, \text{cm}^{-3}$.

Figure 5.1: Schematic drawing of the $^{86}$Sr PAS experiment setup.

The PA beam is derived from a 689 nm master-slave diode laser system (Fig. 5.1) that has a linewidth of approximately 10kHz. Short term stability is provided by locking the laser frequency to a moderate finesse ($\mathcal{F} = 2000$) optical cavity, and long term stability is assured through saturated absorption spectroscopy of the $^1S_0$-
3$^3P_1$ atomic transition in a vapor cell. The PA beam is red detuned with respect to the atomic transition using acousto-optic modulators (AOMs) and transported to the atoms through a single-mode optical fiber. In the interaction region, the beam is linearly polarized, with a waist of 700 µm and peak intensity up to 50 mW/cm$^2$. During the application of the PA beam, to eliminate the ac-Stark shift due to ODT beams, the ODT is modulated with 50% duty cycle, period of 462 µs, and a peak trap depth of 7.2 µK. The PA beam is applied when the ODT is off. Total PA time per single trapped atom sample is varied from 16 to 830 ms, depending upon the transition, to obtain a peak atom loss due to PA of approximately 50% with minimal change in sample temperature. The number of atoms and sample temperature are determined with time-of-flight absorption imaging using the $^1S_0-^1P_1$ transition at 461 nm.

5.2 Line Shape Analysis

In experiments, we measured the number of leftover atoms in the trap after applying a PAS beam for the interaction time of $t$. The time evolution of the number of atoms is given by [20]

$$N(t) = \frac{N_0e^{-\Gamma_1 t}}{1 + \frac{2N_0KV_q}{\Gamma_1 V_1^2}(1 - e^{-\Gamma_1 t})},$$  \hspace{1cm} (5.1)

where $N_0$ is the number of atoms without applying PAS beams, $\Gamma_1$ is the one-body loss rate due to background collisions and off-resonance scattering from the PAS beams, $K$ is the effective collision event rate constant, and $V_q$ ($q = 1$ and 2) are the effective volumes defined by

$$V_q = \int_V d^3r e^{-\frac{qU(r)}{k_BT}},$$  \hspace{1cm} (5.2)
with the trap potential \( U(r) \), and the sample temperature \( T \).

Assuming constant sample temperature, and loss described by \( \dot{n} = -2Kn^2 \) and considering \( \Gamma t \ll 1 \) in the experiment, Eq. 5.1 can be simplified to

\[
N(t) = \frac{N_0}{1 + 2N_0KtV_2/V_1^2},
\]

from which we can get

\[
K = \left( \frac{1}{N(t)} - \frac{1}{N_0} \right) \frac{V_1^2}{2V_2t}.
\]

With the parabolic approximation, the trap potential of an ODT has the form (Eq. (C1) in Ref. [120])

\[
U(r) = \frac{1}{2} m(\omega r)^2 \Theta(R_0 - r) = \epsilon_t \left( \frac{r}{R_0} \right)^2 \Theta(R_0 - r),
\]

where \( \Theta(r) \) is the Heaviside step function, and \( R_0 = \sqrt{2\epsilon_t/m\omega^2} \) is the boundary of the trap with the trap depth \( \epsilon_t \). Eq. (C8) in Ref. [120] gives the analytic expressions with high-\((\epsilon_t/k_B T)\) approximation for effective volumes

\[
V_q = \frac{4\pi R_0^3}{3} \left( \frac{k_B T}{q\epsilon_t} \right)^{3/2} \Gamma(5/2) = \pi^{3/2} R_0^3 \left( \frac{k_B T}{q\epsilon_t} \right)^{3/2} = \left( \frac{2\pi k_B T}{qm\omega^2} \right)^{3/2},
\]

where \( \Gamma(a) \) is the Gamma function.

Substituting \( V_1 \) and \( V_2 \) from Eq. 5.6 into Eq. 5.4 yields

\[
K = \frac{4}{t} \left( \frac{1}{N(t)} - \frac{1}{N_0} \right) \left( \frac{\pi k_B T}{m\omega^2} \right)^{3/2},
\]

which allows us to directly extract spectra of \( K \) from the loss spectra of atoms. Figure 5.2 is an example near \( 0_u(n = -2) \) PA line of \(^{86}\text{Sr}\).

Now we fit spectra of collision event rate constant \( K \) (Fig. 5.2 (b)) using the model introduced in Ref. [20], which gives

\[
K = \frac{1}{V_2} \int_V d^3r \ e^{-\frac{2U(r)}{\hbar^2}} \times \frac{1}{hQ_T} \int_0^{\epsilon_t-U(r)} dE_g |S_{1g}|^2 e^{-E_g/k_B T},
\]
Figure 5.2: Spectroscopy of the $0_u(n_b = -2)$ PA line of $^{86}$Sr. (a) Atom number versus laser detuning from the one-photon $^1S_0 - ^3P_1$ atomic transition for interaction time $t = 59.4\text{ms}$. (b) The effective collision event rate constant derived from the atom loss using Eq. 5.7. (c) Temperature versus detuning shows little variation, supporting the assumption of constant sample temperature.
where $h$ is the Planck constant, $Q_T = (2\pi k_B \mu/h^2)^{3/2}$ is the partition function for reduced mass $\mu = m/2$, the integral of kinetic energy $E_g$ is truncated by the local trap depth $\epsilon_t - U(r)$, and $|S_{1g}|^2$ is the scattering probability. Furthermore, our tests have showed that neglecting truncation of the energy integral results in a very small correction, so we have

$$K = \frac{1}{hQ_T} \int_{-\infty}^{+\infty} dE_g |S_{1g}|^2 e^{-E_g/k_B T}.$$  \hfill (5.9)

Ciurylo et al. [18] point out that the Doppler broadening and photon recoil significantly contribute to the shape of PA lines when the atomic temperature $T$ is lower than the atomic recoil temperature $T_R = (h/\lambda)^2/(mk_B)$ with the wavelength of the PA laser $\lambda = 689\,\text{nm}$, which is the case in our experiments where $T \sim 400\,\text{nK} < T_R = 460\,\text{nK}$. Now Eq. 5.9 can be rewritten as [18]

$$K = \frac{k_B T}{hQ_T} \int_{-\infty}^{+\infty} dy e^{-y^2} \int_{0}^{\infty} dx x e^{-x^2} \mathcal{L}(f, y, x^2),$$  \hfill (5.10)

where $x$ and $y$ are dimensionless variables, and

$$\mathcal{L}(f, y, x^2) = \frac{\eta \gamma_m \gamma_s/(2\pi)^2}{(f + y\Delta_D + x^2\Delta_T - E_b/h - E_{\text{rec}}/h)^2 + (\eta' \gamma_m/4\pi)^2}$$  \hfill (5.11)

with the thermal width $\Delta_T = k_B T/h$, the Doppler width $\Delta_D = \sqrt{k_B T/m}/\lambda$, the natural linewidth of the excited molecular level $\gamma_m = 2\pi \times 15\,\text{kHz}$, and the stimulated width $\gamma_s = 2(\sqrt{2\mu x^2 \Delta_T/h}) \gamma_m \ell_{\text{opt}} \ll \gamma_m$ where $\ell_{\text{opt}}$ is the optical length. Here $hf$ is the energy of the photon, $E_b$ is the fitting PA line center including the molecular ac Stark shift due to PA beams, and $E_{\text{rec}} = (h/\lambda)^2/(4m)$ is the photon recoil energy of an isolated atom. The parameter $\eta \geq 1$ accounts for the extra molecular losses observed in OFR experiments [41,103], and $\eta' \gamma_m = \eta' \gamma_m + \gamma_{\text{laser}}$ with the line width of
Figure 5.3: Fitting of $\ell_{\text{opt}}$ and PA line centers with $I$ for $0_u (n_b = -2)$ PA line. (a) Linear fitting of $\ell_{\text{opt}}$ with the beam intensity $I$ for $0_u (n = -2)$ PA line gives $\ell_{\text{opt}}/I = (1.5 \pm 0.3) \times 10^4 a_0/(\text{W/cm}^2)$. $\eta_{\text{upper}} = 1.5$, and $\gamma_{\text{laser}}^{\text{upper}} = 2\pi \times 7.5\text{kHz}$. The error bars are entirely systematic reflecting the upper and lower limits for $\ell_{\text{opt}}$ as described in the text. (b) Linear fitting of PA line centers with $I$ for the same PA line gives zero-intensity molecular center $\nu_0$ at -44.246 MHz.
the PA laser $\gamma_{\text{laser}}$. We can only determine $E_b$, $\eta' = \eta + \gamma_{\text{laser}}/\gamma_m$, and $\eta \ell_{\text{opt}}$, and cannot independently determine $\eta$, $\ell_{\text{opt}}$, and $\gamma_{\text{laser}}$ because of mutual restriction as showed in Eq. 5.11. However, we can estimate the limits of these parameters. Furthermore, as introduced above, the ODT-induced-shift term [20] disappears since the ODT is modulated with 50% duty cycle and the PA beam is applied when the ODT is off.

