

Quantum Bose and Fermi Gases of Dysprosium: Production and Initial Study

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Mingwu Lu

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Benjamin Lev, Primary Adviser

I certify that I have read this dissertation and that, in my opinion, it is fully adequate in scope and quality as a dissertation for the degree of Doctor of Philosophy.

Leo Hollberg

I certify that I have read this dissertation and that, in my opinion, it is fully adequate in scope and quality as a dissertation for the degree of Doctor of Philosophy.

Hideo Mabuchi

Approved for the Stanford University Committee on Graduate Studies.

Patricia J. Gumport, Vice Provost for Graduate Education

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Thesis advisor

Author

Benjamin Lev

Mingwu Lu

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Abstract

Advancements in quantum manipulation of ultracold atomic gases are opening a new frontier in the quest to better understand strongly correlated matter. By exploiting the long-ranged and anisotropic character of the dipole-dipole interaction in ultracold gases, we hope to simulate or create novel forms of quantum matter. Quantum dilute gases of the most magnetic element, dysprosium, provide an important platform to study this dipolar physics. We present details of experiments that produce strongly dipolar Bose-Einstein condensates of ^{164}Dy and ^{162}Dy , as well as the first dipolar degenerate Fermi gas of ^{161}Dy (0.2 x Fermi temperature) in low magnetic field. Particularly, we demonstrated for the first time the direct evaporative cooling of single component ultracold fermions (^{161}Dy) to Fermi degeneracy, predicted by universal dipolar scattering theory. Due to its large spin multiplicity in the ground state, large effective gauge fields for simulating, e.g., the quantum Hall effect, may be synthesized by exploiting the Dy atomic structure. Moreover, as a non-s-state atom, Dy is predicted to be an ideal candidate for spin-orbit coupling experiments in ultracold gases since the Raman dressing would be largely free from the incoherent scattering problems induced by the additional laser beams.

For my parents

Acknowledgments

When I finished the last chapter of my PhD dissertation, I found I have stayed in graduate schools for almost eight years. These eight years have been spent in the office as a theorist in the beginning and in lab as an experimentalist later, at two different universities, on a continent that I had never been to before. It was a wonderful journey. I would like to take this chance to thank all the people that I am indebted to along the way.

I am very grateful to my PhD thesis advisor Prof. Benjamin L. Lev. I considered him as an advisor second to none. I joined Lev Lab and started building the first Dy cold atom machine with Seo-Ho Youn (we were the only group members then), back at University of Illinois at Urbana-Champaign six years ago. Benjamin's intense enthusiasm for scientific research and hard-working spirit were a paradigm to me. Starting from zero, he taught me the essential technical skills as well as the ways to find answers in lab as an experimentalist; outside the lab he is a deep thinker, showing me what are the interesting questions that should be addressed in research. These were invaluable experiences that I will never forget.

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My first acquaintance with atomic, molecular and optics (AMO) physics stemmed from Urbana, where I took Prof. Brian L. DeMarco's course on modern atomic physics. In his class, I was impressed to know the detectability of the tiny size of a nucleus from atomic spectroscopy¹. Brian has been supportive for my AMO research and even earlier we had collaborations on the disorder experiments while I was still a theorist in a condensed-matter-physics group. He is a mentor to me whom I owe a lot.

¹Interestingly, years after, I could interpret Dy's measured isotope shifts partly due to this fact with a one-out-a thousand precision in Lev Lab.

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Papers and Publications

1. *Observation of low-field Fano-Feshbach resonances in ultracold gases of dysprosium*
K. Baumann, N. Q. Burdick, M. Lu, and B. L. Lev,
Physical Review A, **89**, 020701(R) (2014).
2. *Quantum degenerate dipolar Fermi gas*
M. Lu, N. Q. Burdick, and B. L. Lev,
Physical Review Letters **108**, 215301 (2012).
3. *Strongly dipolar Bose-Einstein condensate of dysprosium*
M. Lu, N. Q. Burdick, S.-H. Youn, and B. L. Lev,
Physical Review Letters **107**, 190401 (2011).
4. *Spectroscopy of a narrow-line laser cooling transition in atomic dysprosium*
M. Lu, S.-H. Youn, and B. L. Lev,
Physics Review A **83**, 012510 (2011).
5. *Dysprosium magneto-optical traps*
S.-H. Youn, M. Lu, U. Ray, and B. L. Lev,
Physics Review A **82**, 043425 (2010).
6. *Anisotropic sub-Doppler laser cooling in dysprosium magneto-optical traps*
S.-H. Youn, M. Lu, and B. L. Lev,
Physical Review A **82**, 043403 (2010).
7. *Trapping ultracold dysprosium: A highly magnetic gas for dipolar physics*
M. Lu, S.-H. Youn, and B. L. Lev,
Physical Review Letters **104**, 063001 (2010).

Chapter 1

Introduction

1.1 Why dysprosium is interesting

Advancements in quantum manipulation of ultracold atomic gases are opening a new frontier in the quest to better understand strongly correlated matter. One interesting field is the study of quantum soft matter phases which resemble the classical liquid crystalline and glassy phases [1, 2]. These non-fermi liquid phases, intermediate between canonical states of order and disorder, manifest in 2D electron gases, iron-based high-temperature superconductors [3] and other condensed matter systems. Ultracold highly magnetic atoms [4, 5, 6] may provide a unique clean platform to study these so-called quantum liquid crystal phases [7, 8], due to the competition of the long ranged, anisotropic dipole-dipole interaction (DDI) with other short ranged atomic interactions. Also, topologically nontrivial superfluid phases may arise from strong DDI induced p -wave BCS pairing in 2D, which may be applicable to topologically protected quantum information processing [9]. Recently, dipolar gases have

become interesting for simulating meson condensation in the dense nuclear matter, a topic conventionally studied in the context of neutron stars [10].

In addition to the rich physics induced by the strong DDI, these magnetic atoms with high spin angular momenta may be used to explore a diversity of fascinating phenomena —non-Abelian spinors, spin-orbit-coupled spinors and high-spin Hall states arising from large Abelian gauge fields [11, 12, 13]. Some of these interesting quantum phases are so unique that they are perhaps not readily realizable in the conventional solid state systems.

Ultracold dysprosium gases, composed of highly magnetic and high spin atoms, form a very desirable candidate in studying the above exotic quantum many-body phenomena. Being extraordinarily magnetic compared to other common ultracold gases, dysprosium gases may also be incorporated in the novel atom chip microscope our group recently built and which may exhibit an unprecedented combination of field sensitivity, spatial resolution, and temperature tunability [14]. In the following sections, I will describe the development of such a quantum gas machine of Dy as my PhD study.

1.1.1 Dysprosium: the most magnetic element

Dysprosium (Dy) is a lanthanide element with an open 4f-electron shell. Unlike alkali and alkaline earth metals, non-*s*-state Dy atoms possess large total angular momenta: $8\hbar$ for bosonic isotopes and $21/2\hbar$ for fermionic isotopes. It has the largest permanent magnetic moment: 10 Bohr magnetons in the ground state, which provides an optimal candidate to explore the DDI in dilute atomic gases. In comparison, Dy



Figure 1.1: Dy metal samples. From MaterialsScientist at en.wikipedia.

is 10 times more dipolar than the earlier studied dipolar atom chromium [15], as measured by dipole lengths. Furthermore, five abundant isotopes of Dy exist in nature. This greatly facilitates the study of quantum Dy gases with different spin statistics as well as Bose-Fermi mixture.¹

1.1.2 Cooling and trapping dysprosium

Reaching the quantum regime in Dy gases is technically challenging, considering its very complicated atomic structure. Laser cooling and trapping techniques sensitively depend on the favorable details of atomic optical transitions; successful evaporative cooling (so far the simplest, most general route to the quantum regime) requires suitable ultracold collisional properties of atoms. Before our experimental studies it was not clear whether dysprosium would meet these criteria.

We chose 421 nm and 741 nm as the relevant laser cooling transitions and performed their spectroscopy [16]. We successfully demonstrated the first 421 nm magneto-optical trap (MOT) of all Dy isotopes [17]. The numerous metastable states above the ground state were long thought to be dangerous loss channels that quenches con-

¹Terbium also possess a dipole moment of $10\mu_B$ but is only a boson and possesses low-lying blackbody coupled states.

tinuous laser cooling. Experimentally, however, we found that this branching ratio is fortunately small enough that atoms in metastable states remain trapped due to their large magnetic moments, and later relax back to the ground state and are recycled in the MOT in much less time than the background gas-limited lifetime. These special properties of Dy make a repumperless MOT possible, as was first demonstrated in Er by the McClelland group at NIST [18].

For further enhancing the phase space density of trapped Dy gases, we cooled Dy atoms down to hundreds of nK via the narrow-line laser cooling at 741 nm (only 2 kHz in natural linewidth). In fact, the 741 nm Dy MOT is among the coldest MOTs based on Doppler cooling [19]. Fully exploiting its potential for low Doppler limited temperatures may lead—when combined with other techniques, e.g., demagnetization cooling [20] or in a dimple trap [21] to efficient generation of quantum gases completely free of evaporative cooling.

1.1.3 Strongly dipolar Bose-Einstein condensation

Efficient evaporative cooling in a loaded crossed optical dipole trap successfully led to the first Bose-Einstein condensate (BEC) of ^{164}Dy gas [5], followed soon by the condensation of ^{162}Dy (which we have not yet reported). In these strongly dipolar Dy BECs, the ratio of the DDI to the mean field energy associated with the isotropic s -wave scattering is on the order of unity. This was demonstrated by the collapse of a trapped Dy BEC when the quantization field is aligned to the weakest trapping direction of a pancake-shaped trap, which tunes the dominant anisotropic DDI to be effectively attractive. Similar collapse was studied earlier in a dipolar Cr BEC,

but Fano-Feshbach (FF) resonance tuning on a s -wave scattering was employed [22]. The fact that the Dy experiment showed BEC collapse without the assistance of FF tuning is a strong indication of the non-perturbative-nature of that DDI that governs the interactions between Dy bosons, even with a typical value of scattering length (100 Bohr radius) measured far away from a FF resonance. Due to the strong DDI between atoms, the expansion of a released Dy condensate is strongly angled towards the direction of an external homogeneous magnetic field. This was the first observation of such a phenomenon, which vividly demonstrated a quantum analog of ferrofluid behavior [23, 5].

1.1.4 First dipolar degenerate Fermi gases

The past decade witnessed technical breakthroughs in producing ultracold polar molecules. So far, the lowest energy scale accessible in trapped molecules is limited to $T/T_F \sim 1$, which is achieved in a KRb system [24]. Highly magnetic atoms, on the other hand, are free from the complexities of chemical reactions or other loss channels in cold molecule gases. Moreover, the magnetic dipole moment exists in the absence of magnetic fields, while the electric dipole moment must be saturated by an electric field that breaks spherical symmetry. This lack of $O(3)$ spontaneous symmetry breaking in polar molecule systems is an important difference when considering this use of dipolar particles for exploring quantum liquid crystal physics.

We produced the first deeply degenerate ($T/T_F = 0.2$) atomic Fermi gas of ^{161}Dy , which constitutes a breakthrough towards quantum simulation with dipolar fermions. We did this by both sympathetically with a bosonic ^{162}Dy gas [25] and by exploiting

universal dipolar scattering. It is well known that spin polarized fermions will form non-interacting gases with a vanishing elastic collisional cross-section in the ultracold regime. However, this is true only if the interparticle interaction is short-ranged, e.g., like the van der Waals force. By contrast, it was predicted that if the long-ranged DDI dominates, the collisional cross section would be a nonzero value which scales as the dipole moment to the fourth power [26]. We experimentally demonstrated that direct evaporative cooling could bring the single component, spin polarized ^{161}Dy gas into the quantum regime ($T/T_F = 0.6$) [25]. This experimental evidence of efficient thermalization clearly shows for the first time that dipolar scattering does not quench for identical fermions even far below the p -wave barrier energy scale and in this case below T_F .

1.2 Summary and Outlook

In summary, our experimental work [5, 16, 17, 25, 27, 28, 29] in Prof. Lev's group demonstrated that quantum Dy gases could be produced, both for bosonic and for fermionic isotopes. Some peculiar features of DDI were unveiled and additional interesting ones are to be explored.

For the simulation of synthesized gauge fields and spin-orbit coupling physics, Dy provides several unique strengths: large spin multiplicity in the ground state enhances the artificial gauge field by almost one order of magnitude compared to Rb atoms [12], which is desirable for simulations of quantum Hall effect [30]; Dy's non- s -state electron structure could considerably lower the heating rate arising from incoherent scattering, a current technical bottleneck, in the Raman beam dressing scheme for

the quantum simulation of spin orbit coupling [31, 32, 12]. Experimental preparations are underway towards this direction.

1.3 Thesis Outline

Chapter 2 describes subsystems of the Dy machine, including the ultra-high vacuum (UHV) chamber, magnetic field control setup and the computer control system. Chapter 3 presents the details of the 780, 421, 741, 1064 nm laser systems as well as rf sources, as part of the Dy machine. Precision servo control of laser systems will be the focus. Chapter 4 discusses details of the experimental detection method for cold atom imaging, as well as spin state detection and control. Chapter 5 describes the general features of Dy's optical transitions and how the 421 nm blue MOT (bMOT) of Dy works. Experimental work of magnetic trapping is also presented. Chapter 6 explains the principle of the narrow-line 741 nm red MOT (rMOT) as well as the experimental implementation. Chapter 7 presents the optical dipole force trapping of Dy at 1064 nm, and discusses Dy's dynamic polarizability. I have included a brief review of ultracold collisions of highly magnetic atoms, as this would be useful to future students even though there are more and more publications covering each part of this broad topic, resulting Chapter 8. Chapter 9 and Chapter 10 are dedicated to the production of Bose-Einstein condensates and degenerate Fermi gases of Dy respectively, together with a preliminary study of their peculiar features.

Chapter 2

Dysprosium Machine I

2.1 Ultra-high vacuum chamber

The Dy cooling and trapping apparatus consists of a high temperature oven, a transverse cooling stage, a Zeeman slower, and the chamber for magneto-optical trapping (Fig. 2.1). This subsection will describe them in sequence.

Dy has a relatively high melting point: 1412 °C. Therefore, a high-temperature oven¹ is used to vaporize Dy nuggets at 1250 °C. Dy atoms effuse from a Ta crucible inside the oven.² The atoms are collimated via a 270-mm long 5-mm wide differential pumping tube that connects the oven chamber and later stages with a low conductance. The chilled-water cooling system in the oven chamber effectively removes the extra heat due to blackbody radiation from the oven; thus the temperature of the chamber body in the oven zone is only slightly above room temperature during op-

¹Custom made from SVT Associates, In. The PID control servos the oven temperature with a precision better than 1°C.

²While molybdenum is another frequently used element for building crucibles, it chemically reacts with Dy at high temperatures and therefore it should not be considered.

eration. For the purpose of atomic beam control, a shutter driven by compressed air is incorporated in front of the crucible aperture to block the beam when needed. An ion pump with a 70 l/s pumping speed in this stage keeps the background pressure in the oven zone under 1×10^{-9} Torr during experiments. A gate valve separates this oven chamber from the following stages so that we may refill the Dy source without disturbing the much lower pressure in the UHV chamber after the gate valve .

The following stage is a 6-way cross chamber with four optical viewports orientated normal to the direction of the atomic beam. In the past, we carried out laser spectroscopy on the atomic beam in this zone and now it is reserved for transverse cooling of the atomic beam. During the 421 nm Dy MOT's loading period, two retroreflected elliptical laser beams come through the four viewports in orthogonal directions to realize 2D transverse cooling on the Dy atomic beam [33]. This results in a more collimated atomic beam, which enhances the beam's flux in the MOT region by a factor of four [27]. A second ion pump is installed in this stage where the vacuum pressure is held at about 9×10^{-11} Torr.

