

RICE UNIVERSITY

**Studies of one- and two-photon photoassociative  
spectroscopy using ultracold strontium**

by

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A THESIS SUBMITTED  
IN PARTIAL FULFILLMENT OF THE  
REQUIREMENTS FOR THE DEGREE

**Doctor of Philosophy**

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## ABSTRACT

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## Acknowledgments

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# Chapter 1

## Introduction

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Should probably mention somewhere that this is long-range PA, in contrast to short-range stuff being explored now.

[1]

### 1.1 Few-body physics

### 1.2 Halo molecules

### 1.3 Properties of strontium

### 1.4 Thesis Outline

$$V(x) = V_{lat} \sin^2(k_L x) \tag{1.1}$$

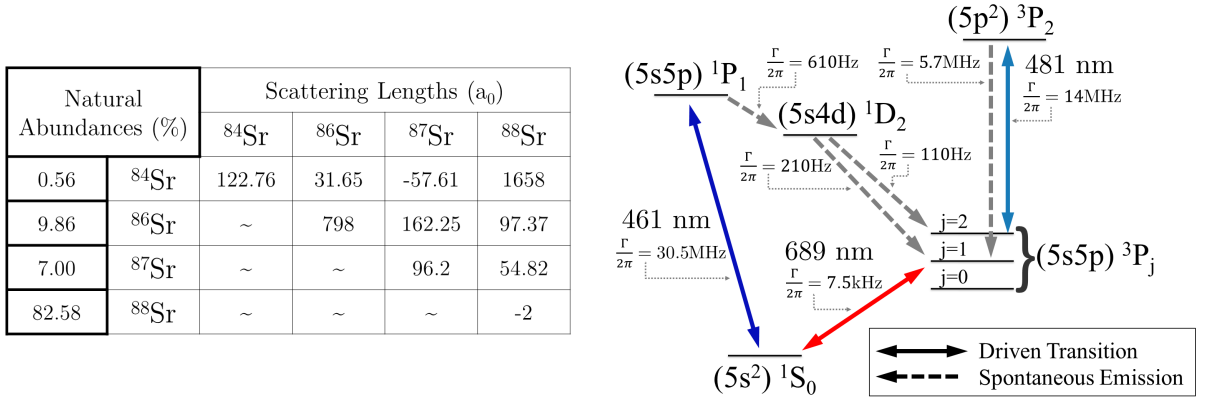


Figure 1.1 : Properties of strontium

Properties of strontium. Left: Natural abundances and s-wave scattering lengths for all mixtures of Sr. Right: Simplified energy level diagram of Sr showing the relevant states used for trapping and cooling of the atomic gas

## Chapter 2

### The Neutral apparatus

#### 2.1 Vacuum system

#### 2.2 Laser systems

##### 2.2.1 Wideband cooling stage: 461 nm

##### 2.2.2 Narrowband cooling stage: 689 nm

##### 2.2.3 Repumping: 481 nm

##### 2.2.4 Optical dipole trap: 1064 nm

##### 2.2.5 Optical toolbox

#### 2.3 Electronics

##### 2.3.1 Computer control system

##### 2.3.2 Standalone systems

#### 2.4 Apparatus benchmarks

## Chapter 3

### Photoassociation spectroscopy: theory and setup

#### 3.1 Introduction

##### 3.1.1 Low energy two-body scattering

consider the two particle system as a single entangled particle long range part of this quasi-particle is just the eigenstates of the separate particles themselves (only composed of two parts) but the short range part is going to be determined by some complex physics (new eigenstates, what is the coupling mechanism?) the vdW point is the boundary distance? coupling is due to the interatomic potential, there is at least the long-range part falling off as  $R^{-6}$ , what are the types of interactions which make up the internal wall?

Think I want to introduce the photoassociation by talking about the collisional wavefunction

what will that do?

I want to build up ideas about the FCF and need the wf for that to get qf I have to go back to scattering theory

ideas of the wavefunction become that basis for how you want to talk about interacting potentials

free atoms scattering as single particle state (different eigenstate) interaction determined by some gnarly stuff From scattering theory we know that the long range behavior is determined by short range physics how do we know this? (the dalibard intro) Can we come up with good enough pseudo potentials to describe the short range physics and then solve the schrodinger equation to extract wavefunctions? we want wavefunctions because that is the full characterization we don't know the right eigenbasis for the short range part but we can make some guesses (in particular Hund cases setup eigen states for various possible internal states) Bohn and Julienne theory guessed based on using quantum defect theory this pre-supposes that the bound and free wavefunctions are similar (I forget in what respect) but that the bound ones must go to zero as  $R \rightarrow \infty$  If we have some notion of the wf then we can construct matrices which define interactions once we add additional coupling to the scattering problem

now in a position where I need to connect scattering theory and the PAS

Once we have the ground state wavefunction of our new particle we can construct the internal structure by considering the internal energy structure of the constituent atoms Can I make a connection that since it is a composite particle we must consider all the various configurations available?

can I save?