Using Eq. 5.10, the fitting results of the spectrum of collision event rate constant $K$ is shown in Fig. 5.2(b). The PA line center shifts linearly with the intensity of PA beams $I$ [18], and we can determine the zero-intensity resonance center of PA lines $\nu_0$ by linearly fitting intensity-dependent data of PA resonance centers (Fig. 5.3(b)). $\eta_{\text{upper}}$, the upper limit of $\eta$ and the lower limit of $\ell_{\text{opt}}$ can be determined by fixing $\gamma_{\text{laser}} = 0$, while $\gamma_{\text{laser}}^{\text{upper}}$, the upper limit of $\gamma_{\text{laser}}$ and the upper limit of $\ell_{\text{opt}}$ can be determined by fixing $\eta = 1$. The error bars in Fig. 5.3(a) reflect these upper and lower systematic uncertainties in $\ell_{\text{opt}}$. Considering the optical length $\ell_{\text{opt}}$ is directly proportional to the intensity $I$ of PA beams [103], $\ell_{\text{opt}}/I$ can be extracted by linearly fitting the values of $\ell_{\text{opt}}$ at different $I$ (Fig. 5.3(a)). Values of these parameters from the fitting are summarized in Table 5.1. It is noted that data for 1_u potential were taken in deep traps and temperatures increased significantly when PA beams were applied, so we cannot assume a constant temperature, which is required in the fitting process described here. We just provide the value with a larger uncertainty.

Now we will estimate the uncertainty of $\nu_0$. The atomic resonance spectrum can be fitted with a simple Lorentz profile to determine the atomic resonance position with an uncertainty of 5 kHz (Fig. 5.4). $E_b$ is extracted by fitting spectra of $K$ (Fig. 5.2(b)) with an uncertainty of 3 kHz. Moreover, there is an uncertainty of 2 kHz from the RF synthesizer. Thus, the total uncertainty of $\nu_0$ is determined to be 10 kHz.
Table 5.1: Values of parameters of $^{86}\text{Sr}$ PA lines extracted from the fitting

<table>
<thead>
<tr>
<th>$n$</th>
<th>$^{86}\text{Sr}$ Potential</th>
<th>$\nu_0$ [MHz]</th>
<th>Theory $\nu_0$ [138] [MHz]</th>
<th>$l_{\text{opt}}/I$ $[a_B/\text{W/cm}^2]$</th>
<th>$\eta^\text{upper}$ $[\text{kHz}]$</th>
<th>$\gamma^\text{upper}_{\text{laser}}/\pi$ [MHz/($\text{W/cm}^2$)]</th>
<th>$\Delta\nu/I$ [MHz/($\text{W/cm}^2$)]</th>
</tr>
</thead>
<tbody>
<tr>
<td>-1</td>
<td>0_u</td>
<td>-1.633(10)</td>
<td>-1.541</td>
<td>$(3.8 \sim 6.5) \times 10^4$</td>
<td>1.7</td>
<td>10.5</td>
<td>7.059</td>
</tr>
<tr>
<td>-2</td>
<td>0_u</td>
<td>-44.246(10)</td>
<td>-44.208</td>
<td>$(1.2 \sim 1.8) \times 10^4$</td>
<td>1.5</td>
<td>7.5</td>
<td>0.948</td>
</tr>
<tr>
<td>-1</td>
<td>1_u</td>
<td>-159.984(50)</td>
<td>-160.02</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>-3</td>
<td>0_u</td>
<td>-348.742(10)</td>
<td>-348.795</td>
<td>$(5.7 \sim 10.3) \times 10^2$</td>
<td>1.8</td>
<td>12.0</td>
<td>0.151</td>
</tr>
</tbody>
</table>
Figure 5.4: Atomic resonance and a series of PA lines.
Chapter 6

Conclusion

Alkaline-earth metal atoms, such as strontium, feature an extremely narrow intercombination, which has facilitated many fields of ultracold physics, for example, the development of the next generation of frequency standards, studies of novel magnetism, and tests of theoretical models.

In the first experiment presented in this thesis, this feature enables us to demonstrate the optical Feshbach resonance by controlling collapse and expansion of a condensate. It is the first time to use an optical Feshbach resonance to control the expansion and collapse of a condensate, similar to experiments that are often done with magnetic Feshbach resonances. In this work we have explored the regime of large-detuning with respect to a PA resonance, and shown that significant interaction changes can be achieved with a low atom loss. Losses are so low that the OFR can be performed for times on the order of the hydrodynamic timescale of condensate expansion.

The original peak density of the condensate is extremely high in this OFR experiment due to the attractive interactions. In samples with density comparable to that corresponding to one atom per site in an optical lattice, it is expected to increase lifetime where larger OFR effect could be achieved. Alternatively, a more deeply bound excited molecular state, for example, the PA line at -1.08 GHz in the same molecular potential [86] with a decent value of $l_{opt}$ can be used in future experiments, which
has the advantage of greater suppression of atomic light scattering and reduced background two-body loss due to the large detuning from the atomic resonance. This paves a way to perform a lot of possible experiments involving optical Feshbach resonances and quantum fluids.

The second experiment also benefits from the intercombination transition of alkaline-earth quantum gases, which opens a new regime for optical production of molecules and create atom-molecule coherence, Rabi oscillations between atomic and molecular condensates, and strongly-interacting many-body systems. Rabi oscillations between atomic and molecular condensates have never been observed with photoassociation. For the first time, achieved this long-sought-after goal using one-color PA, bearing an analogy to molecule production with a magnetic Feshbach resonance. But the PA method provides one more freedom to control the coupling strength, i.e. besides the detuning of a laser, its intensity can also modify the strength of the atom-molecule coupling, which is not possible with a magnetic Feshbach resonance. Furthermore, in the vicinity of the optical resonance, we can approach a new regime of universal two-body physics in which the PA loss of atoms in a condensate is only determined by the atomic mass. We also observe rare transient shifts, broadenings, and asymmetries of the excitation spectrum that are the signal of the coherent processes. These phenomena have never been observed before due to the short lifetime of an excited molecular state in alkali-metal systems. This work demonstrates a novel method to produce molecular condensates, accesses the unexplored regime predicted by theoretical models, and observes striking behaviors related to the new regime.

Similar to the OFR experiments, it would also be interesting to perform similar experiments in an optical lattice so that the molecular loss due to collisions can
be suppressed and the density-inhomogeneity factor can be ignored to simplify the theoretical simulations. Moreover, narrower optical transitions, such as optical clock transitions in strontium and ytterbium or the $^1S_0-^3P_1$ transition in calcium [139], may further expand the possibilities with coherent one-color PA to achieve more significant phenomena.
Appendix A

New Imaging System

A.1 Design

A high quality imaging system is crucial for ultracold physics experiments. Here, a high resolution imaging system is designed, implemented and characterized for the quantum degenerate apparatus.

Figure A.1: Schematic of designed configuration of the new imaging system. All lengths are in mm.

Figure A.1 shows the schematic of designed configuration of the imaging system mounted in a Thorlabs Cage Assembly System. The outer surface of the flange of the vacuum chamber is 10.8 cm away from the center of the chamber where atom clouds are located. A 1:1 optical relay system consists of a pair of achromatic doublet lens, and the focal length of each lens is 150 mm. The relay system forms an image of the atom cloud outside the chamber at point A. A microscope composed of an objective and a tube lens transfer the image to the CCD sensor of the camera. The
objective is removable and can be exchanged for different overall image magnification. Table A.1 lists optical elements shown in Fig. A.1 including two Nikon objectives with the amplification of $1 \times$ and $4 \times$, respectively. The parfocal distances of the objectives are $60 \text{ mm}$. The $1 \times$ objective is appropriate for diagnosing relatively large atom clouds in blue or red MOTs, while the $4 \times$ objective is for small-clouds imaging, especially quantum degenerate gases. Table A.2 lists other accessories not shown in Fig. A.1, such as cage plates, cage assembly rods, and various adapters.

### A.2 Characterization of Imaging System Off Table

Figure A.2 is the schematic of actual configuration of the imaging system where all elements are mounted in cage plates. Figure A.3 is the photograph of the imaging system on the optical table. Firstly, we characterize the imaging system off the optical table. A Standard 1951 USAF Slide Resolution Target (Fig. A.4) is illuminated with a collimated $461 \text{ nm}$ beam. Tables A.3 and A.4 list the number of line pairs per mm and the width of one line in the target, respectively.