The Zeeman slower in the next stage slows down the longitudinal atomic speed for MOT loading. It is a σ^- spin-flip type Zeeman slower with a capture velocity of 480 m/s and an extraction velocity of 30 m/s by design. We adopted coils designed with multiple layer so that the current through the coils is only a few amperes during the operation. For efficient laser slowing, a one-watt slightly focusing 421 nm laser beam is sent through the Zeeman slower, counter-propagating against the Dy atomic beam.

Immediately after the Zeeman slower is the MOT chamber. The main body is a

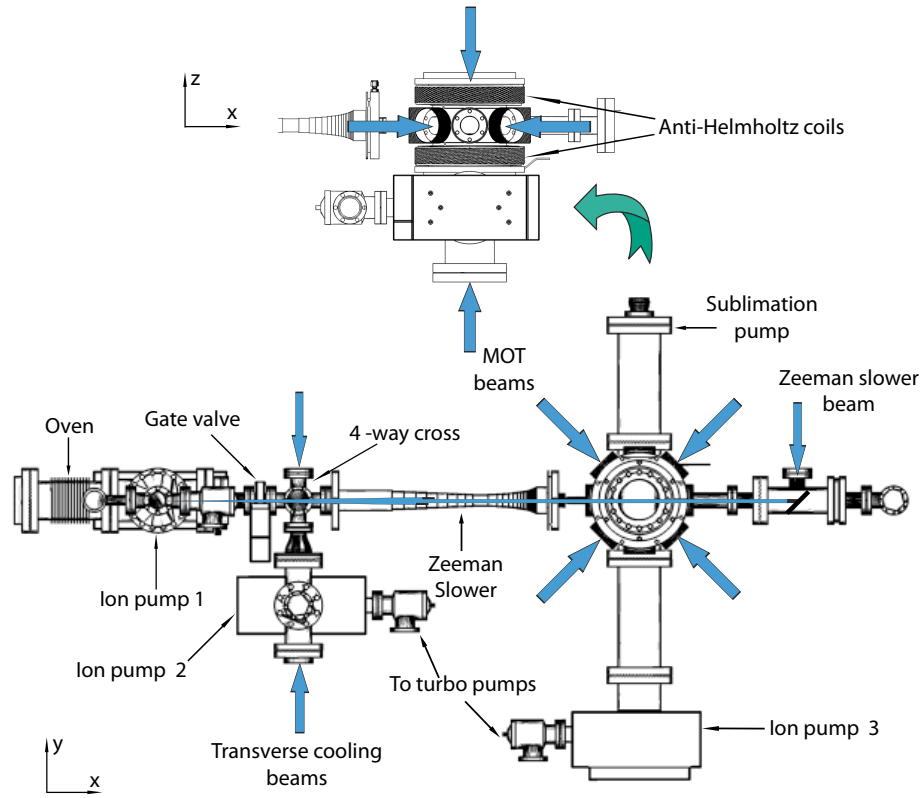


Figure 2.1: Illustration of Dy vacuum system. Upper panel is the side view of MOT chamber and lower panel the side view of whole vacuum chamber.

6" flange octagon sandwiched in the vertical direction by a pair of large MOT coils. The octagon has six anti-reflection-coated³ viewports in the horizontal plane and two 6" ones in the vertical direction. The collinear 421 nm and 741 nm MOT beams are sent through four viewports in the horizontal plane and retroflected, defining the x and y directions in experiments, while the 6" vertical windows offer access for the MOT beams in the z direction. The extra pair of viewports in the horizontal plane is used for the absorption imaging beam. The original viewport on the octagon chamber for the Zeeman beam's entrance was extended and rotated by 90° with a right angle mirror installed behind it [27, 34]. This indirect introduction of the Zeeman beam prevents direct coating on the viewport by Dy atoms which deteriorates transmission efficiency of the Zeeman beam after only a few days of experimental running. A Ti-sublimation pump together with an ion pump of 70 l/s pumping speed keeps a background vacuum of low 10^{-11} Torr in the MOT chamber, after careful baking of the whole vacuum chamber. This leads to a vacuum pressure limited life time of trapped gases of tens of seconds.

2.2 Magnetic field control

Precise, fast and stable magnetic field control is important in experiments. Earth's field is carefully cancelled by three pairs of Helmholtz coils in orthogonal directions with the residual field determined by RF spectroscopy of Zeeman sub-levels. A homogeneous magnetic field is generated by one pair of these Helmholtz coils in the

³The anti-reflection (AR) coating is a custom quad-band coating with less than 1% residual reflectivity at 421 nm, 532 nm, 741 nm and 1064 nm.

vertical direction by offsetting its current value from the one that would cancel out the ambient field in the same direction. Quantization coils are placed in the horizontal plane along the imaging beam's direction. All currents in those coils are servoed in PID (proportional-integral-derivative) control circuits with residual relative current fluctuations less than 5×10^{-5} , which is mainly limited by the 60 Hz main line noise. Carefully grounding the electronics proved to be crucial. To further reduce the influence of main line noise (e.g., in rf spectroscopy or spin rotation in the Bloch sphere experiments), the experimental control sequence is triggered by the main line to synchronize the phase for improved repeatability in results. Most of the coils can be turned on or off fully within a few hundreds of μs [34], after optimizing the transfer functions of the control circuits. The quadrupole magnetic field for Dy MOTs and magnetic trapping (MT) is generated by a pair of anti-Helmholtz coils which is actively cooled by circulating chilled water from inside. Two coils are connected in series to cancel out the possible differential current noise. While the quadrupole field could be turned off within 1 ms, the turn-on time is a few ms mainly due to the considerable inductance of the MOT coils and finite dc supply voltage available.⁴

2.3 Computer control

The custom control system of the Dy experiment consists of digital/analog sequencers and an imaging acquisition system. The sequencer control is realized by MATLAB-based custom written code (together with custom functions written in

⁴Due to historic reasons, we did not adopt a bucket-window design. The coils are widely separated by the MOT chamber and their sizes are correspondingly big. Also, we did not push the MOT current to extreme values in order to reduce the number of turns in the coils so as to reduce the inductance.

C++ for interfacing between hardware and MATLAB codes) which write commands to the National Instruments digital and analog output devices. These channels send electronic signals to trigger the imaging CCD, optical beam shutter, the atomic beam shutter, the turning on and off of acoustic optical modulators (AOM) and the RF coils, and to control the current servo point for the MOT coils and other coils for magnetic field control. The imaging acquisition system consists of the imaging system (see the chapter on experimental detection) and the computer for the imaging data reading and processing. The triggered images are processed in MATLAB-based programs for real time analysis and/or stored on the local drive.

Chapter 3

Dysprosium Machine II

3.1 Introduction of laser systems

The importance of a robust and well-controlled laser system can never be emphasized in producing dilute quantum gases. For an ultracold Dy experiment, it is imperative to build reliable and agile laser systems meeting a variety of stringent requirements because of the features of this species.

For the MOT based on one of strongest Dy laser cooling transitions at 421 nm, it is necessary to generate watt-level blue laser beams due to the high saturation intensity of this line.¹

For the 741 nm narrow line MOT, coherence of the laser system is the prime concern, and stabilization of the external cavity diode laser's (ECDL) frequency to a passively stable optical cavity with sub-kHz linewidth is the key ingredient of this

¹ $I_s = \pi \hbar c \Gamma / 3 \lambda^3$ is a natural light intensity scale indicating the onset of saturation in driving the two-level system of atom under study. Because the wavelength λ is short and the natural linewidth Γ is fairly large for this 421 nm transition, I_s is large.

laser system.

Besides the linewidth control of lasers, the long-time frequency stability of the laser system is also extremely important, and considerable effort was needed to achieve the desired stability. Due to the experimental need to co-trap different isotopes, we require that the laser systems either be agile enough to continuously sweep between the frequencies necessary for different isotopes, or they consist of multiple interlocked laser sources with excellent coherence.

Dy, being a massive element (almost twice as heavy as Rb), is less polarizable in an external near infrared (NIR) electromagnetic field than alkali atoms. Hence, a high power (20-50 W) 1064 nm laser system is very desirable for optical dipole force trapping.

This chapter describes details of each of the laser systems for cooling and trapping, keeping in mind the issues and constraints above, and focusing on laser frequency stability. Imaging light, which also derives from the 421 nm laser system, will be discussed in the chapter for the detection.

3.2 Laser frequency stabilization

3.2.1 Phase and frequency noise

A laser is a relatively coherent light source, but in reality the phase coherence in laser's emission is not strictly maintained. Deviations in phase from pure sinusoidal oscillations occur. Such deviations, or so-called phase/frequency noise² take place in

²They are not the same but they are very closely related as we shall see.

a wide range of time scales and certain distributions. For example, frequency noise could be large in amplitude but narrow in Fourier frequency distribution, or small in amplitude but present in a wide Fourier frequency range (fast and slow frequency jitter), or a combination of the two. In practice, fast frequency jitter is not resolved in many experiments and the laser's center frequency is broadened away from a Dirac delta peak. The corresponding width in frequency distribution is called the laser's linewidth, roughly speaking, and it is a useful figure of merit in judging a laser's coherence. There is also large slow frequency excursion that is readily resolved, often referred to as frequency drift.

The division above is somewhat arbitrary and depends on one's partitioning of time scales. For laser cooling experiments, the important time scale is the microsecond or sub-microsecond regime during which atoms can undergo at least a few cycles of photon scattering for cooling. Many modern single-frequency solid state lasers can provide a linewidth on this time scale comparable to or less than the natural linewidth of cooling transitions in typical atoms ($\Gamma \sim 10$ MHz for alkali atoms). Active servoing of fast noise (or so called linewidth reduction) is not essential. On much longer time scales comparable to an experimental data-taking cycle (minutes typically), the laser center frequency often travels much more than Γ due to thermal effects. A frequency servo is often needed to cancel this drift.

Dy narrow-line laser cooling employs a 1.8 kHz wide optical transition. For such a transition, a single photon scattering takes $180 \mu\text{s}$. Therefore we desire a laser source which maintains good coherence on or beyond the ms scale. To avoid possible conceptual confusions, we will first derive some important relations in a more rigorous

way. Also a quantitative analysis is helpful in diagnostics when we improve the laser frequency stability in experiments. In the following derivation of important frequency noise relations, we mainly follow the notations in [35].

In the absence of amplitude noise, optical oscillations in electric field components can be written in complex representation as:

$$E(t) = E_0 \exp[i(2\pi\nu_0 t + \phi(t))] \quad (3.1)$$

where E_0 is the amplitude of oscillation, ν_0 the nominal oscillation frequency, and $\phi(t)$ the random phase noise as a function of time. To avoid confusion, we denote oscillation frequency as ν and Fourier frequency as f .

From the definition of instantaneous frequency, we can see that phase noise and frequency noise are interrelated, as expressed in the time domain

$$\delta\nu(t) = \nu(t) - \nu_0 = \frac{d\phi(t)}{dt} \quad (3.2)$$

and in the frequency domain

$$S_{\delta\nu}(f) = f^2 S_\phi(f) \quad (3.3)$$

where $S_\phi, S_{\delta\nu}$ are the phase and frequency noise power spectra respectively [36]. Roughly speaking, S_ϕ ($S_{\delta\nu}$) is the Fourier transform of autocorrelation functions of phase (frequency) in time.³ We will refer only to single-sided noise spectra.

The autocorrelation function of the electric field itself in time, which is defined as $R_E(\tau) = \langle E^*(t)E(t+\tau) \rangle_t$ ($\langle \dots \rangle_t$ denotes time average)⁴ can be mathemati-

³There are complexities related to whether the process is stationary or not (which often not in real world), which we do not address here.

⁴Note that it is $\langle E^*(t)E(t+\tau) \rangle_t$, not $\langle \phi^*(t)\phi(t+\tau) \rangle_t$, which is R_ϕ . Failing to notice the difference may lead to confusions conceptually.

cally proven to be related to the frequency (phase) noise power spectrum [37] in the following way:

$$\begin{aligned} R_E(\tau) &= E_0^2 e^{i2\pi\nu_0\tau} \exp\left[-\frac{1}{2} \langle [\phi(t+\tau) - \phi(t)]^2 \rangle_t\right] \\ &= E_0^2 e^{i2\pi\nu_0\tau} \exp\left[-2 \int_0^\infty S_{\delta\nu}(f) \sin^2(\pi f\tau)/f^2 df\right] \end{aligned} \quad (3.4)$$

This important result comes with three parts. E_0^2 could be normalized to one and is therefore not essential. The oscillatory part $e^{i2\pi\nu_0\tau}$ reflects a deterministic phase evolution for an ideal sinusoidal oscillation, and thus is also not a concern. The last part decaying exponentially represents the decoherence due to random frequency/phase noise, which is the essential term. The weight function $\sin^2(\pi f\tau)/f^2$ in the decoherence term, which scales as $1/f^2$, indicates that the frequency noise in the low frequency regime contributes more than those in the high frequency regime. This provides some insight in improving a laser's coherence effectively; combating the low frequency portion of the noise spectrum is essential.

Finally one gets the power distribution of oscillations, or the laser's line shape (in experimentalists' terms) via the Fourier transformation of R_E :

$$S_E(f) = 2 \int_{-\infty}^{\infty} e^{-i2\pi f\tau} R_E(\tau) d\tau \quad (3.5)$$

From the discussion above, we can see that starting with measurable noise power spectra, one Fourier transformation with some weighted function is needed to get the coherence (decoherence) function, and one more Fourier transformation derives the laser's line shape. In general, this must be calculated numerically. Analytical calculation is available only for some simple noise power spectra. For example, in the case of white frequency noise ($S_{\delta\nu} = \text{const.}$), it is known that $S_E(f)$ is Lorentzian

and the full width half maximum $\Delta\nu = \pi S_{\delta\nu}$, which can be obtained directly from the formula above. Another case is in the situation of a narrow noise spectrum distribution near DC with high modulation indices ($S_{\delta\nu}(f) \gg f$), which corresponds to a Gaussian line shape. This noise distribution is often due to the presence of technical frequency noise.

In practice, technical frequency noise often dominates over the white noise at low Fourier frequencies, scaling as $1/f$ or sometimes faster. This makes the integral in eqn. 3.4 converges slowly or even diverge. In practice, the integration limit is always finite and is cut off at some frequency that corresponds to the time scale of interest in experiments. Laser linewidth is therefore integration time dependent, and normally the time scale is clear in different contexts.

3.2.2 PDH cavity lock

The Pound-Drever-Hall (PDH) technique [38] is a powerful tool to extract information about the instantaneous frequency/phase noise of a laser and to provide a dispersive error signal for laser frequency control. Mathematical and physical details are addressed in the literature, e.g., the review by E. Black [39], hence only the principle will be outlined here in order to understand the implementation and experimental results.