### 3.1.2 Modifying interactions

### 3.1.3 PAS in atomic physics

## 3.2 Semi-classical treatment of lineshapes

## 3.3 Observing photoassociative loss

This is a test file for chapter 3

ok, I need to think how I am going to introduce photoassociation theory? It has to be quick. I am never going to get this done.

Okk, first we introduce the potential between atoms

this arises from the interaction of scattering theory with a molecular state. "the interaction potential between two atoms. Which is caused by?

this results in a potential that supports bound states. In atomic physics, our low density cases are mainly within the regime of small interactions. This spatial dependence is mapped onto the internal energy levels of each atom. I want to say dressed state model here (review atom-photon coupling, atomic physics book).

## Chapter 4

### Binding energy of the $^{86}\text{Sr}_2$ halo molecule

Describe and introduce halo molecules here

#### 4.1 Experimental methods

#### 4.2 Theoretical description

#### 4.3 Spectral fitting and determination of the binding energy

##### 4.3.1 Consideration of the trap depth

From the formula for the line shape we can see that it depends on the spatial distribution of the atoms. The standard approximation made when measuring these types of systems is to ensure loss does not cause heating of the atoms during photoassociation. Heating results in re-equilibration of the atomic density distribution, which in turn affects the rate of loss creation. Without independent controls to keep the system in thermal (and therefore spatial density) equilibrium.

What are the things the rate equation deals with?

We need the density distribution.

In a harmonic trap, there is a simple analytic form to the density distribution of a thermal gas. From Mi's work (and others) we know that this is only an approx-



imation that is valid when  $\eta$  is approx greater than 4. When greater than 4 we can apply the high- $\eta$  approx and the trap frequencies along a particular direction reduce to  $j\omega_j$ .

However, the trap we did this experiment in were at  $\eta$ 's of 1 or less so we don't have an analytic solution to the spatial distribution. Since this could be a problem we need to know what the trap looks like.

We measure trap oscillation freq. at several different powers and model the trap using the utility outlined somewhere else.

From the numeric model, we can define a spatially dependent  $\eta$  which is determined by the local trap depth which is simply the difference between the local potential energy and the global depth. This is illustrated in fig something.

The spatial information is not only important for the density determination, but also for the range of available thermal energies. Consider two atoms near the local bottom of the trap. By definition, in equilibrium, a single atom may only have up to the trap depths worth of energy since any additional energy would result in its expulsion from the trap. In this case, in a relative momentum frame, the allowed collision energies range from zero to two times the trap depth. Similarly, as we move towards the edge of the trap the range of accessible collision energies shrinks. This additional weighting factor may be viewed as having a local truncated Boltzmann distribution at every point in space.

Normally the BZ dist goes to infinity but here we have a cutoff at 2 trap depth. The most naive approx would be to simply consider the BZ and harshly

truncate at 2 trap depth. We tried this

We know this is unphysical since we should expect that the probability of observing a certain momenta at a certain point in space, should smoothly tend zero towards as we approach the edge of the trap. To see what this looks like we (and determine how important the effect is) we rederive the relative momentum distribution.

Some stuff about center of mass and relative

What were all the cases and conclusions of having done this? Remember to consider what the different cases are. If the total relative energy can be X then how does that get split up? Use the plots to show this limiting behavior. Like if particle 1 has all the energy then there is only one possible value for particle 2 (and vice versa).

DERIVATION for truncated trap below

Need to lookup references for this molecular chaos assumption. What about ergodicity? How to discuss that we may not be completely ergodic?

What does my potential look like? Can I make it a piecewise function? How should I introduce this part?

Where does the f equation come from? I believe this is just the normalized boltzmann factor for probability to occupy a particular state.