![Schematic of actual configuration of the new imaging system](image)

Figure A.2: Schematic of actual configuration of the new imaging system. All lengths are in mm.
<table>
<thead>
<tr>
<th>Element</th>
<th>Vender</th>
<th>Stock #</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lens 1</td>
<td>Thorlabs</td>
<td>AC508-150-A</td>
<td>( f = 150 \text{ mm}, 2'' \text{ Unmounted Visible Achromat, ARC:} 400-700\text{ nm} )</td>
</tr>
<tr>
<td>Lens 2</td>
<td>Thorlabs</td>
<td>AC508-150-A</td>
<td>( f = 150 \text{ mm}, 2'' \text{ Unmounted Visible Achromat, ARC:} 400-700\text{ nm} )</td>
</tr>
<tr>
<td>Objective 1</td>
<td>Nikon</td>
<td>MRL00012</td>
<td>( 1 \times ) Nikon CFI Plan Achromat Objective, 0.04 NA, 3.2mm WD</td>
</tr>
<tr>
<td>Objective 2</td>
<td>Nikon</td>
<td>MRL00042</td>
<td>( 4 \times ) Nikon CFI Plan Achromat Objective, 0.1 NA, 30mm WD</td>
</tr>
<tr>
<td>Objective 3</td>
<td>Thorlabs</td>
<td>N10X-PF</td>
<td>( 10 \times ) Nikon Plan Fluorite Imaging Objective, 0.3 NA, 16mm WD</td>
</tr>
<tr>
<td>Tube Lens</td>
<td>Thorlabs</td>
<td>ITL200</td>
<td>Infinity-Corrected Tube Lens for Plan Fluorite Objectives</td>
</tr>
<tr>
<td>Camera</td>
<td>CooKe</td>
<td>Pixelfly</td>
<td>High Performance Digital CCD Camera System</td>
</tr>
<tr>
<td>Stock #</td>
<td>Description</td>
<td>Qty</td>
<td></td>
</tr>
<tr>
<td>---------</td>
<td>-----------------------------------------------------------------------------</td>
<td>-----</td>
<td></td>
</tr>
<tr>
<td>LCP01T</td>
<td>60 mm Threaded Cage Plate, 0.9&quot; Thick</td>
<td>5</td>
<td></td>
</tr>
<tr>
<td>ER24</td>
<td>Cage Assembly Rod, 24&quot; Long, φ6 mm</td>
<td>4</td>
<td></td>
</tr>
<tr>
<td>SM1A12</td>
<td>Adapter with External SM1 Threads and Internal M25 × 0.75 Threads</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>SM2A6</td>
<td>Adapter with External SM2 Threads and Internal SM1 Threads</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>SM2A20</td>
<td>Adapter with External SM2 Threads and Internal M38 × 0.5 Threads</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>SM2T2</td>
<td>SM2 (φ2.035&quot;-40) Coupler, External Threads</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>SM1A2</td>
<td>Adapter with External SM1 Threads and Internal SM2 Threads</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>SM1A9</td>
<td>Adapter with External C-Mount Threads and Internal SM1 Threads</td>
<td>1</td>
<td></td>
</tr>
</tbody>
</table>
A common parameter of image quality is contrast defined as

\[ C = \frac{C_{\text{max}} - C_{\text{min}}}{C_{\text{max}} + C_{\text{min}}}, \]  

(A.1)

where \( C_{\text{max}} \) and \( C_{\text{min}} \) are the maximum and minimum signal level, respectively.
<table>
<thead>
<tr>
<th>GROUP NUMBER</th>
<th>ELEMENT</th>
<th>0</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.00</td>
<td>2.00</td>
<td>4.00</td>
<td>8.00</td>
<td>16.00</td>
<td>32.0</td>
<td>64.0</td>
<td>128.0</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>1.12</td>
<td>2.24</td>
<td>4.49</td>
<td>8.98</td>
<td>17.95</td>
<td>36.0</td>
<td>71.8</td>
<td>144.0</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>1.26</td>
<td>2.52</td>
<td>5.04</td>
<td>10.10</td>
<td>20.16</td>
<td>40.3</td>
<td>80.6</td>
<td>161.0</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>1.41</td>
<td>2.83</td>
<td>5.66</td>
<td>11.30</td>
<td>22.62</td>
<td>45.3</td>
<td>90.5</td>
<td>181.0</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>1.59</td>
<td>3.17</td>
<td>6.35</td>
<td>12.70</td>
<td>25.39</td>
<td>50.8</td>
<td>102.0</td>
<td>203.0</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>1.78</td>
<td>3.56</td>
<td>7.13</td>
<td>14.30</td>
<td>28.50</td>
<td>57.0</td>
<td>114.0</td>
<td>228.0</td>
<td></td>
</tr>
</tbody>
</table>

Table A.3: Number of Line Pairs per mm in USAF Resolving Power Test Target 1951
<table>
<thead>
<tr>
<th>ELEMENT</th>
<th>0</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>500.00</td>
<td>250.00</td>
<td>125.00</td>
<td>62.50</td>
<td>31.25</td>
<td>15.63</td>
<td>7.81</td>
<td>3.91</td>
</tr>
<tr>
<td>2</td>
<td>446.43</td>
<td>223.21</td>
<td>111.36</td>
<td>55.68</td>
<td>27.86</td>
<td>13.89</td>
<td>6.96</td>
<td>3.47</td>
</tr>
<tr>
<td>3</td>
<td>396.83</td>
<td>198.41</td>
<td>99.21</td>
<td>49.50</td>
<td>24.80</td>
<td>12.41</td>
<td>6.20</td>
<td>3.11</td>
</tr>
<tr>
<td>4</td>
<td>354.61</td>
<td>176.68</td>
<td>88.34</td>
<td>44.25</td>
<td>22.10</td>
<td>11.04</td>
<td>5.52</td>
<td>2.76</td>
</tr>
<tr>
<td>5</td>
<td>314.47</td>
<td>157.73</td>
<td>78.74</td>
<td>39.37</td>
<td>19.69</td>
<td>9.84</td>
<td>4.90</td>
<td>2.46</td>
</tr>
<tr>
<td>6</td>
<td>280.90</td>
<td>140.45</td>
<td>70.13</td>
<td>34.97</td>
<td>17.54</td>
<td>8.77</td>
<td>4.39</td>
<td>2.19</td>
</tr>
</tbody>
</table>
Figure A.5: In-focus 1951 USAF slide resolution target illuminated with 461 nm laser light and imaged with the $1 \times$-objective imaging system onto a Pixelfly CCD.
Figure A.6: Setup of the microscope composed of the objective (bottom), the tube lens (middle), and the CCD sensor, i.e. image plane (top) for best performance recommended by datasheet.

A.2.1 Data Recorded by Imaging Systems with \( 4 \times \) Objective

We start with the system with the \( 4 \times \) objective. Figure A.6 shows the recommended setup of the microscope composed of the objective, the tube lens, and the CCD sensor (image plane). At first, the distance between the objective and the tube lens is preset at roughly 100 mm, which is within the recommended value of 70-170 mm. Next, since the recommended distance between the tube lens and the CCD sensor is 148 mm, and the CCD sensor is 17.5 mm away from the front surface of the camera (Fig. A.2), the distance between the tube lens and the front surface of the camera is preset at \( 148 - 17.5 = 130.5 \) mm. To get images in focus, the positions of the objective and the tube lens are walked. Coarse adjustment is performed by moving the cage plate where the objective/tube lens is mounted, while fine adjustment is realized by
screwing in/out the adaptor carrying the objective/tube lens. The in-focus position parameters of all optical elements are indicated in Fig. A.2.

Figure A.7: Slices of imaged 1951 USAF target taken by the imaging system with the 4× objective when images are in focus. Subtitles indicate widths of target lines.

Figure A.7 shows in-focus slices of imaged target lines. The pixel size is 6.08 μm, which at 4x magnification corresponds to 6.08 μm of object size per pixel (section A.3.1). It is noted that lines with the width of up to 6 μm can be resolved. Corresponding contrasts of images versus line widths are shown in Fig. A.8 (blue line). The contrast drops sharply at about 6 μm, which indicates the rough limit of the resolution of the system.

Since the two objectives will be switched back and forth during experiments, to check the reproducibility, the 4× objective is removed and remounted to the orig-
Figure A.8: Comparison of contrasts of images by remounting the 4× objective.