For a frequency modulated laser beam, the amplitude is

$$E(t) = E_0 \exp[i(\omega t + \beta \sin(\Omega t))] \quad (3.6)$$

where ω and Ω are the optical frequency and RF modulation frequency respectively, and β is modulation depth. A mathematical expansion [40] into a Bessel series leads

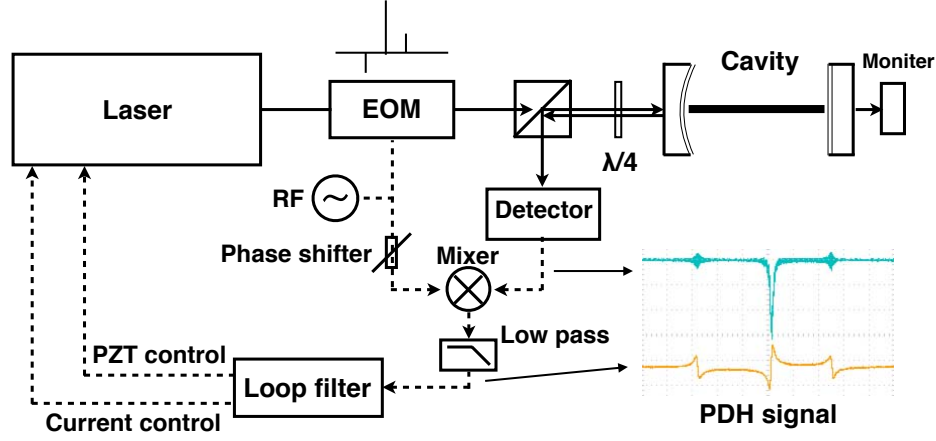


Figure 3.1: Pound-Drever-Hall cavity lock scheme in a very basic form. Solid lines are optical signal paths and dashed lines electronic signal paths. PDH signals in inset are taken from real measurement. See text for details.

to:

$$E(t) = E_0 \left[J_0(\beta) + \sum_{m=1,2,\dots} [J_m(\beta) + (-1)^m J_m(\beta)] \right] e^{i(\omega t + m\Omega t)}, \quad (3.7)$$

where the J_0 term is normally called the carrier, and all higher J_m terms the sidebands. For small modulation depth only the carrier and 1st order sidebands $J_{\pm 1}$ need to be considered. The two sidebands are 180° out of phase,⁵ hence their individual beats with the carrier⁶ will normally cancel out each other.

The reflected beam from an optical cavity derives an extra $180^\circ + \theta(\omega - \omega_0)$ phase change [39], where θ is an odd function of frequency detuning $\omega - \omega_0$ with respect to the cavity's resonant frequency ω_0 . Therefore the two beat signals can partially constructively interfere with each other after cavity reflection, and the sum amplitude is also an odd function of frequency detuning. A frequency-detuning-

⁵This is quite general for pure frequency modulation, while in amplitude modulation they are in phase.

⁶The two sidebands can beat at twice the modulation frequency, but it is a higher order process with low modulation depth which would be filtered out after a frequency mixing.

sensitive, dispersive PDH error signal is obtained after frequency demodulation of the resulting carrier-sideband beats. Additionally, notice that when the carrier frequency is resonant with the cavity, the reflection is zero [39], providing a desirable zero crossing of the PDH error signal at the center of the cavity resonance. After proper filtering, this signal is used for laser frequency feedback control.

The essential advantage of the PDH technique compared to cavity transmission error detection lies in the cavity response. Cavity transmission is the leakage from the intracavity field, and is inherently low-frequency-pass filtered [38, 41]. The PDH technique employs the cavity reflection which can be regarded as the sum of the directly reflected beam that never enters the cavity and the leakage from the cavity build-up field. The information of fast phase⁷ changes in the laser beam is imprinted in the cavity reflection as the result of optical heterodyning. This is particularly desirable for fast frequency servoing of lasers with wide frequency noise distributions (e.g., diode lasers).

3.2.3 PDH signal optimization

In an experiment, to obtain a symmetric, non-drifting PDH error signal with high SNR in a wide bandwidth is not trivial. Much trial-and-error was involved, along with guidance from the literature [42, 43, 44, 45].

The signal contrast at resonance in cavity reflection is never 100%. There is always residual reflection due to imperfect mode matching as well as impedance matching of the cavity. Careful control of the waist and wavefront curvature of the laser

⁷In the high frequency regime which is far beyond the cavity's linewidth, the frequency response of the cavity is essentially a phase response, because of the integration effect of the cavity field [41].

beam via lenses can improve the mode matching between the input beam's mode and the cavity's Gaussian mode [42, 41], but often the contrast is limited by impedance matching [41],⁸ especially for high finesse cavities. For a cavity with identical mirrors, the power ratio of the reflected beam and input beam on resonance is given by $I_r/I_{in} = A^2/(2A + 2T)^2$, where scattering and absorptive loss A , transmission T , together with reflection of cavity mirrors R fulfill the relation: $A + T + R = 1$ [46]. The low scattering and absorption of mirror coating, based on the ion sputtering technique, reduces the residual cavity reflection on resonance. In our experiments, normally 70 – 90% of laser power can be coupled into cavities.

Theoretically, modulation depth of $\beta = 1.08$ maximizes the PDH error signal amplitude [39]. In practice, the modulation coefficient of an electro-optical modulator (EOM) based on a Pockel cell is relatively small, so for normal rf drive, β is usually less than 20-30%.⁹ LC resonant circuits [42] can bring up the modulation voltage to the optimal value under normal driving levels. We use an air core coil as an RF inductor to creat an LC circuit,¹⁰ achieving Q values of 10 - 20 depending on frequencies. To maximize the size of the demodulated signal at fixed modulation depth, the relative phase between the local oscillator and the detector signal is tuned either by a phase shifter circuit, or by changing the modulation frequency itself at fixed electrical path length, or by changing the coaxial cable length between the local oscillator and the mixer input port. For resonant EOMs working at high modulation frequencies (50-100 MHz), the last method is very convenient and was adopted in the experiment.

⁸As for concept in electronic circuits, no reflection occurs when impedance is matched.

⁹This corresponds to the power in a sideband $[J_1(\beta)]^2 \sim (\beta/2)^2 \lesssim 0.1$ of total optical power.

¹⁰The terminals sandwiching the EOM crystal is the capacitor.

Since PDH is an AC detection technique, the modulation frequency is essentially the sampling rate that sets the limit of the fastest frequency/phase noise detection bandwidth, so it should be high enough for fast servo purposes (As a rule of thumb, this should be 10 times faster than the servo bandwidth targeted at). It should also be high in order to avoid the predominating near-dc amplitude noise in the laser. Practically, a modulation frequency higher than 10 MHz is often chosen. In the conversion of the optical signal to an electrical signal, a low noise photodetector with wide bandwidth (larger the modulation frequency) is used. Since the cavity's drift is cancelled by fine tuning a PZT as we will discuss in the section for long time frequency stability, we use 100 μ W of optical power for a good SNR, which is higher than the typical power used in the experiments where the heating of dielectric coating of cavity mirror due to residual absorption of high intra-cavity field is a concern in an absolutely passively stabilized cavity. [44]

Often, the baseline of the PDH error signal is nonzero and time-dependent, which leads to error in the lock center. Theories and practice for minimizing this effect have been addressed in the literature in the context of residual amplitude modulation [43, 42]. In our experiment, several practices seem to be crucial. We use a linear polarizer with a high contrast ratio ($> 10^5$) and carefully align the direction of polarization of the input beam as well as the beam direction with respect to the EOM crystal during the optimization process. Frequency modulation is always preformed *after* a single-mode polarization maintaining fiber if there is one in the path. With these procedures, we can routinely reduce the residual error below 0.5% of total amplitude of error signal.

3.2.4 Building, mounting and testing of the cavity

The main parameters of an optical cavity are free spectral range (FSR) and linewidth $\delta\nu$. Basic relations are:¹¹

$$\text{FSR} = \frac{c}{2L}, \quad (3.8)$$

$$\delta\nu = \text{FSR}/F, \quad (3.9)$$

$$F \sim \pi\sqrt{R}/(1-R), \quad (3.10)$$

where L is spacing between cavity mirrors, and F the finesse of the cavity, which depends on the power reflectivity coefficients $R = R_1 = R_2$ of both mirrors. While small FSR is convenient for bridging the laser frequency and the cavity resonant frequency, as well as pushing the cavity linewidth narrower, a correspondingly longer cavity becomes mechanically less stable. Also, the cavity linewidth should match the free-running laser's linewidth in order to work in linear dispersion regime of the PDH error signal [43].

For the 1.8 kHz-wide 741 nm cooling transition, we desire a laser system with a linewidth of kHz or sub-kHz. The free-running commercial ECDL we use has a linewidth of 200 kHz measured within 5 μs , according to specifications. At the University of Illinois, we used a cavity for laser frequency stabilization with FSR = 1.4 GHz and a linewidth of 150 kHz. The cavity is built on a $3 \times 3 \times 10$ cm³ fused silica spacer. The spacer was bored out with an $r = 0.5$ cm central hole for the cavity mode. A plano-plano mirror and a plano-concave mirror (radius 25 cm) were glued to each side of the spacer to form a stable cavity. In order to compensate

¹¹For identical mirror coatings in high reflection limit, in vacuum.

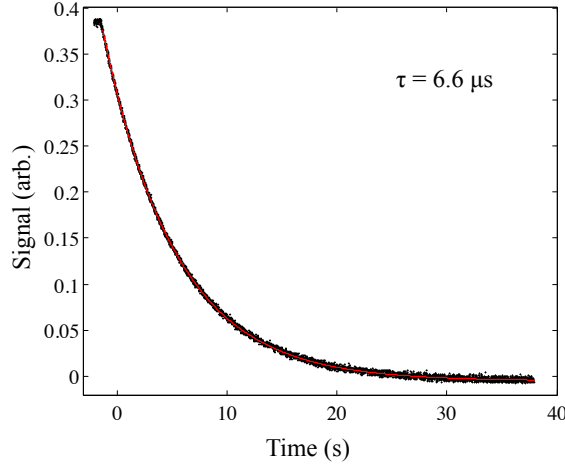


Figure 3.2: Cavity ring-down experiment of the 741 nm cavity. Transmitted light from the cavity is recorded and fitted to an exponential decay function. Fitted time constant is $6.6 \mu\text{s}$ which corresponds to a 24 kHz linewidth of the cavity.

for the inevitable thermal drift of the cavity,¹² a PZT ring was incorporated between the mirror and the spacer on one side for extremely fine length tuning. We chose not to actively servo the temperature of cavity, which normally requires challenging mK stability [44]. The cavity was simply mounted on sorbothane pads inside a small canister pumped to rough vacuum.¹³

When the Lev group moved to Stanford University, we revisited the old 741 nm laser system to see if the cavity was the bottleneck for improving laser coherence which will affect laser cooling efficiency. We built a reference cavity (for clarity called the monitor cavity) for out-of-loop noise measurement purposes. This cavity had the same geometry except for increased nominal mirror reflectivity of 99.995% (22 kHz linewidth for the cavity) for better sensitivity as a frequency discriminator.

¹²A relative length change of $\delta l/l = 1$ ppb leads to an absolute change of 400 kHz in the optical frequency at 741 nm.

¹³Outgassing was not a problem in at this vacuum level, as we found in experiments. Also the long time drift was always cancelled by PZT control. In a later upgrade, we used viton beads instead.

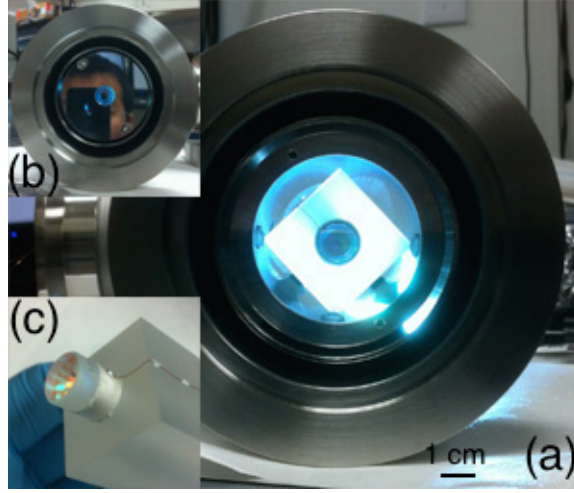


Figure 3.3: Picture of the optical cavity for narrow-line laser cooling. (a) is an overview of cavity once mounted in nested housing in vacuum. Inset (b) is a side view when the cavity is sealed inside the housing with a reflective cap. Inset (c) shows one side of cavity in free space.

Armed with the pre-stablized laser locked to the old cavity, we carried out cavity ring-down experiments [42] to determine the linewidth of new cavity (see Fig. 3.3). The measured value of 24 kHz is very close to the designed value.

We also took the chance to build a second new cavity (for short we will call it the new cavity from now on) identical to the monitor cavity. We now use it permanently for laser frequency stabilization after swapping out the old cavity from the vacuum canister. The new cavity is mounted in a nested housing inside the same vacuum canister for shielding purposes. Four viton beads support the cavity symmetrically from below, as shown in Fig. 3.4. The contacts on both sides are separated at the "Airy" separation which is 0.577 of the total length of the spacer for a reduced sensitivity to the vibrations [47]. Outermost is a box covered with thick plastic foam that covers the whole vacuum canister (Fig. 3.5).

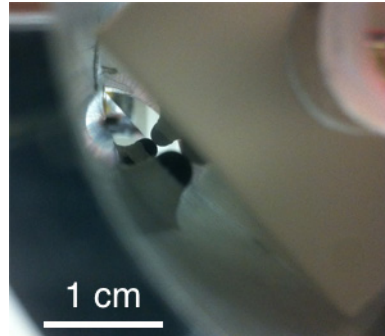


Figure 3.4: Mechanical support of 741 nm optical cavity: close sideview. The cavity is supported from below by small viton beads (black) which form point contacts at calculated positions.



Figure 3.5: Vacuum housing and outer acoustic shielding box for the 741 nm optical cavity. Cover of the shielding box is removed for the photograph.

3.2.5 Laser frequency feedback control

A free-running ECDL has a relatively wide frequency noise spectrum, and two "knobs" are often used for frequency tuning. Varying the voltage across the PZT that controls the external cavity's position can tune the laser frequency widely but slowly. Beyond one to a few kHz the PZT's response drops considerably and the phase changes drastically around mechanical resonances. Comparatively, varying diode current can tune the laser frequency very quickly, and the transfer function is almost a constant far beyond several kHz. Hence it is an important tool for fast noise control and laser linewidth narrowing.

To close the feedback loop, the PDH error signal is sent to a loop filter which modulates the PZT voltage and diode current of the laser. There is a considerable literature on detailed design principles for loop filter engineering in the context of PID (proportional-integral-derivative) control [41, 42, 48]. Therefore, only key ingredients that were important to achieving our tight lock are provided here.

For the purpose of suppressing noise, current branch gain should increase as $1/f$ below unity servo bandwidth, while maintaining the closed loop stability.¹⁴ It is effectively an integrator. To avoid fighting between the PZT control and current control near dc, the gain for the current servo should be limited and correspondingly the current loop filter becomes "P", while the dc lock center is determined by the PZT servo which has a higher gain near dc.

Between a few hundreds of kHz and MHz, where the transfer function of current modulation changes phase considerably or even flips sign because the physical mech-

¹⁴A more drastic transfer function of $1/f^2$ introduces undue phase delay, leading to lock instability.

anism for frequency tuning changes [46], extra phase-delay compensation is desirable. This is the reason to introduce a phase advance circuit working at high frequencies (which is essentially "D"). We found the unity gain servo bandwidth is boosted from about 200 – 300 kHz to above 1 MHz by introducing this part alone, before any other optimization.¹⁵ The op-amps used in the circuit should be fast enough to avoid any unwanted phase delay, as their performance becomes the limiting factor more and more as one pushes the servo speed limit.¹⁶ Further, the signal path (light path as well as electric path) is reduced as much as possible and the loop filter is physically installed next to the laser head. One must also be very careful to choose the correct low pass filter for extracting the PDH error signal after mixing the optical beats with the local oscillator (Fig. 3.1), as it may introduce considerable unwanted phase delay. With all these efforts, we were able to achieve 4 – 4.5 MHz unity gain servo bandwidth.¹⁷

3.2.6 Long-term frequency stability: transfer cavity scheme

The laser's linewidth is narrowed when it is locked to the cavity. But even for a well shielded cavity that is free from mechanical noise, thermal drift will eventually shift its resonant frequency. Heroic work on an ultrastable optical cavity (i.e., as

¹⁵Extending servo bandwidth does not only mean that the faster noise is suppressed, but also leads to higher gain in lower frequencies, as the loop filter's transfer function takes the form of $1/f$ in a wide frequency regime.

