Building Single Molecules from Single Atoms

A dissertation presented
by
Yichao Yu
to
The Department of Physics

in partial fulfillment of the requirements
for the degree of
Doctor of Philosophy
in the subject of
Physics

Harvard University
Cambridge, Massachusetts
March 2021

Thesis advisor: Professor Kang-Kuen Ni — Yichao Yu

Building Single Molecules from Single Atoms

Abstract

Contents

Acknowledgments

,

0

Introduction

1

Apparatus

1.1 Cooling and optical pumping beams

(MOT, OP, fiber back reflection)

(Mention Na Raman beam to be covered in later chapter?)

1.2 Tweezer and imaging

1.3 Molecular Raman frequency generation

(beam path, calibration)

2

Computer control of the experiment

2.1 Overall structure

2.2 Frontend

2.3 Backends

(communication protocol)

2.3.1 FPGA backend

2.3.2 NiDAQ backend

2.3.3 USRP backend

2.4 Automation of scan

3

Raman sideband cooling

3.1 Theory

3.1

3.2 Setup

3.2

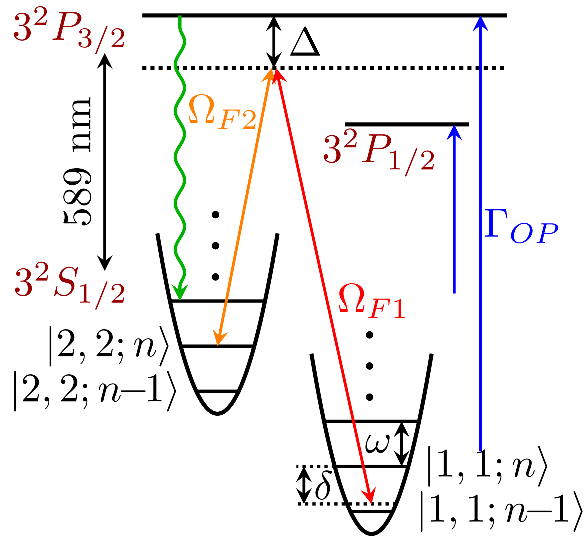

Figure 3.1: Single Na atom Raman sideband cooling scheme. The Raman transitions between $|2, 2; n\rangle$ and $|1, 1; n + \Delta n\rangle$ have a one-photon detuning $\Delta = 75$ GHz below the $3^2S_{1/2}$ to $3^2P_{3/2}$ transition. Two-photon detuning, δ, is defined relative to the $\Delta n = 0$ carrier transition. For optical pumping, we use two σ^+ polarized transitions, one to pump the atom state out of $|1, 1\rangle$ via $3^2P_{3/2}$ and one to pump atoms out of $|2, 1\rangle$ via $3^2P_{1/2}$ to minimize heating of the $|2, 2\rangle$ state.

3.3 Challenge with large Lamb-Dicky parameter

3.3

3.4 Solution: High order sidebands

3.4

3.5 Solution: Simulation based optimization

3.5

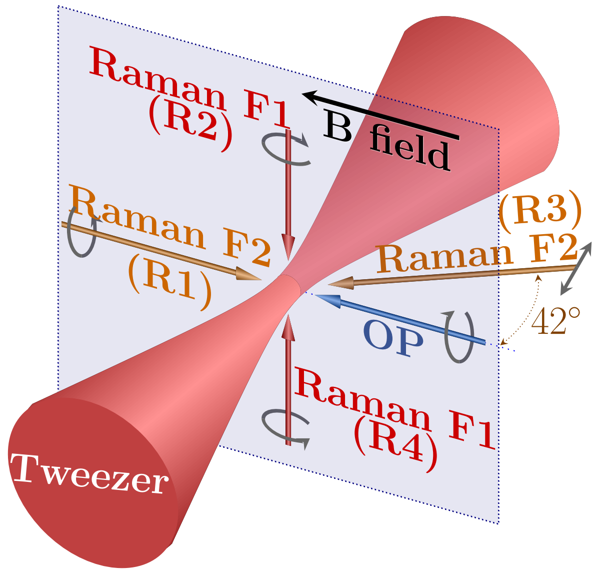

Figure 3.2: Geometry and polarizations of the Raman and optical pumping beams relative to the optical tweezer and bias magnetic field. Raman beams R1 and R4 address the radial x-mode. R1 and R2 address the radial y-mode. R3 and R4 address the axial z-mode, where the beams also couple to radial motion, but this coupling can be neglected when the atoms is cooled to the ground state of motion.