Figure A.8: Comparison of contrasts of images by remounting the 4× objective.

original in-focus position marked by a SM2RR retaining ring, and then contrasts are measured. Figure A.8 compares the data of the original in-focus position and those after remounting the objective once and twice, which proves that the system can be recovered well.

It is necessary to study the sensitivity of the imaging system due to the position of the objective. The internal SM2 thread of the cage plates has 40 TPI (threads per inch), which means that its pitch is 0.025 inches (0.635 mm). Here we define the original in-focus position of the objective as 0 cm, the direction pointing to the camera as positive (+), and that pointing to the target as negative (-). Images are recorded by systems with the 4× objective located at ±0.318 mm (screwing in/out the objective by a half turns) and ±0.159 mm (a quarter turns), respectively. Figure A.9 compares
contrasts in all five cases. The 0 mm curve is selected as the optimal in-focus position because its contrast stays high (larger than 0.8) for the smallest line width and is the highest in the range of line widths smaller than 7 \( \mu \text{m} \). In the range of line widths larger than 8 \( \mu \text{m} \), the contrast falls off faster to the negative side than to the positive side. The line widths at which the contrast decreases rapidly in positive curves are about 8 \( \mu \text{m} \), which are larger than those of around 6 \( \mu \text{m} \) in negative curves. However, the contrasts of all four positive and negative curves at around 6 \( \mu \text{m} \) are very close. To sum up, the effect of 0.159 mm shift (a quarter turns) is not negligible. When optimizing the objective position, a decent step size is screwing the objective by about 30 degrees.

![Graph showing contrast comparison]

Figure A.9: Comparison of contrasts of images by shifting the 4\( \times \) objective. Refer to the text for details.

All above data are recorded in the middle region (vertical pixel range: 412 - 736) of
the CCD sensor of the camera. To characterize the disparity of properties of different regions of the CCD sensor with the $4\times$ objective, images are recorded in the top (vertical pixel range: 4 - 244), middle (vertical pixel range: 412 - 736), and the bottom regions (vertical pixel range: 944 - 1260) of the CCD sensor, respectively. Figure A.10 lists these three groups of contrasts. There are no significant differences at line widths more than $8\,\mu m$, but the bottom data shows worse performance than other two at the range smaller than $8\,\mu m$.

Figure A.10 : Comparison of contrasts of images recorded in the top (vertical pixel range: 4 - 244), middle (vertical pixel range: 412 - 736), and bottom parts (vertical pixel range: 944 - 1260) of the CCD. The $4\times$ objective is used here.

A.2.2 Data Recorded by Imaging Systems with $1\times$ Objective

Now we switch to the $1\times$ objective which is mounted at the original in-focus position of the $4\times$ Objective with the aid of SM2RR retaining ring. Theoretically, the same
Parfocal lengths of the two objectives can guarantee the in-focus condition of the system with the 1× objective. Actually, fine adjustment of the position of the objective is needed when switching between two objectives with the same parfocal lengths. At the original in-focus position of the 4×-objective imaging system, the system with the 1× objective can only resolve lines with the widths of up to 25 μm, although it is sufficient for diagnosing large atom clouds in MOTs. However, we can get images in focus by moving the 1× objective by +1.590 mm (two and half turns). Figure A.11 shows the data recorded by imaging systems with the two objectives at individual in-focus positions, and we can notice the 4×-objective system performs better than the 1× one.

![Graph](image)

Figure A.11 : Comparison of contrasts of images recorded by imaging systems with the 4× and 1× objective, respectively. The 1× objective is shifted by +1.590 mm (2.5 turns) from the original 4×-objective-in-focus position.
A.3 Characterization of Imaging System on Table

The imaging system is mounted back to the optical table and is characterized with the aid of ultracold atoms.

A.3.1 Calibration of Pixel Sizes

Firstly, the pixel sizes of the camera systems with 1× and 4× objectives are calibrated via time-of-flight measurements, respectively. Figure A.12 shows the relative vertical position of atom clouds with a series of drop times, as well as the fitting lines using freely falling formula. The pixel sizes are determined to be 1.52(1) μm/pixel for 4×-objective imaging system, and 6.08(8) μm/pixel for 1× one. As expected, the latter is exactly four times the former.

A.3.2 Characterization of Resolution of Imaging Systems

Firstly, a very tiny $^{88}$Sr thermal-atom cloud is prepared and dropped for the time $t$. We assume a Gaussian density profile of an atom cloud

$$n(r) = n_0 \exp \left[ -\frac{r^2}{2\sigma^2} \right]$$

(A.2)

where $n_0$ is the peak density, and the width after expansion $\sigma$ can be described by [87]

$$\sigma^2 = \sigma_0^2 + (vt)^2,$$

(A.3)

with the original size of the atom cloud before expansion $\sigma_0$, and the speed of thermal atoms $v$.

Now the resolution of the imaging system $\sigma_{res}$ will be taken into account. The
Figure A.12: Calibration of pixel sizes of imaging systems with (a) 1× or (b) 4× objective via time-of-flight measurements. Pixel sizes are extracted by fitting data using freely falling formula.
point spread function (PSF) of the imaging system is given by

\[
L(r) = \frac{1}{2\pi\sigma_{\text{res}}^2} \exp \left[ - \frac{r^2}{2\sigma_{\text{res}}^2} \right],
\]

(A.4)

which is also Gaussian.

Thus, the observed density profile can be received by convolution

\[
n_f(r) = n(r) \otimes L(r) = \frac{n_0\sigma^2}{\sigma^2 + \sigma_{\text{res}}^2} \exp \left[ - \frac{r^2}{2(\sigma^2 + \sigma_{\text{res}}^2)} \right] = n_0 f \exp \left[ - \frac{r^2}{2\sigma_{\text{obs}}^2} \right]
\]

(A.5)

with the observed size of the atom cloud after expansion

\[
\sigma_{\text{obs}}^2 = \sigma^2 + \sigma_{\text{res}}^2.
\]

(A.6)

Substituting Eq. A.3 into Eq. A.6 yields

\[
\sigma_{\text{obs}}^2 = \sigma_0^2 + (vt)^2 + \sigma_{\text{res}}^2.
\]

(A.7)

When the drop time \( t \) is long enough (e.g. 1-4 ms) so that \( vt \gg \sigma_0, \sigma_{\text{res}} \), we have \( \sigma_{\text{obs}} \simeq vt \). The temperature of thermal atoms \( T \) can be extracted according to

\[
\frac{1}{2}k_B T = \frac{1}{2}mv^2 \simeq \frac{1}{2}m \left( \frac{\sigma_{\text{obs}}}{t} \right)^2,
\]

(A.8)

with the Boltzmann constant \( k_B \) and the mass of atoms \( m \).

With the parabolic-trap approximation, the original size of the thermal could before expansion \( \sigma_0 \) can be determined from

\[
\frac{1}{2}k_B T = \frac{1}{2}m(\omega\sigma_0)^2,
\]

(A.9)

where the oscillator frequency of the ODT \( \omega = 2\pi f \) with trap frequencies \( f \), which can be predicted by the trap model at the final power of the input ODT beam before the vacuum chamber of 8.2 W (Table A.5).
Combining Eqs. A.8 and A.9 yields

$$
\sigma_0 = \frac{\sigma_{\text{obs}}}{2\pi ft},
$$

(A.10)

Figure A.13: Observed sizes of atom clouds $\sigma_{\text{obs}}$ in horizontal (Left) and vertical (Right) axes after a series of long drop times. $\sigma_{\text{obs}}/t$ is extracted by linear fit of data.

Figure A.13 shows the time-of-flight data with a series of drop times, from which $\sigma_{\text{obs}}/t$ is determined to be 7.7 $\mu$m/ms in both vertical (Y) and horizontal (X) axes. Thus, the original size of the atom cloud $\sigma_0$ can be calculated according to Eq. B.2 (Table A.5). Figure A.14 shows an example of the data with an expansion time of 2 ms, which can be well fitted using a gaussian distribution.

Then we measure the observed size of thermal cloud with extremely short expansion time of 100 $\mu$s $\sigma'_{\text{obs}}(X) = 6.1 \mu$m and $\sigma'_{\text{obs}}(Y) = 5.5 \mu$m (Fig. A.15), so that $vt \ll \sigma_0, \sigma_{\text{res}}$. Therefore, eq. A.7 can be rewritten as

$$
\sigma'^2_{\text{obs}} = \sigma'^2_0 + \sigma'^2_{\text{res}},
$$

(A.11)
from which the resolution of the imaging system can be extracted by

$$\sigma_{\text{res}} = \sqrt{\sigma_{\text{obs}}^2 - \sigma_0^2}. \quad (A.12)$$

Table A.5 lists measured parameters for calculation of the resolution, which is determined to be $4.8(2) \, \mu m$ and $4.8(1) \, \mu m$ in the horizontal and vertical directions,
respectively, which is improved a little bit compared to that of the old imaging system of about 5 to 6 \( \mu m \).