¹⁶Commercial fast analog loop filters, e.g., Toptica FALC110, are available these days, with only 45° phase delay at 10 MHz when set to be pure P (according to the specifications). One is being used in this experiment, for fast response and ease in tuning corner frequencies and gain limits.

¹⁷The intrinsic transfer function of the laser diode also influences the fastest servo possible. We used two types of diodes from different fabrications and with slightly different emission peak positions (745 nm and 730 nm) and empirically found a 1 MHz change in servo bandwidth when swapping between the two, even after fully optimizing the transfer function of the loop filter, while the performances among diodes in each category are much more similar.

in [44]) may reduce this drift rate enough to be acceptable within experimental data taking periods, but it is demanding technically. We decided to use an atomic reference to obtain long-term frequency stability.

Dy has a very high melting point of 1400°C , which makes conventional atomic vapor cell building too challenging. We desire a stand-alone frequency standard, so in-situ atomic spectroscopy on the Dy atomic beam in the UHV chamber for experiments [49, 18] was not considered. A Dy hollow cathode lamp is another convenient atomic reference source, but its severe collisional broadening in linewidth limits the lock frequency accuracy to $\sim 1\text{ MHz}$ [50], while Dy 741 nm transition is only 1.8 kHz wide. We decided to try sub-Doppler spectroscopy on the Rb D2 line as the frequency standard.

However this line is 40 nm away from the 741 nm narrow line. The transfer cavity technique allows us to bridge the two optical frequencies that may be hundreds of THz apart. By designing a proper dielectric coating for the cavity mirror, the cavity can be resonant for two injected laser beams with different colors. In this scheme one of the laser beams is pre-stabilized and the cavity itself can be locked to it via PDH detection and PZT tuning, while the other laser is locked to the stabilized cavity. In the end, the frequency stability is transferred from one laser to the other.

To shift the frequency of the locked laser in a wide range, we use a waveguide based fiber EOM to create sidebands GHz (comparable to FSR of cavity) away from the carrier, before sending it into the transfer cavity. We apply a PDH cavity lock (20-80 MHz frequency modulation) on one of the GHz sidebands, and by tuning the GHz rf frequency the carrier frequency is stably scanned based on the locked sideband

frequency. This is the scheme for all transfer cavity locks to bridge the targeted optical frequency and fixed cavity resonance frequency, as well as to scan the laser frequency while the lock is maintained.

3.3 Optical frequency reference: Rb 780 nm D2 line

Saturation absorption spectroscopy (SAS) [51] is a typical sub-Doppler spectroscopy that can provide sub-MHz frequency precision, and is widely used in cold atom experiments for laser frequency locks. The optical frequency standard for the Dy experiment is illustrated in Fig. 3.6. A 780 nm ECDL laser beam is partly injected into a 780 nm cavity for frequency stabilization (servo bandwidth 1 MHz) on short time scales. The pre-stabilized 780 nm laser beam is then used to probe the Rb cell for sub-Doppler spectroscopy to acquire an atomic frequency reference. The resultant lock error signal, which is generated through the frequency modulation technique, is properly filtered and sent to a high voltage amplifier that finely controls the PZT of the transfer cavity, cancelling out long term drift. Once this composite servo loop is complete, the rest of the laser beam is sent to a fiber array to distribute the optical frequency standard that can be used in transfer cavities for other laser systems. To best take advantage of existing optics, we designed the 780 nm cavity also as a transfer cavity that can accept an 842 nm laser beam for frequency locking of the Ti:Sapph laser.

The most essential figure of merit for this frequency standard is the long time stability. At the University of Illinois, we built the first version of this standard, using SAS, as illustrated in Fig. 3.7. Its frequency drift of ~ 50 kHz/h was too large

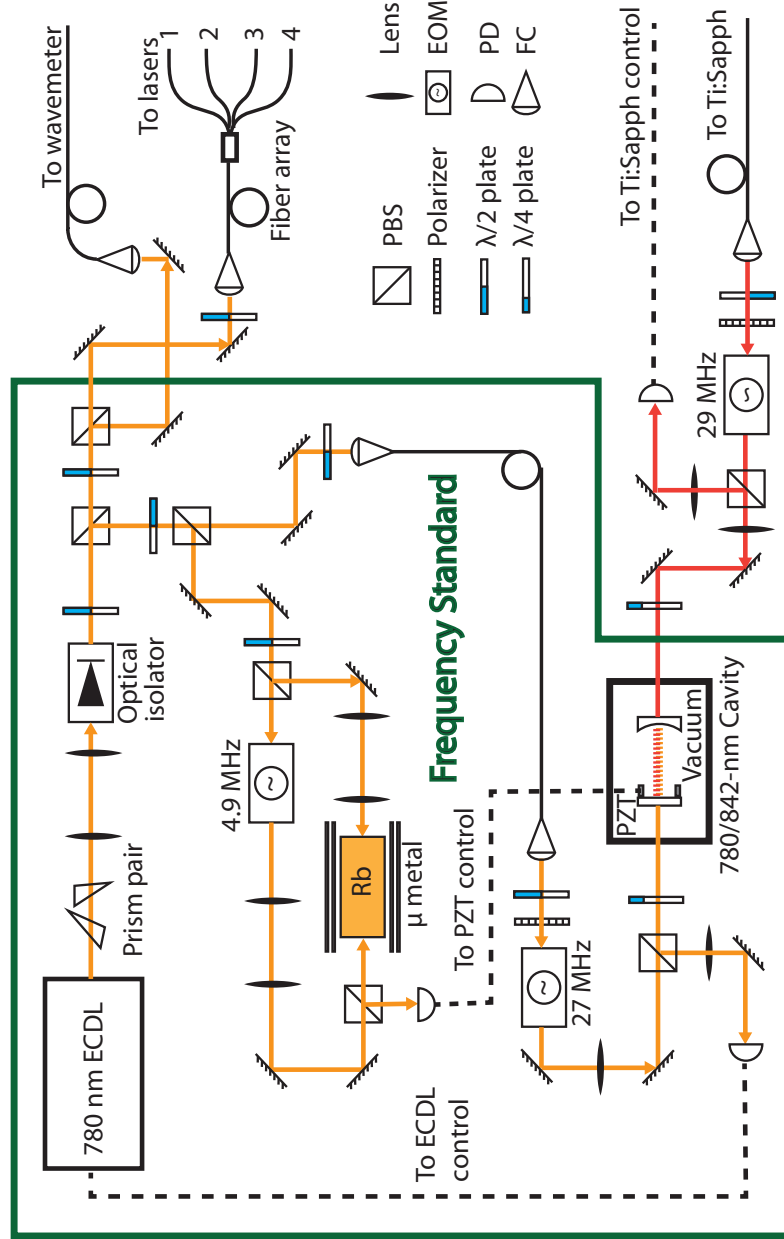


Figure 3.6: Optical setup for the 780 nm laser system. PBS: polarizing beam splitter; $\lambda/2$, $\lambda/4$ plates are retarders with specified retardance; PD: photo detector; FC: fiber coupler; AOM: acoustic-optic modulator; EOM: electric-optic modulator.

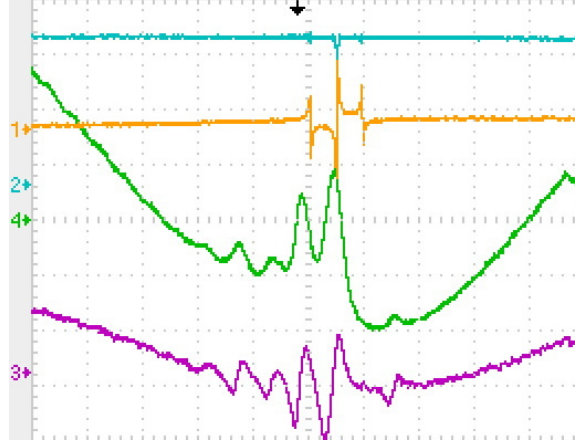


Figure 3.7: First version of the frequency standard: saturation absorption spectroscopy and PDH signals at 780 nm. Channel 1 (brown) and 2 (blue) are PDH related signals. Channel 3 (purple) and 4 (green) are saturation absorption spectrum and error signal for ^{85}Rb $F = 3$ hyperfine state respectively. The strongest cross-over peak of $F = 3 \rightarrow F' = 3$ and $F = 3 \rightarrow F' = 4$ was used for frequency lock.

for the experiment. The major systematic source was from SAS itself and details of its implementation. The error signal base line is intrinsically not flat and the zero crossing shifts up and down as the optical alignment changes. In order to originally achieve multiple purposes simultaneously, we used an AOM for frequency modulation in the SAS instead of EOM, which introduced noticeable residual amplitude modulation. On the cavity lock side, some details mentioned in the PDH optimization section were not yet introduced, and the shift of base line was clearly seen in Fig. 3.7.

At Stanford University, we upgraded via the major change to modulation transfer spectroscopy (MTS) [52, 53]. MTS is basically a four-wave mixing process that transfers the frequency modulation of a pumping beam to an unmodulated probe beam via a nonlinear atomic medium (Rb vapor). When the Doppler free resonance condition is not met, modulation is not transferred to the probe beam and therefore the baseline of the error signal is strictly zero. This is desirable for precision determination of the

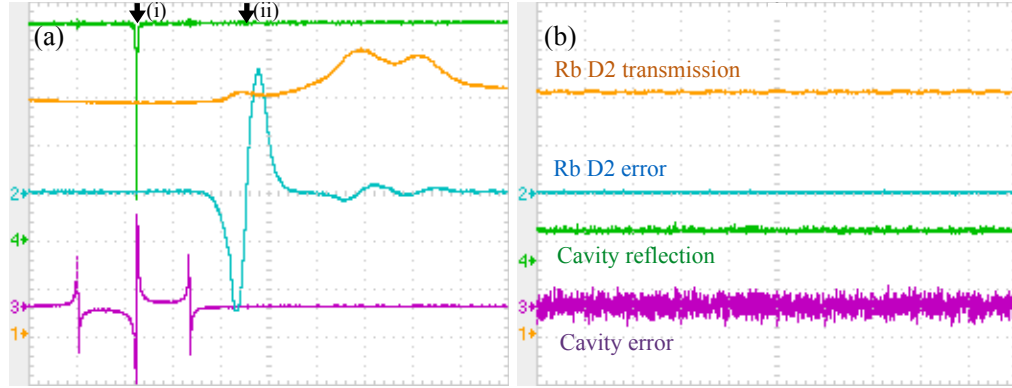


Figure 3.8: Second version of the frequency standard: modulation transfer spectroscopy and PDH signal at 780 nm. Taken from snapshots of oscilloscope triggered by laser frequency scanning signal, with arbitrary units. Channel 1 (brown, 1 V/div) is the modulation transfer spectroscopy of Rb 780 nm D2 line; channel 2 (blue, 500 mV/div) is the error signal; channel 3 (purple, 1 V/div) and channel 4 (green, 1 V/div) are the PDH error signal ($\times 10$ from the monitor port of our home-built loop filter) and reflection signal from cavity. (a) and (b) are for frequency scanning mode and locked mode respectively. Cavity resonance (i) and atomic resonance (ii) are offset for clarity. The strongest error peak for $F = 3 \rightarrow F' = 4$ cycling transition is used for lock. Ripples in channel 1 are the high index rf frequency modulation imprinted on the peak of atomic transition which is aliased when shown on a digital oscilloscope.

lock center.

To obtain the steepest error slope, different modulation frequencies and amplitudes were explored [54]. We used 4.9 MHz rf frequency (close to half of the slightly broadened linewidth of Rb) with an LC resonator for modulation depth enhancement.¹⁸ We also empirically adjusted the power of pump and probe beams for high SNR, with their power ratio close to unity in practice. A decent photodetector was chosen for low noise background and a low noise preamplifier was used.

We noticed that back reflection from the cavity may enter the ECDL which leads

¹⁸It is a nonlinear process, hence sensitive to the product of the powers in the carrier and in the sidebands.

to instability of the laser, even with 50 – 60 dB isolation. Optical isolation of 100 dB is now employed for the smallest optical feedback possible.

To guarantee lock center precision, we used an EOM instead of an AOM. The Rb vapor cell is carefully shielded from external magnetic field, using multiple layers of mu-metal, so that Zeeman shift is negligible. Saturation and DC drift of the rf double-balanced mixer was carefully monitored and minimized by using the optimum rf power level.

To fully take advantage of the high SNR error signal obtained,¹⁹ we employ a double integrator [48, 55] in the near-DC frequency regime to further suppress low frequency noise.

The frequency noise distribution for the cavity lock and the atomic lock are different. The atomic lock has better long time stability but deteriorates in the higher frequency range because of limited SNR and much lower slope of the error signal;²⁰ the cavity lock has better short time stability but poorer long time stability due to mechanical vibrations and thermal drift. We optimized overall performance of this composite loop servo by adjusting the relative gain ratios of the two locks so that the hand-over frequency f_0 is appropriate: below it the atomic lock dominates and above it the cavity lock dominates [56].

Assessment of the upgraded frequency standard was not trivial. There is no identical one to compare with and normal optical wave-meters are far from the precision required. However, the 741 nm narrow-line cooling experiment provides a natural

¹⁹In 2 kHz bandwidth, SNR exceeds 1500, mainly limited by the discrete 180 Hz power line harmonics.

²⁰The atomic natural linewidth is about 10 MHz and the cavity's linewidth in this setup is about 300 kHz.

sensitive tool to check the frequency. The observed frequency drift of the 741 nm laser system referenced to the 780 nm standard is 10 kHz/h on average. This provides the upper limit of long time instability of 780 nm standard. We shall discuss this in detail in the section on the 741 nm laser system.

In summary, we developed an optical frequency standard with < 10 kHz/h drift and 20 kHz fast linewidth;²¹ It indicates that we can determine the center position of the alkali D2 line to within 1‰.

3.4 Blue 421 nm laser

The Dy 421 nm broad transition is used for Zeeman slowing, transverse cooling, magneto-optical trapping and imaging. Due to the high saturation intensity of this transition, a high power laser is required, while requirements on the laser's linewidth are much less stringent so that only a fairly slow frequency lock is needed.

The laser source is provided by a commercial Ti:Sapphire laser (Coherent MBR 110) that is frequency doubled through nonlinear optics. The beta barium borate (BBO) crystal, mounted in a bow-tie type cavity (Coherent MBD) for intra-cavity power enhancement, delivers a 1.6-1.7 W 421 nm laser beam from 4.5 W 842 nm pumping beam. The Ti:Sapphire laser is pumped by an 18 W frequency doubled multi-mode solid state laser (Coherent G18) at 532 nm. The linewidth of 421 nm laser beam is < 50 kHz according to in-loop measurements of the Ti:Sapphire and doubling cavity error signals. A small fraction of 842 nm laser power is picked off for frequency control of the whole 421 nm laser system. Details are discussed for the rest

²¹Measured on 1 ms time scale.

of this section.

The majority of the 421 nm laser power is split into branches for various purposes (Fig. 3.9). The Zeeman slower beam is 650 MHz detuned from atomic resonance, using a custom-made high frequency AOM, with an efficiency of 65–70%. The AOM has a finite extinction ratio when used to turn laser beams on or off, therefore additional mechanical shutters are used for critical experiments where absolute darkness is important.