We can truncate this single particle distribution by

$$f_{\mathbf{r}}(\mathbf{p}) = A \left( \frac{1}{2\pi k_B T} \right)^{3/2} e^{\left( \frac{-p^2}{2mk_B T} \right)} \Theta \left( \epsilon_{max} - U(\mathbf{r}) - \frac{p^2}{2m} \right) \quad (4.1)$$

where A is a normalization constant which ensures  $\int_0^\infty f_{\mathbf{r}}(\mathbf{p}) d\mathbf{p} = 1$  and  $\Theta(x)$  is the Heaviside function defined by

$$\Theta(x) = \begin{cases} 1 & \text{if } x \geq 0, \\ 0 & \text{if } x < 0 \end{cases} \quad (4.2)$$

#### 4.4 Discussion of binding energy

We got a certain answer with the way shown in the paper.

We can also use a completely different method that ignores all the considerations of the last few sections. As was done in the calcium paper, we could simply fit the blue edge of the feature using a model function which can capture the high level features of the lineshape. Get the same answer. SHOW PLOTS TO THIS AFFECT AND COMPARE

#### 4.5 Calculating the bound-bound Frank-Condon factor

Maybe go a little into the isolated resonance model (or at least recall), then tie into how we can measure the susceptibility across several different detunings which can give us the coupling to intermediate level. The first order analysis of this data suggest a bound-bound rabi frequency of **BLAH**.

Point out the curling up at the end and say how the simple isolated resonance model cannot predict. A full coupled channel calculation probably could but in the spirit of the Bohn and Julienne semi-classical approach, we set out to derive an approximate analytic expression to determine the binding energies. This is presented in the next chapter.

Typical derivation of relative momentum probability distribution function

We begin by considering the single particle momentum probability distribution function (gotten how)? Single particle momentum probability distribution

$$f^1(\mathbf{p}) = \left( \frac{1}{2\pi k_B T} \right)^{3/2} e^{\left( \frac{-p^2}{2mk_B T} \right)} \quad (4.3)$$

Extension of this simple Boltzmann equation into the two-particle regime is complicated due to dependence of each particle on the others. If however, we make the assumption that particle collisions are rapid (on some timescale) we can approximate the two particle momentum distribution as the product of two single particle functions. This is known as the molecular chaos assumption and is important for what???

The two particle distribution for a homogeneous system is then

$$\begin{aligned} f^2(\mathbf{p}_1, \mathbf{p}_2) &= f^1(\mathbf{p}_1) f^1(\mathbf{p}_2) \\ &= \left( \frac{1}{2\pi k_B T} \right)^3 \exp \left( \frac{-(p_1^2 + p_2^2)}{2mk_B T} \right) \end{aligned} \quad (4.4)$$

Next, we'd like to consider a center-of-mass frame for the distribution. So we'll define

we define the relative and center-of-mass momenta of the two particles by defining

$$\mathbf{P}_c = \mathbf{p}_1 + \mathbf{p}_2 \quad M = m_1 + m_2 = 2m \quad (4.5)$$

$$\mathbf{p}_r = \frac{\mathbf{p}_1 - \mathbf{p}_2}{2} \quad \mu = \frac{m_1 m_2}{m_1 + m_2} = \frac{m}{2} \quad (4.6)$$

Now take Eq.4.4 and redfine in terms of Eq.4.5

Need to make a connection between dipole matrix element, wigner threshold, and infinite squarer step potential. This infinite square step can be viewed as a dilute ideal gas.

To prove this assumption I want to show that using the square step I can get the same equations like in Eq. 1 of the 99 paper. Then once we know the infinite energy behavior (valid for only a particular portion of energy due to s-wave constraint) then we can ask what happens if  $f(p)$  is truncated.

In the s-wave limit I need to write  $K$  as a function of  $f(p)$  (should do this in the appendix proof and reference in body). Given the form of the loss rate constant  $K$ , our problem reduces to determining the form of  $f(p)$  when  $\eta$  is finite.

Ok, so need to reference [2] to motivate usage of center of mass. Then use [3] Eq. 43 to reference the particular form

what is the throughline I want to make? Develop  $K_{in} - i$

## Chapter 5

### Strongly coupled PAS of a weakly bound molecule

#### 5.1 Introduction

#### 5.2 Experimental methods

#### 5.3 Three level model

#### 5.4 Resonance positions

#### 5.5 Lineshape

#### 5.6 Emergence of multi-photon Raman coupling

## Chapter 6

### Progress towards studies of quantum magnetism

A straightforward extension of the work presented in this thesis would be to control interparticle spacing via an optical lattice. For these and additional experiments using quantum degenerate fermionic strontium we purchased and installed an optical lattice system. Our lattice is implemented using a Coherent Verdi V-18 which is shapped and propagated to our science chamber in free space. Fig shows the optical path for each arm of our cubic lattice.