<table>
<thead>
<tr>
<th>Axes</th>
<th>( f ) [Hz]</th>
<th>( \sigma_0 [\mu m] )</th>
<th>( \sigma'_{\text{obs}} [\mu m] )</th>
<th>( \sigma_{\text{res}} [\mu m] )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Horizontal (X)</td>
<td>325(25)</td>
<td>3.8(3)</td>
<td>6.1</td>
<td>4.8(2)</td>
</tr>
<tr>
<td>Vertical (Y)</td>
<td>465(15)</td>
<td>2.6(1)</td>
<td>5.5</td>
<td>4.8(1)</td>
</tr>
</tbody>
</table>

The diffraction limit \( d_s \) of the 1:1 relay system is given by \[140\]

\[
d_s = 2.44\lambda M f_0 / D \simeq 3.3 \mu m,
\]

where the wavelength of the imaging laser \( \lambda = 461 \text{nm} \), the magnification of the relay system \( M = 1 \), the focal length of the two achromatic doublet lens \( f_0 = 150 \text{mm} \), and the diameter of the lens \( D = 2 \text{inches} \).

It is noticed that the resolution of the imaging system of 4.8 \( \mu m \) is a little larger than the diffraction limit of 3.3 \( \mu m \). It could be from the relay system. For example, a little deviation in the distance between atom clouds and Lens 1 may result in the deviation of the magnification from 1. It also could be attributed to the microscope part. The objective may not be exactly at the best in-focus position due to the extremely sensitive objective-position dependence of the system. Furthermore, Ref. [141] points out that the typical tolerances of optical elements’ displacement transverse to the optical axis is on the order of tens of \( \mu m \), and the possible displacement in our imaging system could degrade performance.
A.3.3 Imaging $^{88}$Sr BECs

At last, $^{88}$Sr BECs are prepared and dropped by 20 ms, and are recorded by imaging systems with $1 \times$ and $4 \times$ objectives, respectively. Figure A.16 shows the 2D absorption images and 1D profiles of BECs in both cases. As expected, BEC component occupies more pixels in images recorded by the $4 \times$-objective imaging system, which improves the property of the imaging system significantly when imaging tiny atom clouds.

![1x Objective](image1.png) ![4x Objective](image2.png)

Figure A.16: 2D absorption images (Top) and areal density profile (Bottom) of $^{88}$Sr BECs. Data corresponding to 20 ms of free expansion are recorded by imaging systems with $1 \times$ objective (Left) and $4 \times$ objective (Right), respectively.
Appendix B

Background Fitting Routine

B.1 Method

During experiments, an intensity pattern with atom clouds present $I_{\text{atoms}}$, and a background pattern without atom clouds $I_{\text{back}}$ are recorded in each measurement cycle. Figure B.1 is an example of $I_{\text{atoms}}$ with $^{88}$Sr BECs and background pattern $I_{\text{back}}$, both of which are squares with sides of $D$. We are interested in the region $I_{\text{atoms}}^{\text{in}}$ (Fig. B.1(a)) inside the dashed square with sides of $d$ where atoms are located, as well as the region $I_{\text{back}}^{\text{in}}$ (Fig. B.1(b)) inside the dashed square, which has the same dimension and position as $I_{\text{atoms}}^{\text{in}}$. The remainder region outside the dashed square is denoted as $I_{\text{atoms}}^{\text{out}}$ and $I_{\text{back}}^{\text{out}}$, respectively. In the old routine, 2D absorption images are generated from $\ln(I_{\text{atoms}}^{\text{in}}/I_{\text{back}}^{\text{in}})$. The drawback of the routine is that different random noise added to $I_{\text{atoms}}$ and $I_{\text{back}}$ which makes images fuzzy, especially when imaging tiny atom clouds in quantum degenerate with the small number of atoms.

To reduce vagueness due to random noise, an alternative routine of processing background signals is applied, which directly takes into account the random noise from $I_{\text{atoms}}$ itself.

In the new background fitting routine, instead of $I_{\text{back}}^{\text{in}}$, the effective background pattern $I_{\text{back}}^{\text{in, eff}}$ is introduced, which is obtained with the aid of $N$ background patterns $I_{\text{back}}^{(j)}$ ($j = 1, ..., N$). Firstly, the outside region of the pattern with atoms $I_{\text{atoms}}^{\text{out}}$ is
Figure B.1: Squared intensity patterns $I_{\text{atom}}$ with atom clouds (a), and $I_{\text{back}}$ without atom clouds (b). Regions inside the dashed squares are denoted as $I_{\text{in}}$, while remainder regions, which are outside the dashed squares, are denoted as $I_{\text{out}}$. 
considered as a linear combination of a series of $I_{\text{back}}^{\text{out}(j)}$ ($j = 1, ..., N$), and can be fitted using
\begin{equation}
\sum_{j=1}^{N} P_j I_{\text{back}}^{\text{out}(j)},
\end{equation}
where $P_j$ are undetermined coefficients, which is the weight of each $I_{\text{back}}^{\text{out}(j)}$ contributing to $I_{\text{atoms}}^{\text{out}}$.

It is an intuitive approximation that each $I_{\text{back}}^{\text{in}(j)}$ contributes to $I_{\text{atoms}}^{\text{in}}$ with the same weight $P_j$. Thus, the effective background pattern $I_{\text{back}}^{\text{in,eff}}$ can be expressed as a linear combination of $I_{\text{back}}^{\text{in}(j)}$ with the weight of $P_j$ determined by the fitting
\begin{equation}
I_{\text{back}}^{\text{in,eff}} = \sum_{j=1}^{N} P_j I_{\text{back}}^{\text{in}(j)},
\end{equation}

Figure B.2 shows 2D absorption images and areal profiles of $^{88}$Sr BECs using the old and new background fitting routines, respectively. There are $N = 11$ background $I_{\text{back}}^{\text{in}(j)}$ used in the new routine. It is clear that the profile processed by new routine is cleaner than old one, especially in thermal parts (wings) of the profiles. To balance the effect and time cost in fitting programs, usually $N$ is selected between 10 to 20, and the dimension $D$ is 3 to 4 times larger than $d$. 
Figure B.2: Comparison of 2D absorption images (Top) and areal profile (Bottom) of $^{88}$Sr BECs using the old (a) and new routines (b), respectively. There are $N = 11$ background $I_{\text{back}}^{\text{in}(j)}$ used in the new routine.
B.2 Codes

The corresponding Matlab codes are attached here, which were written by Mi Yan, and modified by Jim Aman. There are 4 Matlab files allowing someone to run the background program:

(1) AnalysisVariables.m is the settings file;

(2) get_indiv_batch_data.m reads batch specific information and passes to fitting functions;

(3) imagefit_gaussianBimodalAndHistogramFittingA.m does the least square difference calculation in parallel;

(4) WeightedBackgroundFunction.m computes the current linear decomposition of the background basis set and passes back the squared difference.
function analyVar = AnalysisVariables()
% Master file to control all similar variables to be passed between
% background analysis, cloud fitting, and graphing routines.
% INPUTS:
% none
% OUTPUTS:
% analyVar - Structure containing all the variables defined in
% this function.
% MISC:
% %ok<*NASGU> - suppress all instances of 'this variables may not be used'
% because the who() builds a structure with all variables
% defined in the workspace.
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
%% Initialization and Options
%2012.12.04
BECweight    = 10^6;
cloudWindowRadius = 35;
roiWindowRadius   = 150; % Usually, roiWindowRadius = cloudWindowRadius*2.5

% Fitting options
BimodalFitPara  = 4; % Parameter for Bimodal fitting, usually it is 4. However, for some data, if the fitting is bad, you can adjust this value.
PureBECBegin    = 0; % The value n here means pure BECs emerge from the nth data point; 0 means there is no pure BEC in the scan
Plotdetail      = 1; % 1 plot details, 0 not plot details
fit             = 1; % 1 to fit condensate or simply plot;
Fugacityfit     = 1; % 1 will fit the thermal part with Fugacityfit and 0 with Gaussian
BECfit          = 0; % 1 will use TF function to fit BEC...only on first pass...not on residuals
BECsizeDisplay  = 1; % 1 means BEC sizes displayed are ONE sigma WITH resolution correction; 2 means they are TWO sigma WITHOUT resolution correction
BECfuncFit      = 0; % 1 means Fit BEC part to the BEC function; 0 means Fit BEC part to the Gaussian function