To co-trap different Dy isotopes, different frequencies of laser beams are needed. We either employ a two-laser system to address isotopes simultaneously (741 nm), or sweep the laser frequency in a controlled way for sequential loading of isotopes. The latter is used for the 421 nm laser system, considering the cost and space of a Ti:Sapphire laser system.²²

Among Dy isotopes, the largest isotope shift is between ^{164}Dy and ^{161}Dy at ~ 2200 MHz [57], and at least a 1.1 GHz frequency shift is needed at 842 nm before frequency doubling. By designing the 780/842 nm transfer cavity with an FSR of 3.3 GHz, the carrier can scan 1.1 GHz with one sideband locked while the other sideband scans 2.2 GHz without hitting any cavity resonances (see Fig. 3.10).²³ In practice, it was found that one cavity resonance (already fixed by 780 nm D2 line in frequency absolutely) was in just the wrong position for allowing continuous carrier scanning between Dy atomic resonances, therefore a double-passed AOM was added to shift this continuous

²²The narrow-line MOT cannot run in a sequential loading scheme because after shifting the MOT frequency for loading the next isotope, the earlier loaded ultra-cold and dense cloud would undergo inelastic collisions rapidly as well as suffer from Majorana loss in our quadrupole magnetic trap. The 421 nm MOT, being much hotter and more sparse, can be safely held in the quadrupole magnetic trap in darkness for hundreds of ms.

²³In practice, any double resonance from carrier and sidebands throw the laser out of lock.

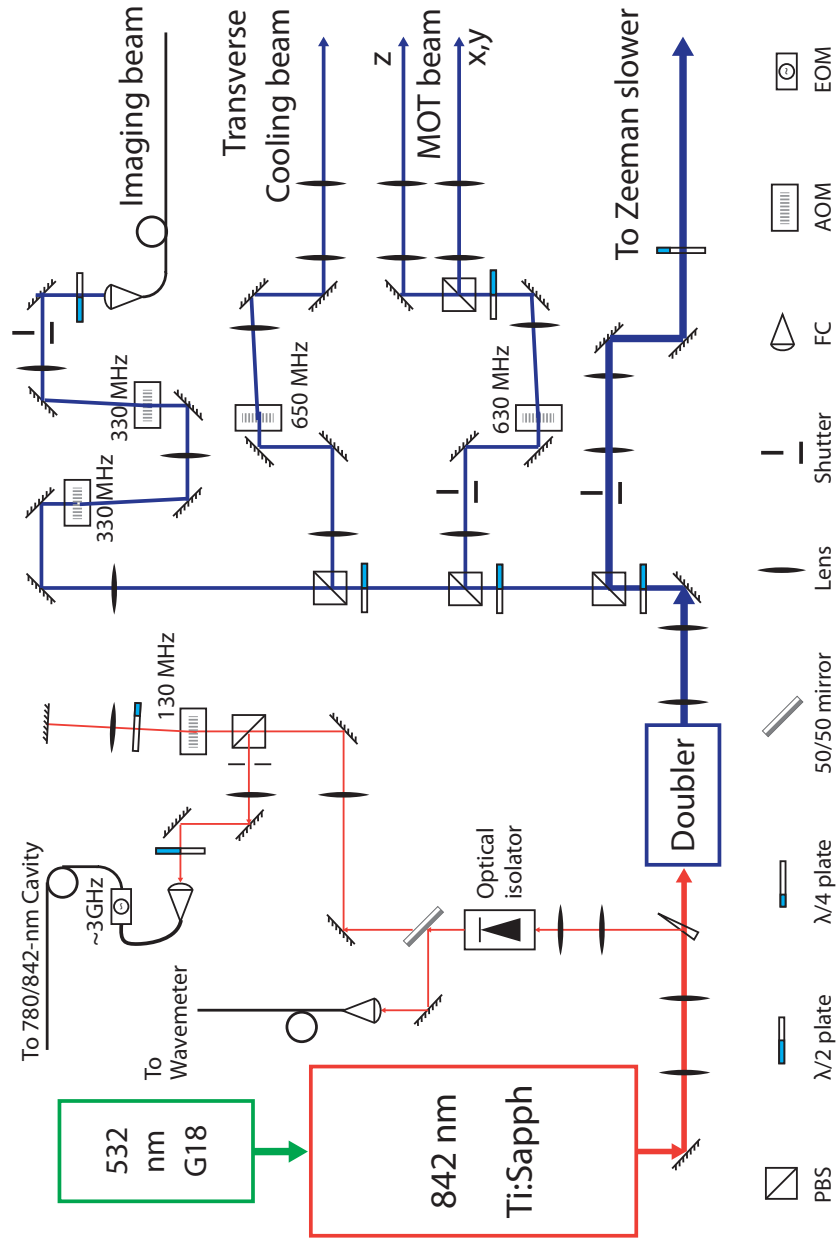


Figure 3.9: Optical setup for the 421 nm laser system. PBS: polarizing beam splitter; $\lambda/2$, $\lambda/4$ wave-plates are with specified retardance; FC: fiber coupler; AOM: acoustic-optic modulator; EOM: electric-optic modulator.

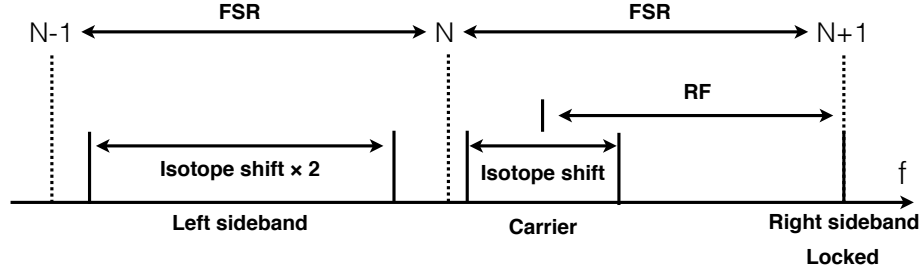


Figure 3.10: Frequency lock scheme for the 842 nm laser frequency scanning. Dashed vertical lines are different orders of cavity resonances separated by FSR in frequency; solid lines are laser frequency components of carrier and two sidebands.

frequency scan range by 260 MHz so as to avoid the offending cavity resonance.

The fastest continuous frequency scan achievable is about 1GHz/100 ms with the Ti:Sapphire laser locked. This is limited by the highest scan speed of input allowed by the processor that regulates the internal reference cavity of the Ti:Sapphire for frequency stabilization. This speed is fast enough for hopping between two frequencies for co-trapping any pair of Dy isotopes before the first isotope suffers considerable loss in the magnetic trap.

3.5 Red 741 nm laser

The 741 nm laser system is a master and slave dual-laser system. The master laser is frequency locked to the 741/780 nm transfer cavity, and the slave laser is phase locked to the master laser with an offset frequency in GHz regime (Fig. 3.11). The two lasers (18 mW output each) are used for co-trapping different isotopes simultaneously, and most of their outputs are coupled into an optical fiber array that distributes MOT

beams into the vacuum chamber on another optical table. The added phase noise from properly positioned optical fibers (10 m long) were expected to be negligible for the 1.8 kHz transition linewidth [58], therefore we do not actively cancel out phase noise in optical fibers as carried out in optical clock experiments [59].

For the 741 nm lasers, frequency stabilization is the central challenge. After all the efforts to narrow the laser linewidth that have been described in earlier sections, we want to know how coherent the lasers are once locked. One direct way is to measure beat signals of two identical but independently locked lasers, but we do not have another independently locked laser to check against. Delayed self-heterodyne measurement is suitable when a single laser is available [60], but to introduce enough time delay comparable to the coherence time of a 1 kHz laser, the optical fiber delay line is impractically long. Since we are focused on quantum simulation with cold atoms, accurate linewidth measurement is not important. We decided to use an independent monitor cavity as the frequency discriminator to estimate the linewidth of 741 nm locked laser.²⁴ Our goal is to know whether 741 nm narrow-line cooling is limited by our laser's coherence or not.

The measurement result for the master laser locked to the old 741 nm cavity (150 kHz linewidth) is shown in Fig. 3.12. Frequency noise power density is obtained from Fourier transformation of the PDH error signal recorded on the independent monitor cavity with a linewidth of $\delta\nu = 24$ kHz (section 1.1.5), corrected with a multiplicative factor of $1 + (2f/\delta\nu)^2$ based on cavity's transfer function [41]. Qualitatively, frequency noise at medium to high frequencies (high 100 kHz) is effectively suppressed

²⁴We would rather perform an out-of-loop measurement instead of an easier in-loop measurement, since the former reflects the performance in an objective way.

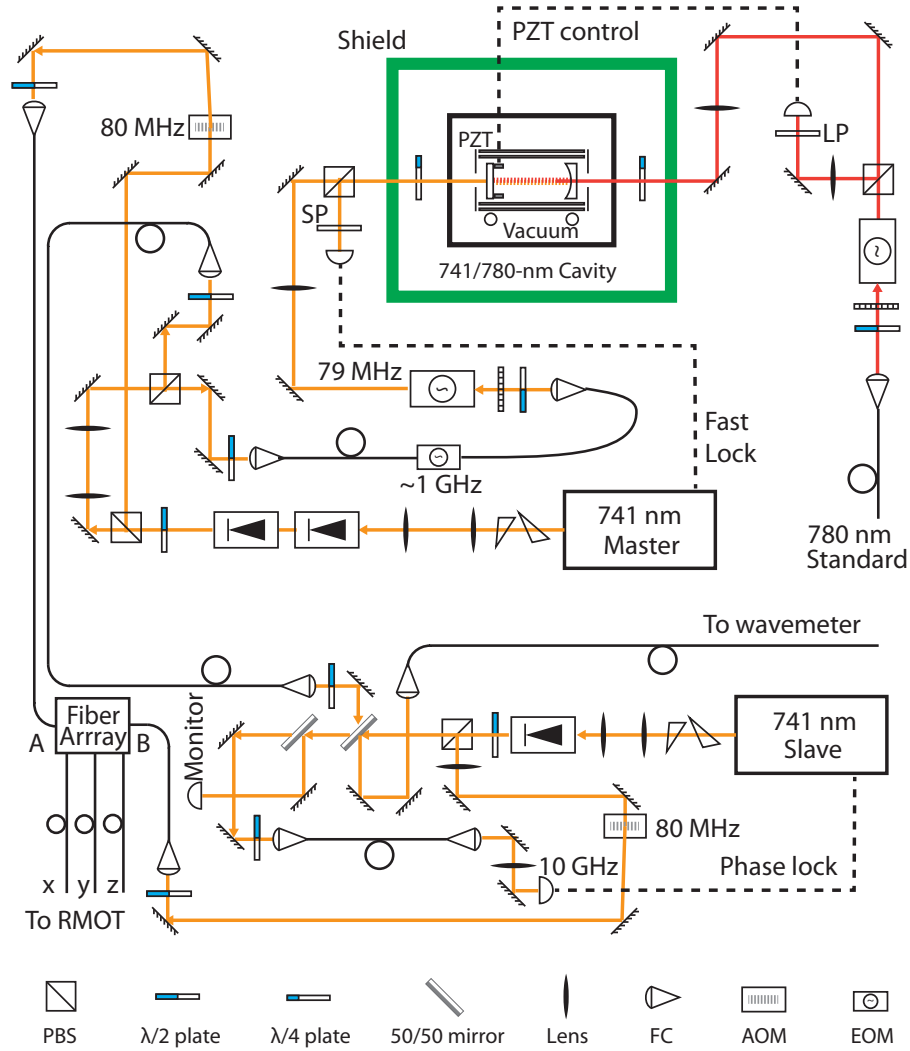


Figure 3.11: Optical setup for the 741 nm laser system. PBS: polarizing beam splitter; $\lambda/2$, $\lambda/4$ wave-plates are with specified retardance; FC: fiber coupler; AOM: acoustic-optic modulator; EOM: electro-optic modulator. RMOT: 741 nm red MOT. 10 GHz PIN detector is Electro-Optics Technology ET-4000.

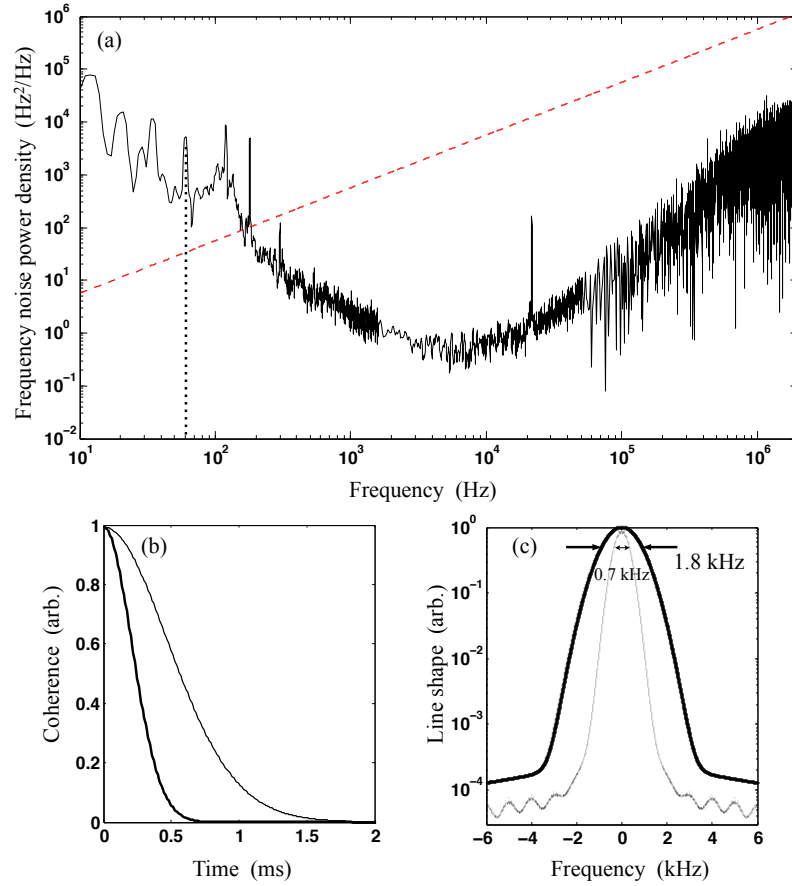


Figure 3.12: Linewidth determination of 741 nm master laser. (a) measured frequency noise power spectrum. Red dashed line is β line (see text); black dotted line is at 60 Hz. (b) Calculated coherence term in eqn. 3.4 based on (a). Thick line and thin line are results based on noise spectrum including and excluding noise below 60 Hz respectively. (c) Calculated line shapes based on (b) respectively. Wiggles on the tails are artifacts due to discrete sampling frequency spacing in measurements.

in Fig. 3.12, while the limiting factor for laser linewidth is the low frequency noise below about 150 Hz where the power density spectrum meets the line separating low/high modulation index regimes ($8\ln(2)f/\pi^2$ or the so-called β line in [35]). This is consistent with the sharp differences in the derived linewidths of 1.8 kHz and 0.7 kHz when including and excluding frequency noise below 60 Hz, derived from measured noise power spectrum. Note that the Gaussian line shape obtained is another indication of technical noise dominating at low frequencies.

Similar results were obtained from locking the laser to the new 741-cavity (24 kHz linewidth) with improved mounting in the vacuum canister (section 1.1.5). Based on this, we tend to believe that the measured linewidth most probably reflects the stability of the monitor cavity itself, which was mounted in air without decent vibrational or acoustic isolation.²⁵ Therefore, we believe the locked 741 nm laser has a linewidth lower than the 1.8 kHz natural linewidth of 741 nm transition, and perhaps is already in the sub-kHz regime, measured on a 10 ms time scale.

In the 741 nm MOT experiment, sidebands with MHz-wide distribution are created for efficient loading, and frequency scanning capability is desired in order to collect atoms at different positions (see the chapter for 741 nm MOT). These functions are realized by an 80-MHz AOM that controls the laser beam before it is sent to the MOT (Fig. 3.11).