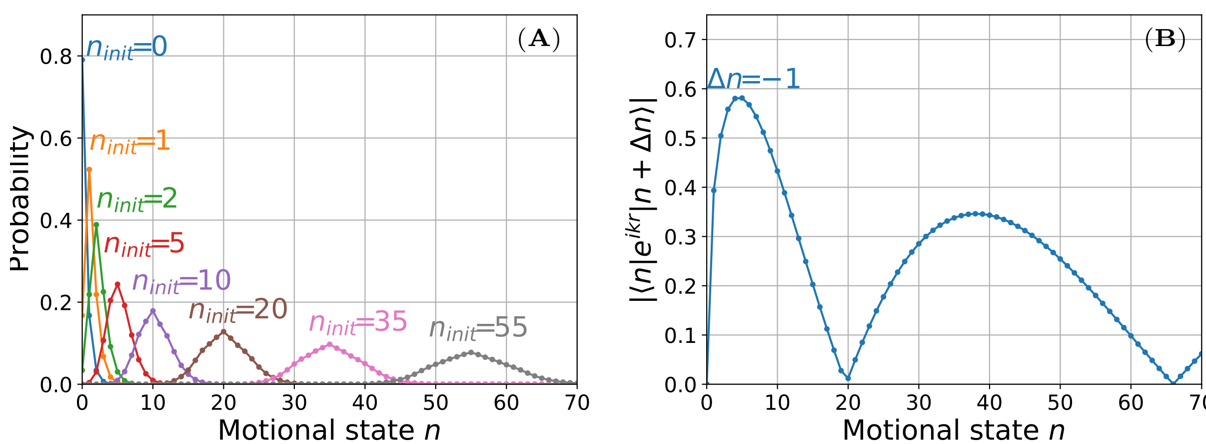

Figure 3.3: Optical pumping motional-state redistribution and Raman coupling for large LD parameters for the axial direction (z). The range plotted covers 95% of the initial thermal distribution. (A) Motional state distribution after one OP cycle for different initial states motion, n_{init}. Due to photon-recoil and the large LD parameter, $\eta_z^{\text{OP}} = 0.55$, there is a high probability of n changing. (B) Matrix elements for Raman transition on the first order cooling sideband deviate from $\sqrt{n}$ scaling with multiple minima.

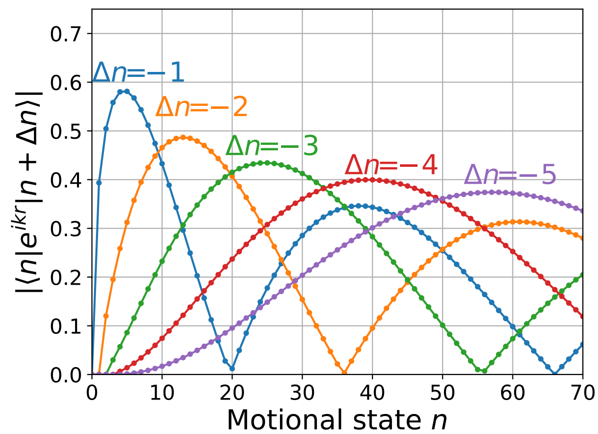

Figure 3.4: Matrix elements for Raman transition including high order sidebands. During cooling, we utilize the fact that high motional states couple most effectively to sidebands with large $|\Delta n|$ in order to overcome the issue with variation and dead zone in the coupling strengths.

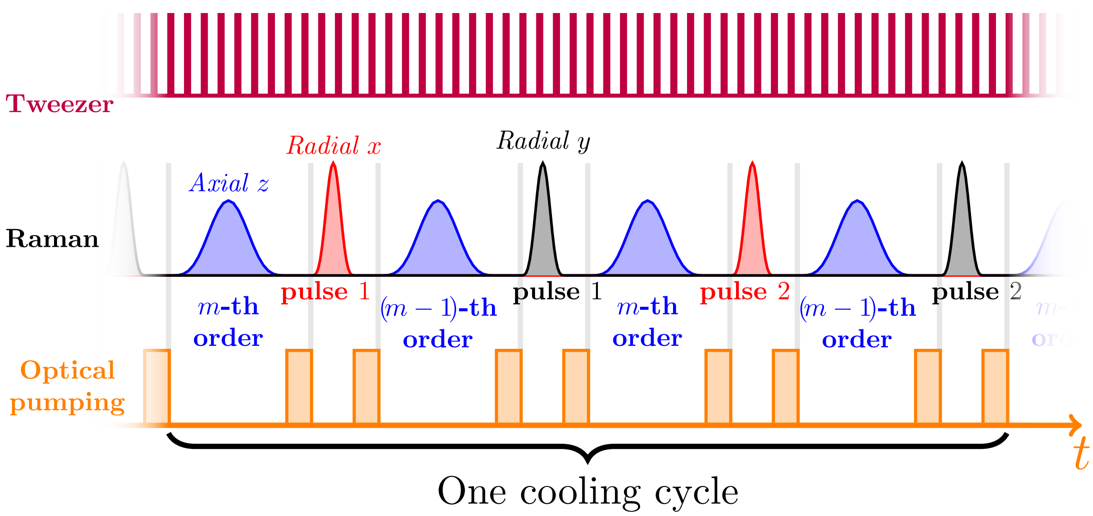

Figure 3.5: Schematic of the cooling pulse sequence. The tweezer is strobed at 3 MHz to reduce light shifts during optical pumping. Each cooling cycle consists of 8 sideband pulses. The four axial pulses address two sideband orders. The two pulses in each radial direction either address $\Delta n = -2$ and $\Delta n = -1$ or have different durations to drive $\Delta n = -1$, at the end of the cooling sequence when most of the population is below $n = 3$. The Raman cooling and spectroscopy pulses have Blackman envelopes to reduce off-resonant coupling, while the measurement Rabi pulses in Fig. and have square envelopes to simplify analysis.

3.6 Cooling performance

4

Interaction of single atoms

4.1 Scattering length

(Importance/relation with binding energy etc.)

4.2 Energy levels of two interacting atoms in an anisotropic trap

4.3 Interaction shift spectroscopy

(motional sideband, scattering length result)

4.4 Summary and Outlook

(Motional state selection)

5

Photoassociation of single atoms

5.1 Energy levels

5.2 Effect of the trap

(light shift, broadening)

5.3 Photoassociation spectroscopy

(v=0, 12, 14, etc)

6

Two-photon spectroscopy of NaCs ground state

(N=2, different HF states)

7

Coherent optical creation of NaCs molecule

8

Conclusion