% Variables defined in lines of each dataset in the master batch file.
masterBatchAtomVar = {'basenamevectorAtom' 'timevectorAtom' 'OFRdetuneAtom' 'dummy02Atom' 'droptimeAtom' 'roicol1Atom' 'roicol2Atom' 'isotopeAtom'};
masterBatchBackVar = {'basenamevectorBack' 'timevectorBack' 'OFRdetuneBack' 'dummy02Back' 'droptimeBack' 'roicol1Back' 'roicol2Back' 'isotopeBack'};

% Variables defined in lines of the individual batch files
indivBatchAtomVar = {'fileAtom' 'imagevcoAtom' 'vcovoltageAtom' 'motdetuningAtom' 'ODTVoltageAtom' 'fugacityguessAtom' 'BECamplitudeParameterAtom' 'sigParamAtom' 'sigBECParamAtom' 'WeightedBECPeakAtom' 'sampleholdAtom' 'wavemeterAtom'};
indivBatchBackVar = {'fileBack' 'imagevcoBack' 'vcovoltageBack' 'motdetuningBack' 'ODTVoltageBack' 'fugacityguessBack' 'BECamplitudeParameterBack' 'sigParamBack' 'sigBECParamBack' 'WeightedBECPeakBack' 'sampleholdBack' 'wavemeterBack'};

% Output Parameter filenames
BackFitParamsFilename = 'BackgroundfitparamsA.txt';
ODimageFilename = 'ODimagebatch.txt';
cutImageAtomFilename = 'cutImageAtom.txt';
cutImageBackFilename = 'cutImageBack.txt';
BECfitFilename = 'Backgroundfitparams';
BECfitGaussFilename = 'Gauss';
BECfitFileExt = '.txt';
BECNumCombineFilename = 'BEC-Holdtimes-Mean_Number-RMS_error.txt';

% PHYSICAL QUANTITIES
mass = 88*1.672*10^(-27); % strontium mass
kBoltz = 1.38*10^(-23);
templow = 1*10^(-6);
hbar = 1.05*10^(-34);
lambda = 461*10^(-9);
detuning = 0; % image beam detuning in Hz (as of 3/1/13)
timefactor = 10^3; % scales time from ms to s
BohrRadius = 0.53*10^(-10); % Bohr Radius in meter
NaturalWidth = 32*10^6; % ion fwhm in Hz
CrossSection = 6*pi*(lambda/(2*pi))^2;
AbsCross = CrossSection*1/(1 + (2*detuning/NaturalWidth)^2); %%% Pascal's Doctorate
thesis (Eq. B.2)

%All frequencies here ONLY go into alldata(k,1); frequencies.
dzeeman = -251.43; % constant overnight
dPAS = -250; % constant overnight
dshift = -41; % constant overnight
databs = 0; % 0 1st run, 260.5 for 2nd run
catseyesign = 0; % depends on peak
ODTPowerFactor = 1.01; %ODTpower = 1.01 * ODT Voltage, measure on 09/07/2010, see Lab Notebook for detail

% SPECIFICS OF FITTING AND PLOTTING ROUTINES
textsize = 16;
axisfontsize = 14;
SmoothPlotSize = 2; %number of points to average for plotting
guessSmoothSize = 1; %This variable is used in fitting, and also smoothes out the linear density cuts

plotFits = Plotdetail; % 1 to plot fits/data, 0 to not plot fits; 2D Gaussian
plotEvolution = Plotdetail; % 1 to show surface plots for each data point on one plot
plotDensity = Plotdetail; % 1 to show plots of density as function of x position (slices along y); helps to spot BEC, fig 2000,3000
fallingAtom = 0; % 1 if cloud position changes from image to image
centFiltWght = 50; % factor to decide smoothness of applied filter when identifying cloud center (findCenterRawImage)
PASResonance = 0; % in MHz (previous value 114.1)
CameraRes = 5; % in micron
SavePlotData = 0; % 1 to allow aggregation of variables from plotting into output structure, 0 to disable output
plotRawImage = 0; % 1 to plot processed raw images (trimmed and binned)
becweight = 1; % 1 will set small weights to a region selected with HalfBEC, xoffset, etc
binaryread = 1; % 0 if the files are ASCII; 1 if the files are binary
AppearBEC = 0; % 1 will plot the appearance of BEC including 4 steps during evaporation
Bimodal = 1; % 1 will plot one bimodal figure for specified data
Bimodalk = 3; % Specify which data, i.e. the sequence number, will be plotted in bimodal figure
save2places = 0; % 1 means we save output files to both primary and secondary directories for backup purposes
TimeOrDetune = 0; % 1 means in the scan time is changing, 0 means Detuning (i.e. DAC to VCO) is changing which is used in BEC size measurement
sliceAverage = 3; % For plotting the 1-D fits through cloud, if sliceaverage=1 there is no averaging,... % if sliceaverage=3 the plot is the average of three pixels perpendicular to slice.

plotResidualLineDensity = 1; % 1 to show line cuts of residual density and fit...must have fitresiduals=1
fitResiduals = 1;
centerFindBinLimit = 5; % Limit to determine whether further binning is required to find the cloud center
centerFindBinSize = 4; % Bin size for finding the position of the cloud
centerFor1DSlice = 0; % For plotting the 1-D fits through cloud, 1 means take center through which the slice... % goes as your center guesses, 0 means take it as the center of the gaussian fit.

COLORS = [0 0 0; 1 0 0; 0 0 1; 0 0 1; .25 .25 .25; 0 .75 0; 0 .75 .75; .5 0 .5; .75 0 .75; 0 1 0; 0 1 0; 0 .75 0; 0 .75 .75; .5 0 .5; .75 0 .75; 0 0 1; 0 1 0; .25 0 .75; .5 0 .5; 0 1 0; 0 .75 0; 0 .75 .75; .5 0 .5; .75 0 .75; 0 0 1; 0 1 0; .25 0 .75; .5 0 .5; 0 1 0; 0 .75 0; 0 .75 .75; .5 0 .5; .75 0 .75; 0 0 1; 0 1 0; .25 0 .75; .5 0 .5; 0 1 0; 0 .75 0; 0 .75 .75; .5 0 .5; .75 0 .75; 0 0 1; 0 1 0; .25 0 .75; .5 0 .5; 0 1 0; 0 .75 0; 0 .75 .75; .5 0 .5; .75 0 .75; 0 0 1; 0 1 0; .25 0 .75; .5 0 .5; 0 1 0; 0 .75 0; 0 .75 .75; .5 0 .5; .75 0 .75; 0 0 1; 0 1 0; .25 0 .75; .5 0 .5; 0 1 0; 0 .75 0; 0 .75 .75; .5 0 .5; .75 0 .75; 0 0 1; 0 1 0; .25 0 .75; .5 0 .5; 0 1 0; 0 .75 0; 0 .75 .75; .5 0 .5; .75 0 .75; 0 0 1; 0 1 0; .25 0 .75; .5 0 .5; 0 1 0; 0 .75 0; 0 .75 .75; .5 0 .5; .75 0 .75; 0 0 1; 0 1 0; .25 0 .75; .5 0 .5; 0 1 0; 0 .75 0; 0 .75 .75; .5 0 .5; .75 0 .75; 0 0 1];
MARKERS = {'-o','-s','-^','-d','-v','-'p','-'<','-'h','->','-x','-+','-.','-o','-s','-^','-d','-v','-'p','-'<','-'h','->','-x','-+','-.','-o','-s','-^','-d','-v','-'p','-'<','-'h','->','-x','-+','-.','-o','-s','-^','-d','-v','-'p','-'<','-'h','->','-x','-+','-.','-o','-s','-^','-d','-v','-'p','-'<','-'h','->','-x','-+','-.','-o','-s','-^','-d','-v','-'p','-'<','-'h','->','-x','-+','-.','-o','-s','-^','-d','-v','-'p','-'<','-'h','->','-x','-+','-.','-o','-s','-^','-d','-v'};