²⁵Another likely "suspect" is that the voltage noise from the PZT driver that modulates the 741/780 nm transfer cavity. With the known voltage-length conversion coefficient of the PZT, the corresponding rms voltage noise is 0.6 mV for 2-kHz wide frequency jittering, which is well above the rms value of voltage noise measured from the output of our high voltage amplifier. We further tested the performance by replacing the high voltage amplifier with batteries, which showed no noticeable change. Lastly, the finite linewidth of 780 nm standard may bring extra frequency jitter within the 741/780 nm transfer cavity's PZT servo bandwidth, but our detailed calculation based on the information of our frequency standard's noise spectrum indicates that this is not the limiting factor.

As we shall see in Chapter 6, the position of the 741 nm MOT is linearly dependent on the frequency detuning. We use this feature to estimate the frequency drift of the 741 nm laser. The typical observed drift rate is random and about 10 kHz/h on average. Contributions may come from the drift of the 780 nm standard, the lock center drift of the 741/780 transfer cavity, or drift in the voltage controlled oscillator (VCO) frequency that drives the AOM. In particular, the VCO has a few kHz/h drift rate, which may be improved by a phase locked loop in the future, but it seems not be the limiting factor at the moment. This measurement also offers an upper limit on the largest drift rate possible of our 780 nm standard, assuming all drifts are statistically random.

The slave laser needs a similar coherence as the master laser. A phase offset lock [61] was employed by comparing the optical beat signal between the master and slave lasers with a stable rf local oscillator that sets the offset frequency (Fig. 3.11). The mixed signal of the two is properly filtered and serves as the error signal for fast frequency/phase control, as in the PDH cavity lock. The error signal for phase comparison from a mixer is sinusoidal as a function of the relative phase and a loose lock may lead to phase slips as the system hops between different zero crossings as the different lock centers. A fast analog loop filter, similar to the one used for the fast frequency servo of the master laser with a few MHz-wide servo bandwidth, keeps the slave laser locked at the same lock point for many hours. Figure 3.13 shows the locked beat signal both in the frequency and time domains. As servo gain increases, close-in phase noise is increasingly suppressed and more power collapses onto the carrier. The locked rms relative phase error between the master laser and slave laser, drops below

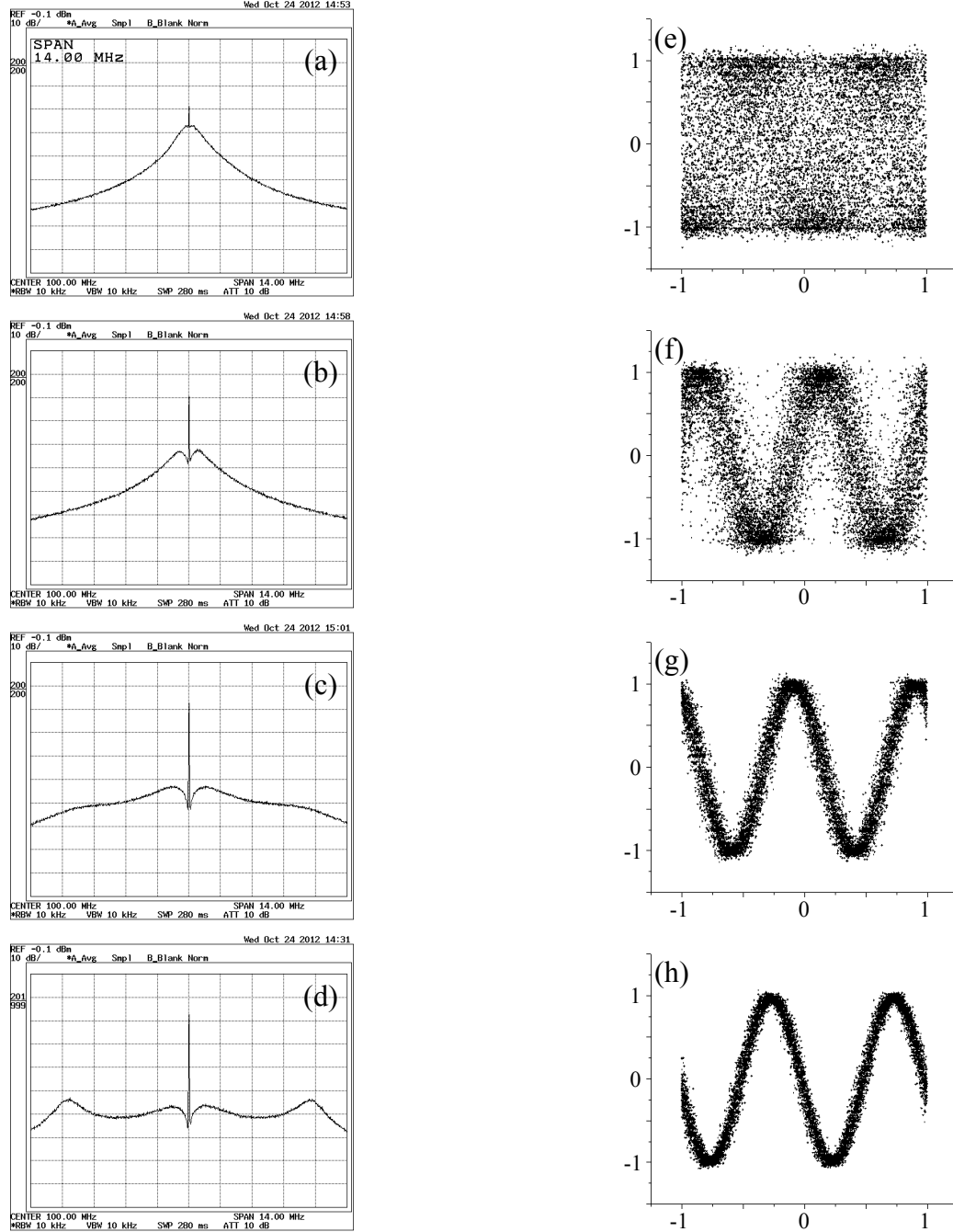


Figure 3.13: Phase lock out-of-loop measurement between the 741 nm master and slave lasers. (a)-(d) measured optical beat signals, resolution bandwidth is 10 kHz, frequency range is 14 MHz, and sweep time 200 ms, 200 average. Vertical scale is 10 dB/division. (e)-(h) corresponding time domain laser beat signals mixed down to Hz level. x-axis unit is second.

1 rad (a crucial turning point) roughly at a servo bandwidth of $\sim 200\text{-}250$ kHz (see Fig. 3.13 (b) and (f)). We demonstrated a minimum rms value of 0.14 rad in relative phase error.²⁶ with a 5.8 MHz servo bandwidth when everything was fully optimized. Therefore, we conclude that the slave laser has almost identical coherence properties to the master laser, for the purpose of 741 nm narrow-line cooling.

3.6 NIR 1064 nm laser

We use 1064 nm fiber lasers for optical dipole force trapping (ODT) of Dy atoms. Since the dynamic polarizability is several times smaller than that for alkali atoms at the same wavelength, high power is needed for tight trapping. In this laser system, a single mode 1064 nm fiber laser with sub-10 kHz linewidth is used as a seed laser. Its output is distributed to 50 W 1064 nm fiber amplifiers. To avoid interferences between different beams in crossed ODTs, the laser frequencies are shifted by AOMs at ± 80 MHz, which also serve as optical shutters. We noticed that thermal effects in the AOM crystal make the laser beams unrepeatable during on/off cycles. The AOM's quartz crystal is actively water cooled to minimize this effect.

The intensity of the ODT beam is servoed, as the noise effectively modulates the trapping frequency and hence parametrically heats trapped atoms [62]. The typical servo bandwidth in our experiment is 10 – 20 kHz, and the lifetime of the trapped cloud is found not to be limited by the intensity noise of the 1064 nm laser beams.

²⁶Measured in ± 5 MHz bandwidth, a rms phase error of $\Delta\phi = 0.14$ rad indicates that a fraction of $\eta = \exp(-\Delta\phi^2) = 98.1\%$ out of the total power is in the carrier [61]. The rms phase error is 0.26 rad when measured in a ± 10 MHz bandwidth and starts to increase fairly slowly as measurement bandwidth increases where we reached the white noise floor.

3.7 Radio frequency generation and control

The Dy experiment needs many rf frequencies in several frequency bands. RF excitation between Zeeman states is typically between 1 to 10s of MHz. Agile frequency control and scan shape engineering, as well as kHz resolution and stability are needed. Direct digital synthesizing (DDS) is used for this purpose. From 80 – 600 MHz, where laser frequencies needs to be shifted constantly, VCOs with a stable DC voltage reference are applied. In the hundreds of MHz to GHz regime, where precision and agility of rf frequency are both desired, i.e., in our phase offset lock, or sideband tuning in our transfer cavity lock, we employ phase locked loops (PLL).

3.7.1 Frequency standard

The RF standard in the experiment is provided by a stable 10 MHz source. This is given by a Rb clock (SRS PRS10)²⁷ which has low phase noise (< -130 dBc/Hz at 10 Hz) and an Allan variance of 2×10^{-11} at 1 s. Beyond 10^4 s the Allan variance starts to rise. As recommended by the manufacturer, we plan to use the GPS signal to control the long time drift of the Rb clock (internal PLL is provided). At all relevant time scales, the relative frequency stability of this GPS-servoed 10 MHz source is predicated to be better than 5×10^{-12} , which is far more than good enough for our current Dy experiments.²⁸

²⁷All the technical parameters in this section were provided from commercial specifications and we did not perform independent tests.

²⁸The stable clock signal is split into well isolated multi-channels via a distribution amplifier (SRS FS730), which are sent to different experiments in parallel in Lev Lab. The PRS10 is locked to the 1-pulse-per-second (PPS) output of a GlobalSat ET-332 GPS board, as the author tested independently in office. The ET-332 is connected to an "ANT-555" antenna that is mounted by office windows during the test. Such a GPS servo has not been implemented in lab yet.

3.7.2 DDS and Phase-Locked Loop

DDS is realized with an evaluation board from analog devices. Basic frequency generation, frequency hopping or phase continuous linear frequency scanning are the most frequent purposes. With the help of external single-board microprocessors like BeagleBone, arbitrary frequency generation is also possible.

In the GHz regime, we generate rf via integer PLL. We obtain continuous scanning with fixed frequency division/multiplication settings, by scanning the reference frequency for PLL which we provide by DDS. The output of the PLL can be tuned continuously over a GHz-wide range, as long as it is within the VCO's output. This is how we prepare the agile rf signal for 1.1 GHz wide scanning for the 421 nm laser system. The wide band Mini-circuits ZX95-2770A+ VCO together with the Analog Devices ADF4108 PLL evaluation board were used in this application. Recent development of ultrafast DDS in the GHz regime²⁹ could replace the PLL in this application eventually.

²⁹Analog Devices AD9106 with maximum output of 1.4GHz.

Chapter 4

Experimental Detection

4.1 Introduction

In this chapter, details of experimental detection of cold atoms are discussed. We begin with the features of the 421-nm imaging transition. Particularly fluorescence imaging and absorption imaging system are addressed. Besides the information of the atom distribution, knowing the distribution in internal states of the atoms is also important. Stern-Gerlach (SG) experiment setup is built and tested for this purpose. Lastly, the discussion of principle and implementation of adiabatic rapid passage completes the topic of manipulation of spin states both for preparation and detection.

4.2 Imaging transition at 421 nm

The 421 nm blue transition of Dy is used for imaging ultracold Dy atoms. This strong transition, with a broad 32 MHz linewidth [16], can drive fast photon scattering for imaging purpose. A broad transition also has a relatively low sensitivity to laser detuning and Zeeman shifts, which brings experimental convenience. Its short wavelength, which is almost half of that of the Rb 780 nm D2 line, decreases the theoretical diffraction limit of imaging by almost a factor of 2. Lastly, its resonant photon-scattering cross section $\sigma = 3\lambda^2/2\pi$ is therefore also almost four times smaller.

Both the ground state and excited states of the 421 nm transition have high spin multiplicities, as illustrated in Fig. 4.1. For Dy atoms in "the stretched" states ($m = \pm 8$ or $m = \pm 21/2$ for bosonic isotopes or fermionic isotopes), the transition strengths for σ^+ , π , σ^- transitions have the extremely unbalanced ratios of 1 : 0.11 : 0.0065 for bosons, and 1 : 0.087 : 0.004 for fermions. Full calculations for all m states are represented in Fig. 4.1.¹ This leads to spin sensitivity in absorption imaging. In comparison, π light transition maintains bulk part of transition strengths for most of spin states.

4.3 Imaging system

The imaging system consists of a telescope and a CCD camera with associated input and output signal channels connected to the control computer. The imaging axis is perpendicular to the axial direction of MOT anti-Helmholtz coils. For absorption

¹As one can see in sub-figure (c-d), Dy is approaching the high spin distribution limit.

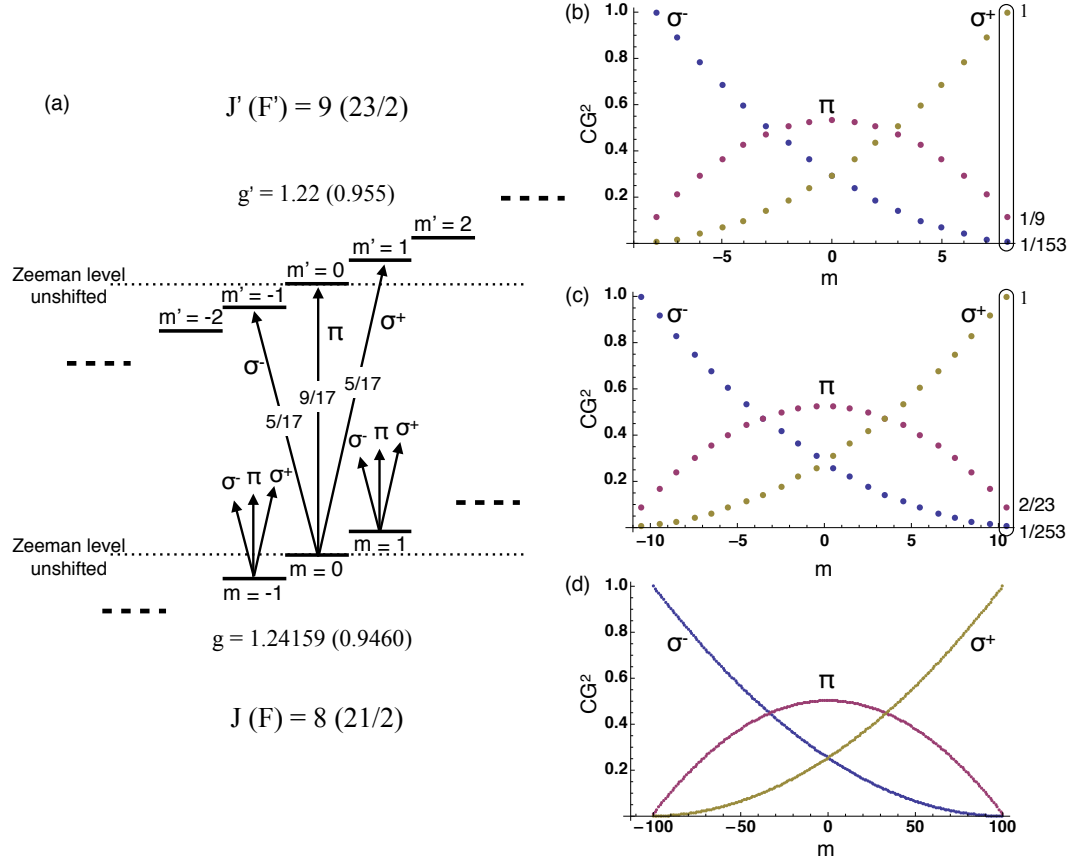


Figure 4.1: (a) Zeeman sublevel structure of Dy for ground state and 421 nm excited state in external magnetic field. J and F are total angular momenta in ground state for bosonic isotopes and fermionic isotopes respectively. m ($|m| \leq J(F)$) is the z component of angular momenta along quantization direction. g the Landé g factors for boson (fermion). σ^+ , π , σ^- denote transitions linking states with $\Delta m = m' - m = 1, 0, -1$, together with corresponding transition strengths (square of Clebsh-Gordon coefficients (CG)). (b),(c),(d) are the transition strengths (CG^2) for each individual m Zeeman levels in ground state undergoing σ^+ , π , σ^- transitions. Particularly, (a) is for boson ($|m| \leq J = 8$), (b) is for fermion ($|m| \leq F = 21/2$), and (c) is limiting form calculated at high spin ($|m| \leq J = 100$). The transition strengths for stretch states ($m = 8, 21/2$) are highlighted.

imaging, an additional a pair of quantization coils is used to generate locally homogeneous magnetic field to define the polarization of the imaging light. Otherwise, both fluorescence and absorption share the same imaging optics and camera.