Unfortunately, complications due to heating when loading the lattice has limited our success in this optical trap. I want to go over what we have been able to do so far with the lattice.

How did we characterize? Kaptiza-dirac extension

What convinced us we were having problems?

What are some ideas we could do in the lattice? Zeno faster cooling via stimulated raman potentially? (can I model this somehow?) repulzively bound molecules? use interaction control in lattice with the zeno thing

## 6.1 Optical lattice: installation and characterization

### 6.1.1 Background

### 6.1.2 Setup

### 6.1.3 Measurement and results

## 6.2 Spin manipulation of $^{87}\text{Sr}$

Here is where I need to introduce and characterize the LCR

Averaging images together (how to use this code specifcally)

## 6.3 Search for narrowline PA molecules using various spin mixtures



## Chapter 7

## Conclusion

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- [1] J. A. Aman, J. C. Hill, R. Ding, K. R. A. Hazzard, T. C. Killian, and W. Y. Kon, “Photoassociative spectroscopy of a halo molecule in  $^{86}\text{Sr}$ ,” *Physical Review A*, vol. 98, p. 053441, nov 2018.
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- [3] T. L. Nicholson, S. Blatt, B. J. Bloom, J. R. Williams, J. W. Thomsen, J. Ye, and P. S. Julienne, “Optical Feshbach resonances: Field-dressed theory and comparison with experiments,” *Physical Review A - Atomic, Molecular, and Optical Physics*, vol. 92, p. 022709, aug 2015.

## Appendices

## Appendix A

### Repair of 922 Lynx master

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## Appendix B

### Imagefit analysis routine

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#### B.1 Background removal

Would like to remove noisy fringes to fit more easily

##### B.1.1 Principal component analysis

#### B.2 Fitting the spatial distribution

#### B.3 Evaluating fit parameters

##### B.3.1 How to write a new plugin

## Appendix C

### neuKLEIN - Killian lab experimental interface

During my time working on the neutral apparatus, Joe Whalen began a rewrite of the Labview based experimental control software which had grown organically through the first decade of the neutral apparatus' existence. Following this refactor, the user interface was also revamped to help reduce human errors and improve overall data collection efficiency. This chapter will outline the major components of the neuKLEIN software package and how this system integrates with the hardware control system and the software analysis algorithm.

#### C.1 Labview code

Need to have description of state machine.

Need to

Use of references for updating front panel

Need to get references for LV documentation for this stuff

Discuss triggered waveform oddity (retriggerable setup)

## C.2 FPGA code

The versatility of FPGA led us to want to simple system for setting static voltages and switching them at will. We built such a system using an National Instrument FPGA device (Xilinx something). The hardware details and circuitry are available in [appendix blah](#). This section will focus on the software side of programming and using the FPGA system.

This was originally a project started by a summer student named Weixuan Li in summer 2018. He did a good job.

Talk about special programs (the custom operation builder specifically)

## C.3 Possible future improvements

Specifically thinking about the ability to do network shared variables. Then we could dedicate a mahcine to an instrument and share the data using a networking layer instead of a hard connection.

Movement to full state machine

Standardization of the triggered waveform VI

implementation of coupled scanning mechanism (how is this currently handled)

## Appendix D

### Experimental control computer hardware

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#### D.1 Overview of status

#### D.2 Migration to a new machine



## Appendix E

### Custom circuitry

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#### E.1 AC line zero crossing trigger

#### E.2 Hard drive shutters

#### E.3 Power locks

#### E.4 Photodiodes

#### E.5 Infinite sample and hold

##### E.5.1 Reduced intensity fluctuations for sub-hertz exposures

## Appendix F

### Miscellaneous tips and tricks

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#### **F.1 Alignment of GHz AOM**

#### **F.2 Using the Picoscope in Labview(TM)**

#### **F.3 Liquid crystal retarder**

#### **F.4 Newport(TM) optomotion control**

#### **F.5 Fast analog lock for 689 nm**

#### **F.6 Porta CoM technique**

#### **F.7 Measuring Rabi frequencies**

## Appendix G

### Two-particle momentum probability distribution

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