%% Image Processing Options
%----------------------------------------------------------------------%
%PIXEL SIZE CHANGED FROM 10.4e-6 TO 9.2e-6 ON October 12, 2009 FOR BEC DATA
%ANALYSIS FROM DATASET WITH TIMESTAMP 2539 ONWARD (FILES FOUND IN ODT\2009.10.10 DATA FOLDER)
%SEE PG. 19YELLOW OF NEUTRAL NOTEBOOK H FOR MORE DETAILS
pixelsize = 0.0000105;
pixelconv = 10.5; %converts pixels to microns %.05*10^-1;
%------------------------------------------------------------------------%
softwareBinSize = 1; %converting pixels to microns %1.05*10^-1;
softwareBinSize*=softwareBinSize
CCDbinning = 1; %Number of pixels square to bin (i.e. area of bin, in # of pixels = number of pixels binned when first recording data
cutBorders = 0; %number of column and rows to trim on either side of the data matrix; same as roi parameters, 'cept inverse way of doing it.
NoiseNumVec = 5; %number of column and rows at the corners to find the uncertainties/noise
scaleBaseLevel = 0; % 1 to scale the base level of atom and background image, 0 for not numberForScale = 5; % number of column and rows to scale the base level of atom and background images
matrixsize = [1280 1024]; %matrix size for binary reading.
sizfactor = pixelsize*softwareBinSize*CCDbinning; % effective pixelsize
NeutExpDir = 'Neutral_Experiment';
analyPrefix = 'BECfit_';
analyOutputName = 'Analysis_Output';

% Two assumptions are made here,
% (1) - The batch directory is at the same folder level as the dataDirectory
% (2) - The batchhead files output by Labview are label as Files_yyyymmdd
% (Files_yyyymmdd_Bg for background)
% First need to determine file structure of Analysis folder
analyDir = pwd; % save Analysis Folder location
% Determine directory where all raw data files are saved (expected to mirror folder
% structure of Analysis folder
dataDir  = [strrep(strrep(analyDir,analyPrefix,''),[filesep 'Analysis' filesep],[filesep 'Raw_Data' filesep]) filesep];
dataDirName = regexp(dataDir,filesep,'split'); dataDirName = regexp(dataDirName{end - 1},'_','split'); dataDirName = dataDirName{1};
% Check if data directory exists
if not(exist(dataDir,'dir'))
    error('
Data Directory: %s
not found. Please check analysis directory and try again.\n',dataDir)
end
% Check if analysis output directory exists, if not create
analyOutDir = [pwd filesep analyOutputName '_' dataDirName filesep]; % Output directory for analysis
if not(exist(analyOutDir,'dir'))
    mkdir([pwd filesep analyOutputName '_' dataDirName])
end
% NOTE: Win 7 has a filename limitation of 260 characters so if you get weird errors with dlmwrite look at the path name length

% Master batch file containing all datasets for the day
% If combining datasets to plot across multiple days create a new master batch file that shares the name of the folder without periods (i.e. \Data\...2012.12.14\_15_Combine\Files_2012121415_Combine.txt )
basenamelistAtom = [dataDir 'Files_' strep(dataDirName,'.',',','') '.txt']
basenamelistBack = [dataDir 'Files_' strep(dataDirName,'.',',','') '_Bg' '.txt']

% Read in variables for each data set defined in the master batch file
masterBatchAtomData = textscan(fopen(basenamelistAtom),'%s%f%f%f%f%f%f%f%f','%'); %data set batch files for atoms
masterBatchBackData = textscan(fopen(basenamelistBack),'%s%f%f%f%f%f%f%f%f','%'); %data set batch files for backgrounds

% Should there be a warning if the size is > 10?
numberOfbasenamesAtom = size(masterBatchAtomData{1},1); % max of ten for figure 200 to work
numberOfbasenamesBack = size(masterBatchBackData{1},1); % max of ten for figure 200 to work
%% What kind of files are expected?
if binaryread == 1
    dataAtom = char('atoms.bny');
dataBack = char('back.bny');
elseif binaryread == 0
    dataAtom = char('atoms.dat');
dataBack = char('back.dat');
end

%% CALCULATED VARIABLES
% Create grid of the ROI to find number of elements in cloud and around cloud
[cloudY,cloudX] = meshgrid(-roiWindowRadius:roiWindowRadius,-roiWindowRadius:roiWindowRadius);
roiNotCloud = numel(find(cloudX > cloudWindowRadius | cloudY > cloudWindowRadius));
% Finds the number of elements that will be outside cloudWindowRadius
roiCloud = numel(find(not((cloudX > cloudWindowRadius | cloudY > cloudWindowRadius))));
% Finds the number of elements within cloudWindowRadius

% From imagefit_GaussianBimodalAndHistogramV2012 options
OFRdetune = masterBatchAtomData{3};
DetuningList = zeros(1,(length(nonzeros(diff(OFRdetune))) + 1)); % Determine the number of time the detuning is changed
NumberOfEachDetuningFile = zeros(1,(length(nonzeros(diff(OFRdetune))) + 1)); % in order to initialize the vectors
for i = 1:length(OFRdetune)
    if i == 1
        NumberOfDetuningList = 1;
        OFRdetuneTemp = OFRdetune(1);
        DetuningList(NumberOfDetuningList) = OFRdetune(1);
        NumberOfEachDetuningFile(NumberOfDetuningList) = 1;
    else
        if OFRdetune(i) == OFRdetuneTemp
            NumberOfEachDetuningFile(NumberOfDetuningList) = NumberOfEachDetuningFile(NumberOfDetuningList) + 1;
        else
            OFRdetuneTemp = OFRdetune(i);
            NumberOfDetuningList = NumberOfDetuningList + 1;
            DetuningList(NumberOfDetuningList) = OFRdetune(i);
            NumberOfEachDetuningFile(NumberOfDetuningList) = 1;
        end
    end
end

DetuningListFit = [0.5 1 1.5 2 2.5 3 3.5 4 4.5 5 5.5 6 6.5 7 7.5 8];
BECnumFitBest = [5059.64 6196.85 6541.54 6689.29 6766.2 6811.43 6840.34 6859.96 ... 6873.91 6884.19 6891.99 6902.86 6906.74 6909.91 6912.54].^.*.95;
BECsizeFitBest = [3.6290105e+01 3.3365628e+01 3.1770458e+01 3.0972874e+01 3.0972874e+01 ... 2.8978981e+01 2.8845981e+01 2.8713050e+01]./2;

%% Create structure of variables
% First need to match the batch filename variables to their values
masterBatchVars = cell2struct(cat(2,masterBatchAtomData,masterBatchBackData),cat(2,
masterBatchAtomVar,masterBatchBackVar),2);

% Concatenate masterBatch structure with another containing all other variables. Lists fields alphabetically.
analyVar = catstruct(v2struct(cat(1,'fieldNames',who())),masterBatchVars,'sorted');

%% Cleanup Workspace
fclose all;
function indivDataset = get_indiv_batch_data(analyVar)
% Reads the master file list provided from AnalysisVariables (in analyVar) and
% opens each dataset batch file to create the dataset variables and the
% BackgroundAll matrix
%
% INPUTs:
%   analyVar - Structure from AnalysisVariables which enumerates all the
%              variables needed for the analysis
%
% OUTPUTs:
%   indivDataset - a structure containing the dataset variables and
%                  BackgroundAll matrix for each dataset.
%
% MISC:
%#ok<*PFBNS> - suppress all instances of 'this variables is indexed but not
%              alled'. Passing large arrays may incur unnecessary communication
% overhead to the workers but we are only reading from the
% analyVar and indivDataset structures and not changing them.
% If there is a performance issue look into the function
% WorkerObjWrapper
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
%% Loop through each batch file listed in analyVar.basenamevectorAtom
indivDataset = cell(analyVar.numberofbasenamesAtom,1);
for basenamenumber = 1:analyVar.numberofbasenamesAtom
    batchfileAtom = [analyVar.dataDir char(analyVar.basenamevectorAtom(basenamenumber)) '.
    batch']; % current atom batchfile name
    batchfileBack = [analyVar.dataDir char(analyVar.basenamevectorBack(basenamenumber)) '.
    batch']; % current background batchfile name

%read in all atom files, no limit
    indivBatchAtomData = textscan(fopen(batchfileAtom), '%q%f%f%f%f%f%f%f
s','commentstyle','%');
%read in all background files, no limit
    indivBatchBackData = textscan(fopen(batchfileBack), '%q%f%f%f%f%f%f%f
s','commentstyle','%');
% create structure with the variables from the filename
    indivBatch = cell2struct(cat(2,indivBatchAtomData,indivBatchBackData),cat(2,analyVar.
    indivBatchAtomVar,analyVar.indivBatchBackVar),2);

    indivBatch.CounterAtom = size(indivBatch.fileAtom,1); % determine how many atom files
    in batch associated with this basename
    indivBatch.CounterBack = size(indivBatch.fileBack,1); % determine how many background
    files in batch associated with this basename

% Subplot number for plotting program
    [indivBatch.SubPlotRows, indivBatch.SubPlotCols] = optiSubPlotNum(indivBatch.
    CounterAtom);

% Decide whether to look for center once or everytime
    if analyVar.fallingAtom == 1;
    diffCloudCenters = indivBatch.CounterAtom;
    else
    diffCloudCenters = 1;
    end