We currently use a Point Grey Research Dragonfly2-13S2 (with Sony ICX445 sensor) as the camera. Its quantum efficiency is above 40% at 421 nm according to the specification, and the pixel size is $3.75 \mu m$. Its digital output has an effective depth of 12 bit, indicating a theoretical contrast limit of $2^{12} \sim 4000$ for each pixel. In practice, it is reduced by dark counts and readout noise.² We are preparing to switch to Princeton Instruments PIXIS: 1024BR CCD, for lower dark counts and other enhanced functionality.

The imaging telescope is made of a pair of 2" achromatic doublets. The focal length of front lens is f_1 is 25 cm, which is limited by the size of vacuum chamber, and the focal length of the rear lens is f_2 can be either 15 cm or 40 cm, providing lower and higher magnifications ($M = f_2/f_1$). The exact magnification is calibrated from fitting the free fall trajectory of ultracold atoms in situ. We noticed that the imaging resolution is within a factor of two of the diffraction limit ($\sim 3 \mu m$), with some residual imaging aberrations.

4.3.1 Fluorescence imaging

During fluorescence imaging, the near resonant light from the MOT beams are scattered by cold atoms and collected by the imaging system. It is a dark-field measurement, in the sense that the background is dark and the atoms are bright.

²The imaging system is not photon shot noise limited.

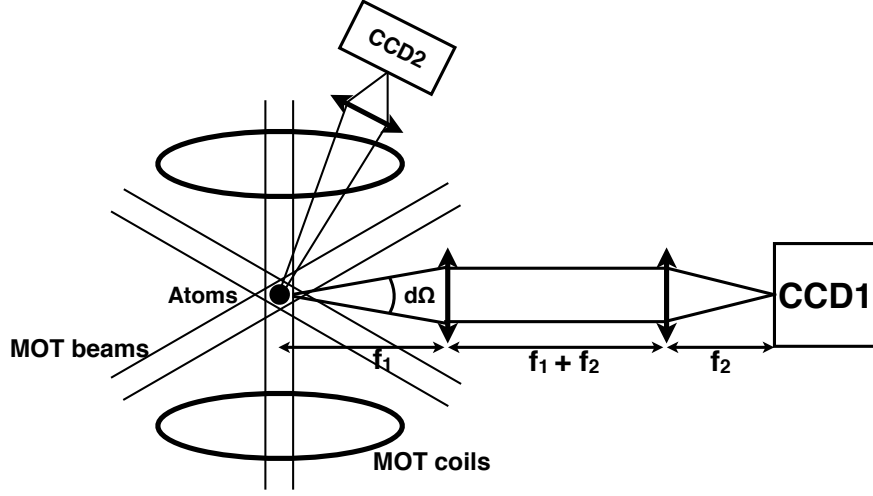


Figure 4.2: Illustration of fluorescence imaging. f_1 and f_2 are focal lengths of front and rear lenses. CCD1 is the major camera for data analysis and CCD2 is for additional diagnostics. The illustration is not to scale.

From the accumulated photons in the imaging region, the atomic density distribution and total atom number are deduced.

Atomic fluorescence depends on the laser intensity I , polarization and frequency detuning Γ externally, and population distribution in spin states of atoms internally. A detailed treatment involves multi-state coupled optical Bloch equations [63]. We note that an empirical functional form can simplify the treatment with reasonable accuracy:

$$\Gamma_{\text{ph}} = g(I/I_s^*, \Delta, \Gamma) N_{\text{atom}} = \left\{ \frac{\Gamma}{2} \frac{I/I_s^*}{1 + 2I/I_s^* + 4(\Delta/\Gamma)^2} \right\} N_{\text{atom}} \quad (4.1)$$

where Γ_{ph} is the photon scattering rate, N_{atom} is atom number, Γ the transition linewidth, Δ laser detuning from resonance, and I_s^* is a constant with a similar role to the saturation intensity I_s . By varying the experimental parameters I and Δ , we found that the functional form of g is a reasonably good fit [17, 27]. Physically, it

can be understood because under the illumination of 6 counter-propagating MOT beams from $\pm x, \pm y, \pm z$ directions with roughly equal intensity and opposing light polarizations, the atom is effectively being excited by isotropic, unpolarized light. The effective transition strength is averaged among $\sigma^\pm, \pi$ excitations and weakened compared to that of a two-level system. With this assumption, we find $I_s^* = \beta I_s$ where the correction factor of $\beta = 2.7$ (see appendix) and I_s is the saturation intensity. For the Dy 421 nm transition, $I_s = 58 \text{ mW cm}^{-2}$.

Considering the fraction of photons collected by the optics ($\frac{d\Omega}{4\pi}$ within solid angle $d\Omega$, assuming emission is isotropic), the transmission efficiency throughout vacuum chamber window and imaging lenses f , and the CCD's quantum efficiency η , the photoelectron generation rate is $\Gamma_e = \eta f \frac{d\Omega}{4\pi} \Gamma_{\text{ph}}$. By reading the signal size integrated within an exposure time $S = \alpha_T \Gamma_e = (\alpha_T \eta) f \frac{d\Omega}{4\pi} \Gamma_{\text{ph}}$, where α_T is a constant for given CCD settings within exposure time T , we could deduce the photon flux rate and hence the atom number. In our experiment, the coefficient $\alpha_T \eta$ is independently calibrated by a light source with known flux intensity.

Due to the complexity of the calibrations and the approximation made, fluorescence imaging can not give a high accuracy measurement of atom number. As a cross check, calibration-free absorption imaging that drives Dy's cycling transition between the $m = 8 \rightarrow m' = 9$ stretched states indicates agreement with fluorescence imaging within a factor of 2, in terms of atom number. On the other hand, fluorescence imaging does not require turning on of a quantization field and is insensitive to spin state. It is useful in some applications when absorption imaging is not appropriate.

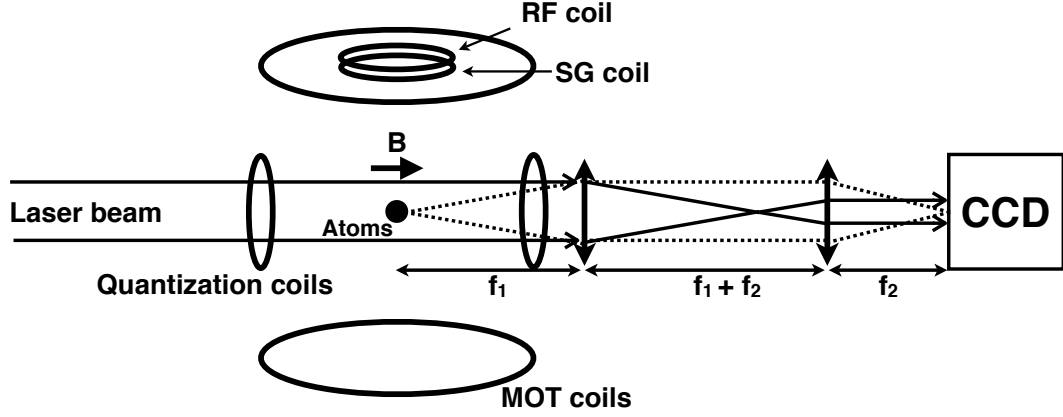


Figure 4.3: Illustration of absorption imaging. f_1 and f_2 are focal lengths of front and rear lenses. Solid lines denote light paths and dashed lines are shadow paths. The illustration is not to scale.

4.3.2 Absorption imaging

Absorption imaging is based on the resonant absorption of light through cold atoms which cast a shadow that is to be imaged. In this sense, it is bright-field imaging. The absorption rate is described by the Beer-Lambert law [64]: $dI/dz = -N\sigma I$, where N is the 3D spatial density of atoms, σ is the interaction cross-section depending on frequency detuning, polarization of light and spin state of atoms, in the low light intensity limit. Optical density (OD) due to absorption is the result of integrated absorption along the viewing direction z , which leads to a 2D column density distribution.

In practice, each absorption imaging consists of three shots: one taken with the imaging light and atoms ($I_{A,ph}(x, y)$), one with only the imaging light $I_{ph}(x, y)$, and one taken with no light and atoms ($I_0(x, y)$). The last one is for calibrating back-

ground noise. The measured OD is extracted as:

$$OD(x, y) = -\ln \frac{I_{A,ph}(x, y) - I_0(x, y)}{I_{ph}(x, y) - I_0(x, y)} \quad (4.2)$$

Given a known cross-section σ , the 2D density of atoms $n(x, y) = OD(x, y)/\sigma$ is obtainable. In this process, the magnification of the imaging optics should be carefully accounted in to faithfully reflect the density in object space.

In the experiment, we used reasonably pure polarized imaging light and a well defined quantization field (Fig. 4.3) to selectively drive certain transitions among spin states. The imaging light is on resonance.³ With 4 mW of power ($I/I_{sat} = 0.02$) for imaging, it is very far from saturation. Under these conditions, the cross-section σ depends only on spin states. For circularly polarized $\sigma^\pm$ light and a stretched spin state $m = \pm 8$ ($\pm 21/2$) (or cycling transition), the atom is simply equivalent to a two-level system $\sigma = 3\lambda^2/2\pi$. For other spin states, the corresponding cross-section should be compensated by the ratio of the transition strengths that are proportional to square of CG coefficients [65, 66] (see Fig. 4.1). The value is highly sensitive to the spin state and light polarization. For example, the atoms in the stretched state $m = 8$ show the highest absorption possible for σ^+ light and are essentially transparent to σ^- light because the CG coefficient (0.0065) is essentially zero.

The physical picture during imaging is slightly more complicated than we have depicted. The atoms are being optically pumped among spin states during imaging, and absorption is evolving along with the spin population distribution. For example, atoms initially in the $m = -8$ state imaged by the "wrong" probe light are pumped towards higher m value spin states. The evolution of population in spin states are

³For cloud with high OD, we sometimes choose to detune the frequency so as to reduce absorption, paying the price of distortion of cloud's shape due to lensing effect out of dispersion.

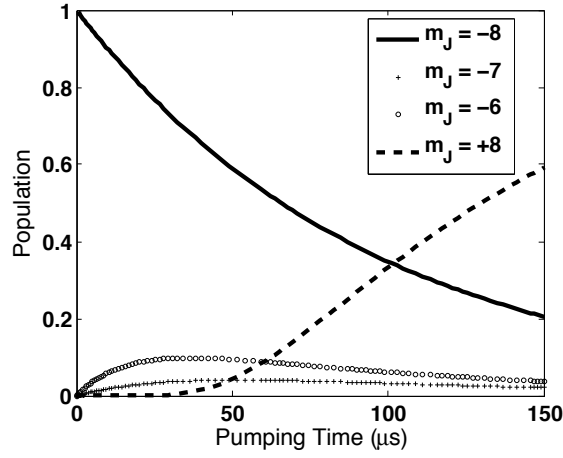


Figure 4.4: Zeeman state optical pumping via the 421 nm transition. Initial state is stretched state $m = -8$. Atoms are optically pumped into higher m_J Zeeman states under σ^+ light. For clarity only populations of a few lowest m_J spin states and other opposite stretched state $m = 8$ are shown. Simulation is based on normal experimental conditions: optical pumping intensity $I/I_{sat} = 0.02$ and magnetic field 3 G with optical frequency on resonance with $m = 8 \rightarrow m' = 9$ transition (cycling transition for σ^+ light).

simulated in Fig. 4.4. By tracking the population evolution, one can calculate the effective cross-section for each spin state, based on averaging the absorption of all resultant spin state components during exposure time, as in Fig. 4.5. In our experiment, exposure time is usually within 20 - 60 μs , which implies spin states with large negative m values are essentially dark states. We flip the sign of polarization of the imaging light to image these states more effectively. To simultaneously see all spin states we use fluorescence imaging (absolute value calibrated from absorption imaging for stretched state) or use π light for absorption imaging.

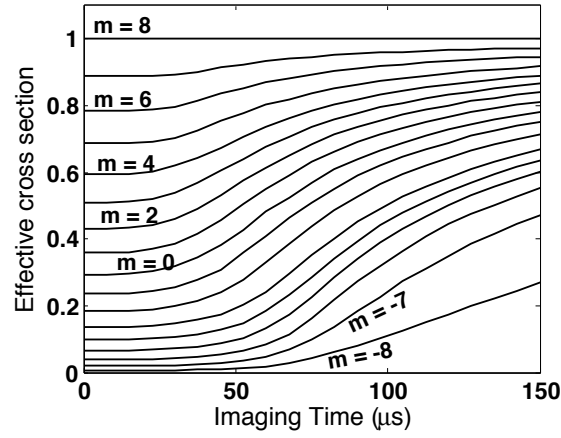


Figure 4.5: Effective cross sections of the 421 nm resonant σ^+ absorption imaging for different spin states. Curves represent for each spin state ($m = +8, +7, \dots, -7, -8$ from top to bottom) the normalized effective cross section averaged over finite imaging time, based on the time evolving population of each spin state under optical pumping. Unity stands for the absorption with full transition strength. The effective cross sections of all spin states asymptotically approach unity which is equal to that of $m = 8$ state, due to σ^+ pumping effect towards this stretched state. Calculation is based on conditions depicted in Fig. 4.4.

4.4 Spin manipulation

4.4.1 Detection: Stern-Gerlach experiment

For a spin mixture ensemble, Stern-Gerlach type experiments [67] are important tool to topographically study the diagonal populations of spin states. The key ingredient lies in the difference of the magnetic moment of each spin state, which leads to a differential "tidal" force in a magnetic field gradient that separates the cloud into spin components.

Separating atoms with different spin states is a competition between free thermal expansion of thermal atoms and a magnetic force kick in a certain direction. For ultracold Dy atoms at low hundreds nK, a constant field gradient of 20 G/cm is sufficient to separate atoms after 10-ms time of flight. A higher field gradient is desirable when probing atoms in the μK regime within shorter times of flight. Therefore we targeted at turning on a 50 G/cm field gradient within 1 ms in the design. The gradient coil is placed 6 cm away from the cloud, which is limited by our vacuum chamber size. We decided to take advantage of the pulsed discharging current from a large capacitor bank. It is much cheaper than an expensive high power DC power supply and active water cooling is not required due to the low duty cycle.

For this purpose, we built a near critically damped LCR circuit with a half period of a few ms, as in Fig. 4.6. The capacitor bank⁴ provides a total capacitance of 0.18 C, the gradient coil (21 rounds with a mean diameter of 5.2 cm) is determined to have an inductance of 30 μH , and the whole circuit is optimized to bring the total

⁴EPCOS B41456-B8229-M 22000 μF , 63V is particularly suitable for its low equivalent series resistance and high ripple current limit.