% Initialize roi boundaries
[roiRowStartVec, roiRowStopVec, roiColStartVec, roiColStopVec] = deal(zeros(1, diffCloudCenters));

% Aggregate background files into 2 matrices, 1 of background behind cloud and the other around the cloud
% Initialize matrices for speed
AtomsCloud = zeros(indivBatch.CounterAtom, analyVar.elCloud);
AtomsNotCloud = zeros(indivBatch.CounterAtom, analyVar.elNotCloud);
for k = 1:indivBatch.CounterAtom
    s = [analyVar.dataDir char(indivBatch.fileAtom(k)) analyVar.dataAtom]; sFID = fopen(s, 'rb', 'ieee-be');
    if analyVar.binaryread == 1;
        % Read in binary file created in LabView
        % LabView saves binary in "big endian" ('be') format: most significant bit in lowest memory address. Matlab needs this info to import binary file correctly.
        % LabView and Matlab treat matrix coordinates differently.
        % (0,0) for LabView is lower left corner; for Matlab is upper left corner.
        fullRawImageAtom = fread(sFID, analyVar.matrixsize, '*int16');
    elseif analyVar.binaryread == 0;
        fullRawImageAtom = transpose(dlmread(s));
    end; fclose(sFID);
    % Only determine center for number of atom images
    if k <= diffCloudCenters
        % Determine center of cloud from full image and save
        [roiRowStartVec(k), roiRowStopVec(k), roiColStartVec(k), roiColStopVec(k)] = ...
            findCenterRawImage(analyVar, indivBatch.fileBack(k), fullRawImageAtom);
        % Assign roi boundaries
        [roiRowStart, roiRowStop, roiColStart, roiColStop] = ...
            deal(roiRowStartVec(k), roiRowStopVec(k), roiColStartVec(k), roiColStopVec(k));
    end
    % Region of interest in Atoms defined by Cloud Center +/- ROI Window Radius
    roiRawImageAtom = ...
    fullRawImageAtom(roiRowStart:roiRowStop, roiColStart:roiColStop);  
    % Separate Atoms into cloud part and around cloud part
    AtomsNotCloud(k, :) = ...
        double(roiRawImageAtom(analyVar.cloudX > analyVar.cloudWindowRadius | analyVar.cloudY > analyVar.cloudWindowRadius)');
    AtomsCloud(k, :) = ...
        double(roiRawImageAtom(not(analyVar.cloudX > analyVar.cloudWindowRadius | analyVar.cloudY > analyVar.cloudWindowRadius))');
end

% Aggregate background files into 2 matrices, 1 of background behind cloud and the other around the cloud
% Initialize matrices for speed
BackgroundNotCloud = zeros(indivBatch.CounterBack, analyVar.elNotCloud, diffCloudCenters);
BackgroundCloud = zeros(indivBatch.CounterBack, analyVar.elCloud, diffCloudCenters);
for j = 1:indivBatch.CounterBack
    t = [analyVar.dataDir char(indivBatch.fileBack(j)) analyVar.dataBack]; tFID = fopen(t, 'rb', 'ieee-be');
if analyVar.binaryread == 1;
    fullRawImageBack = fread(tFID, analyVar.matrixsize, 'int16');
else if analyVar.binaryread == 0;
    fullRawImageBack = transpose(dlmread(t));
end; fclose(tFID);

for q = 1:diffCloudCenters
    % Assign roi boundaries
    [roiRowStart, roiRowStop, roiColStart, roiColStop] = ...
        deal(roiRowStartVec(q), roiRowStopVec(q), roiColStartVec(q), roiColStopVec(q));

    % Region of interest in Background defined by Cloud Center +/- ROI Window
    roiRawImageBack = ...
        fullRawImageBack(roiRowStart:roiRowStop, roiColStart:roiColStop);

    % Separate Background into part behind cloud and part around cloud
    BackgroundNotCloud(j,:,q)  = ...
        double(roiRawImageBack(analyVar.cloudX > analyVar.cloudWindowRadius | ...
            analyVar.cloudY > analyVar.cloudWindowRadius));

    BackgroundCloud(j,:,q) = ...
        double(roiRawImageBack(not(analyVar.cloudX > analyVar.cloudWindowRadius | ...
            analyVar.cloudY > analyVar.cloudWindowRadius));
end %%% end of j = 1:CounterBack

% Save Atoms and Background into the indivBatch structure
indivBatch.AtomsNotCloud      = AtomsNotCloud;
indivBatch.AtomsCloud         = AtomsCloud;
indivBatch.BackgroundNotCloud = BackgroundNotCloud;
indivBatch.BackgroundCloud    = BackgroundCloud;

%% Save the variables for each dataset into a variable containing data from all
% datasets listed in analyVar.basenamevectorAtom
indivDataset{basenamenumber} = orderfields(indivBatch);
end

%% Clean Workspace
fclose all;
function imagefit_GaussianBimodalAndHistogramFittingA(varargin)
%This program is designed to read several datafiles and corresponding background files, plots these normalized data sets on the same graph.
% INPUTS:
% varargin - variable input argument to allow passing of analysis variables from BEC_Analysis_Runner. If not passed, the program will call AnalysisData and get_indiv_batch_data itself. It is important to follow the input construction below for varargin to retrieve variable data from other programs. -- first argument - analyVar -- second argument - indivDataset
% OUTPUTS:
% none
%
% MISC:
%#ok<*PFBNS> - suppress all instances of 'this variables is indexed but not sliced'. Passing large arrays may incur unnecessary communication overhead to the workers but we are only reading from the analyVar and indivDataset structures and not changing them. If there is a performance issue look into the function WorkerObjWrapper
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
```
%% Workspace specific options
optimOptions = optimset('Display','off'); % Suppress warning of MaxFunEvals from fminsearch
warning('off','all') % Suppress parfor warnings in command window

%% Load variables and file data
if nargin == 0 % If run without arguments
    analyVar = AnalysisVariables;
    indivDataset = get_indiv_batch_data(analyVar);
else
    analyVar = varargin{1}; % if arguments are passed analyVar must be first
    indivDataset = varargin{2}; % indivDataset must be second
end

%% Initialize parallel workers
%Open matlabpool if none open
if matlabpool('size') == 0
    matlabpool open
end

%% Loop through each batch file listed in analyVar.basenamevectorAtom
for basenamenumber = 1:analyVar.numberofbasenamesAtom
    % this will keep track of all the files analyzed in all the batches
    fprintf('
Background fitting batch file %g of %g\n',basenamenumber,analyVar.basenamevectorAtom)

    % InitializeCondition = ones(1,indivDataset{basenamenumber}.CounterBack)/indivDataset{basenamenumber}.CounterBack; % vector for fminsearch
    parfor k = 1:indivDataset{basenamenumber}.CounterAtom %this loop fits the background for all atom files in this dataset
        % Check whether cloud center changes
```
if (analyVar.fallingAtom); q = k; else q = 1; end;

% Minimization procedure to find coefficients of the background images to fit the AtomNotCloud (background around cloud image)
A = fminsearch(@(A) WeightedBackgroundFunction(A, indivDataset{basenamenumber}.AtomsNotCloud(k),
    indivDataset{basenamenumber}.BackgroundNotCloud(:,:,q)),
    InitialCondition, optimOptions);

% Write output to disk
dlmwrite(fullfile(analyVar.analyOutDir, char(indivDataset{basenamenumber}.fileAtom(k)),'analyVar.BackFitParamsFilename'), A, '	');
end %loop through files in dataset (one batchfile)
end %end loop through batches

%% Cleanup Parallel workspace
warning('on','all') %% Reenable warnings

%% Wrap Up
fclose('all'); % Close any file handles which may be open
fprintf('The background fitting is completed.

')
function f = WeightedBackgroundFunction(A,AtomsExclusion,BackgroundAll)
% Weight background images by coefficients in A then compare the squared
% difference. Fminsearch will attempt to minimize the squared difference by
% changing coefficients in A.
% In linear algebra terms, we're attempting to decompose each atom
% background into a linear combination of the background image basis set.
%
% INPUTS
%   A              - Coefficients of the background basis set
%   AtomsExclusion - Background image of atoms (minus the cloud)
%   BackgroundAll  - All background images of dataset which define the basis
%                    set we'd like to decompose the atom backgrounds
%                    into
%
% OUTPUTS
%   f - squared difference between current linear decomposition and atom
%        background image
%
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
% Define current linear decomposition of background based on coefficients A
SumBackgroundAlltmp = sum(bsxfun(@times,BackgroundAll,A'));
% Determine squared difference between current decomposition and atom
% background in order to minimize.
f = sum((AtomsExclusion - SumBackgroundAlltmp).^2);
Bibliography


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