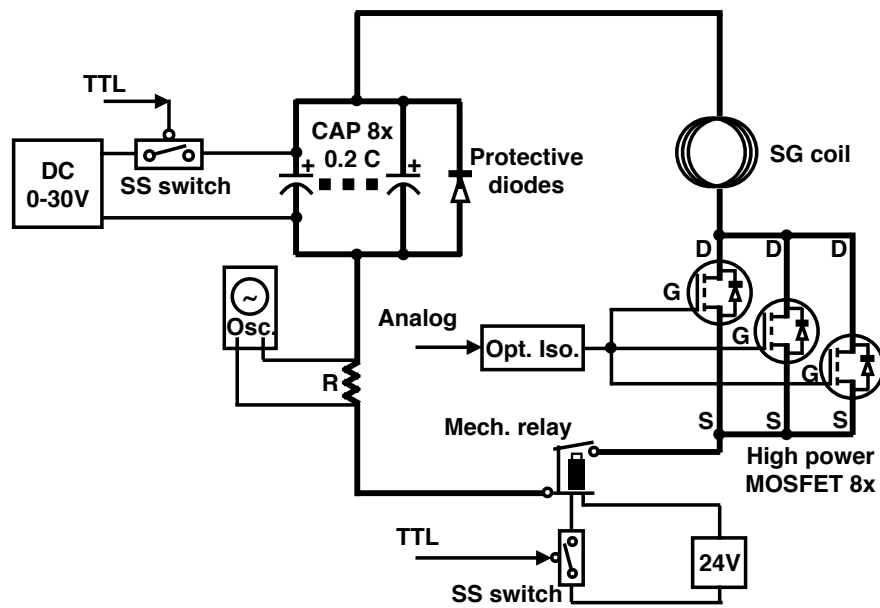


Figure 4.6: Circuit for Stern-Gerlach experiment. Thin wires represent for weak signal or low current paths, and thick wires represent high current paths. Opt. Iso. is the analog optical isolator. SS switch: solid state switch; Mech. relay: mechanical relay; CAP: capacitor bank with 0.2 C total capacitance. G,S,D are gate, source drain terminals of MOSFETs. Osc.: oscilloscope, measures the voltage drop on $R = 1 \text{ m}\Omega$ to monitor current.

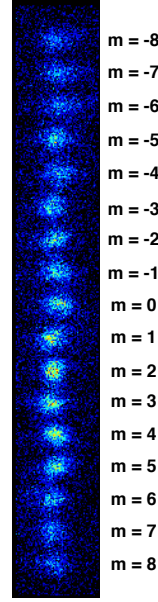


Figure 4.7: Time of flight imaging of ^{162}Dy spin mixture of 17 spin states in Stern-Gerlach experiment. Each spin component is separated and imaged after 14 ms time of flight in a field gradient with the peak value of 18 G cm^{-1} . Brighter color indicates higher density. The spin mixture are prepared by quickly moving magnetic field null point through the spin-polarized Dy cloud. To image each spin state with roughly equal sensitivity, fluorescence imaging is employed instead of absorption imaging. Field of view is about $6 \times 0.8 \text{ mm}$.

resistance down to $30 \text{ m}\Omega$. High power MOSFETs⁵ in parallel are used to pinch the current at controllable holding times. The unipolar capacitors are protected by diodes. Currents are monitored by a non-inductive resistor R ($1 \text{ m}\Omega$) with a high power rating.⁶ Both turn-on speed and peak current are controlled by charge voltage. The highest peak current tested in open space is 650 A .⁷

By design, this SG setup provides a field gradient around the cloud of 12 G/cm/100 A

⁵IXYS: IXFN280N085, N-channel enhanced mode.

⁶OHMITE: TGHGCR0010FE, 1% tolerance, 100W.

⁷It is highly recommended to remove the coil from the vacuum chamber for high current test, to avoid any unnecessary magnetization of ferromagnetic materials nearby. Recall that Dy is highly magnetic.

and usually currents below 200 A are enough for experiments with ultracold bosonic or fermionic Dy isotopes. Figure 4.7 shows a typical TOF image of a SG experiment, for bosonic isotope ^{162}Dy , after release from the optical dipole trap. From the separation of the cloud, we deduced the local field gradient at the Dy atoms, and checked it against theoretical calculation. The measured value is slightly lower than expected, which might be due to counter-rotating eddy currents in the vacuum chamber.

Due to our geometric constraints, the RF coil and SG coil are closely packed together in the Dy machine (Fig. 4.3), and we noticed the coupling between the two reduces the RF field generated by the former. It is a bit surprising that it occurs even when the SG circuit is open by setting the gate voltage of the MOSFETs to zero. One possible explanation could be that the MOSFETs are effectively capacitive at rf frequencies. An ad hoc solution is to physically disconnect the circuit loop during RF excitations by introducing a fast, high-current-grade mechanical relay.⁸ The price is the added complexity of the circuit and a possible dead time of 3-5 ms due to the relay's response time.

4.4.2 RF adiabatic rapid passage

It is often desirable to prepare atoms in targeted spin states starting from one spin state. For example, the 741 nm MOT prepares spin-purified ultracold atoms in $m = +8$ ($+21/2$) states for bosonic (fermionic) isotopes, but the absolute ground states $m = -8$ ($-21/2$) are more suitable for evaporative cooling.

Flipping the spin vector in the Bloch sphere by sending $\pi/2$ or π pulses can

⁸Tyco Electronics, V23071-A1010-A132.

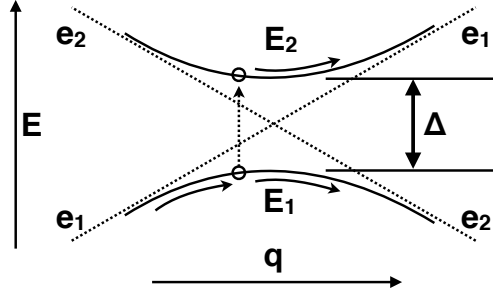


Figure 4.8: Illustration of adiabatic rapid passage. Dashed lines are eigenvalues of Hamiltonian without interaction term V and solid lines are eigenvalues of full Hamiltonian. See text for details.

transfer atoms to different spin states and are widely used in AMO physics [68]. Another powerful tool to transfer the spin state is to use the adiabatic rapid passage (ARP) technique. It is robust and insensitive to technical noise. The details of this technique are described in the literature [69, 70, 71, 72].

In a two-level Hamiltonian, the eigenenergies of two eigenstates e_1, e_2 form an avoided crossing if there is an interaction V that couples the two states, as in Fig. 4.8. As some tuning parameter q in the Hamiltonian varies, the eigenvalues of the new eigenstates E_1, E_2 approach each other at the V -dependent smallest energy gap value Δ , where e_1, e_2 mix maximumly. Away from this resonance, $E_{1/2}$ asymptotically approach the limit form of $e_{1/2}$ both in eigenvalues and Hilbert space. By starting with the prepared state e_1 and adiabatically scanning the q value from one side of the far detuned regime to the other, one follows a trajectory of $e_1 = \lim_{q \rightarrow -\infty} E_1(q) \rightarrow E_1(q) \rightarrow e_2 = \lim_{q \rightarrow +\infty} E_1(q)$ which swaps the eigenstate. This is insensitive to the tuning precision of q and amplitude noise in eigenvalues of E_1 as long as the gap Δ is large enough. Very fast scanning of q can lead to loss to the E_2 state.

The picture above is general, and one can apply the model in the RF regime by mapping $e_i \rightarrow m_i$ (spin states) $q \rightarrow \omega$ (or B field as they are on equal footing in rotating frame), $V \rightarrow \Omega$ (Rabi frequency). The model can also be extended to handle a multi-level system, as in the case of an ARP from $m = +8 \rightarrow m = -8$.

In our experiment several points are worth mentioning. The magnetic dipole transition's selection rule requires that rf magnetic field to have nonzero normal component in the quantization direction, as is easily seen from the interaction term $V \propto J_x B_z = \frac{1}{2}(J_+ + J_-)B_z$. The rf frequency scanning speed $\dot{\omega}$ needs to adiabatically slow so as to lower the Zener-Landau tunneling probability P:

$$P = 1 - \exp(-2\pi \frac{\Omega^2}{\dot{\omega}}) \quad (4.3)$$

For Dy, there are inelastic collisions between atoms in different spin states. Therefore the frequency sweep time should be smaller than the mean inelastic collisional time [72]. On our Dy experimental machine, a scanning range of ~ 100 kHz within 6 – 10 ms transfers atoms from $m = +8(21/2)$ state to $m = -8(21/2)$ with as close to unity fidelity as we can measure within the SNR in spin-sensitive absorption imaging as well as in a Stern-Gerlach experiment. The result is consistent with the Rabi frequency value that was independently estimated. The RF frequency sweep is linear in time and the Rabi frequency is turned on and off exponentially and held constant around the center of the sweep range. Also, the center of scanning range is magnetic field dependent, and we use it as local field probe.

For fermionic isotopes, the quadratic Zeeman shifts in a high magnetic field lift the degeneracy of resonance frequencies of adjacent spin transfers. This provides us with

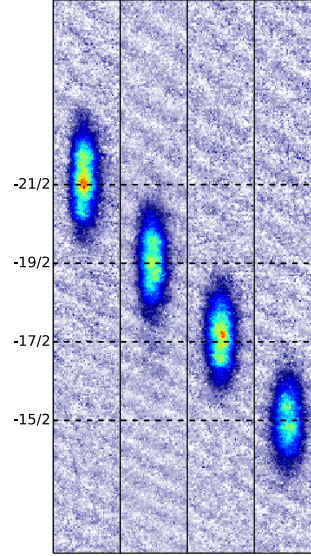


Figure 4.9: Fluorescence images of spin polarized ^{161}Dy in high m_F spin states. Spin polarized clouds with different $m_F = -21/2, -19/2, -17/2, -15/2$ are imaged at fixed TOF in each experimental run (each column). They are separated by SG coil mounted vertically top with different displacements due to varying sizes of the magnetic moment along z direction. Five sample average for each imaging.

the degree of freedom for arbitrary spin state transfer, and Fig. 4.9 illustrates the pure spin state preparation of $m_F = -21/2$ (ground state), $-19/2, -17/2, -15/2$ via ARP in a 30 G magnetic field. Preparation of spin mixture has also been implemented.

Chapter 5

The First Magneto-Optical Trap of Dysprosium

5.1 Cooling and trapping dysprosium: a historical review

Laser cooling and trapping neutral atoms plays a crucial role in the procedure for Bose-Einstein condensation of dilute atomic gases. It is certainly no coincidence that the first experimental BEC of a neutral gas was realized not very long after laser cooling and trapping techniques became available for neutral atoms even though BEC was conceived more than half a century beforehand and observed in superfluid helium long before.

Laser cooling techniques are not without limitations. Scattering at least 10^5 photons is required before typical atoms are cooled from room temperature to the ultra-

cold regime. The ideal case of a true two level system for a cycling transition is rare, and so if there are other states in addition to the lower state of the simplified two level system that atoms in the excited state can decay to, and the probability of such a decay is considerable (branching ratio $> 10^{-5}$), the photon scattering will stop once the atoms are lost in those states (i.e. dark states). In practice, another laser beam, a so-called repumper, is used to pump atoms out of the dark states and back into the quasi-closed two-level system. Repumping becomes impractical if there are too many dark states. In fact, except for a few exceptions [73, 74], successful laser cooling of molecules remains a challenge due to countless rovibrational states within the electronic ground state manifold. For atoms, alkali and alkaline earth dilute gases set the paradigms of laser cooling because of their relatively simple optical transitions. The presence of multiple valence electrons makes finding an effective two-level system exceedingly hard for most other atoms.

Besides laser cooling, cryogenic buffer gas cooling is another route for preparing cold atoms. Atoms that lack suitable laser cooling transitions may be trapped in a magnetic trap and sympathetically cooled by cryogenic helium buffer gas, provided they have a magnetic dipole moment. Indeed, quite a few species of dilute atomic gases [75] were trapped in this way, including Dy. However, due to Dy's bad elastic-to-inelastic collisional ratio at cryogenic helium temperatures (< 50 , [76]), evaporative cooling ceased to lower temperatures below 50 mK, which remains deeply in the thermal regime for typical dilute gases.

In 2006, McClelland's group at NIST reported the repumperless magneto-optical trapping (MOT) of the lanthanide (rare earth) erbium [18]. It was quite unexpected,

considering the many possible dark states that could stop laser cooling. As will be explained in this chapter, the low branching ratio to the dark states from the cooling excited state and the highly magnetic nature of those dark states provide an effective recycling mechanism for the dark state atoms back to Er MOT. While there was no indication that this effect would work for the other lanthanide atoms such as Dy due to the fortuitous nature of these low breaching ratios and favorable collisional properties, we were sufficiently encouraged to take the risk of trying a similar procedure with Dy, an atom slightly more electronically complicated yet more magnetic.

5.2 Features of dysprosium's optical transitions

The Dy atom, atomic number 66, has an electronic ground state configuration $[\text{Xe}]4f^{10}6s^2$. The ten identical f -electrons and two s -electrons form many terms from the different couplings, and the ground term has total orbital angular momentum $L = 6$ and total spin $S = 2$, within the Russell-Saunders (LS) coupling scheme, and the lowest level within this multiplet is $^5\text{I}_8$ as the ground state.¹ As the ground state Landé g factor is 1.24159 [77], Dy has a large magnetic dipole moment of 9.93 Bohr magnetons in the ground state. In fact it is the most magnetic stable atom.²

The atomic spectrum of Dy I in visible and NIR regimes is very complex (Fig. 5.1).

¹In LS coupling scheme, $L = 6$ and $S = 2$ would couple together forming a multiplet of $J = 4, 5, 6, 7, 8$ and Hund's rule predicts the level $J = 8$ with highest angular momentum possible is the ground state. It turns out that for the ground state and low excited states of Dy and other lanthanides, LS coupling is still a good approximation even though electronic spin-orbit coupling is large in such heavy metals. An indication of the validity of LS coupling is that the ratios of energy spacing among the levels in the multiplet $^5\text{I}_{J=4,5,6,7,8}$ fulfill the the interval rules predicted by LS coupling [64].

²Terbium has a comparably large magnetic moment, within 0.5% of Dy's, but it has only one isotope, ^{159}Tb , which is a boson and has a low-lying black-body coupled state which is only 9 THz above the ground state.

This follows from several reasons. The 12 valence electrons can be excited to many different atomic subshells with low excitation energies, because the $4f$, $5d$, $6s$, $6p$ orbitals have small energy differences, as is quite typical in lanthanides [78]. The most common types of transitions are $4f^{10}6s^2 \rightarrow 4f^95d6s^2$ (mainly for low excited states), $4f^{10}6s6p$ (medium to high excited states), $4f^95d^26s^1$ (mainly for high excited states). For each configuration, valence electrons couple themselves together with a huge number of possible levels associated with different L, S, J . With such a large quantity of levels, the number of optical transitions grows quadratically (mathematically equivalent to the handshake problem). Even after decades of spectroscopic work, there was work reported recently on classifications of some spectral lines and new levels of Dy [79].

Natural Dy has seven stable isotopes (see Table 5.1).³ There are five bosonic isotopes (with nuclear spin $I = 0$) as well as two fermionic isotopes ($I = \frac{5}{2}$, see Fig. 5.2 for the hyperfine structure). This facilitates the study of quantum degenerate gases of both flavors in spin statistics as well as Bose-Bose, Fermi-Fermi and Bose-Fermi mixtures. The large number of isotopes also provides us with a greater chance of finding isotopes with favorable ultracold collisional properties. Finally, the generous natural abundances of the major isotopes eliminate the need to use expensive isotope-enriched samples for laser cooling and trapping [80].

The strongest optical transition of Dy is the 421-nm transition between the even parity ground state and the odd parity excited state $4f^{10}(^5I_8)6s6p(^1P^o_1)(8, 1)^o_9$ with

³We note that all lanthanides with even proton number (Ce, Nd, Sm, Gd, Dy, Er, Yb) have far more naturally abundant stable isotopes than ones with odd proton number (La, Pr, Pm, Eu, Tb, Ho, Tm, Lu).

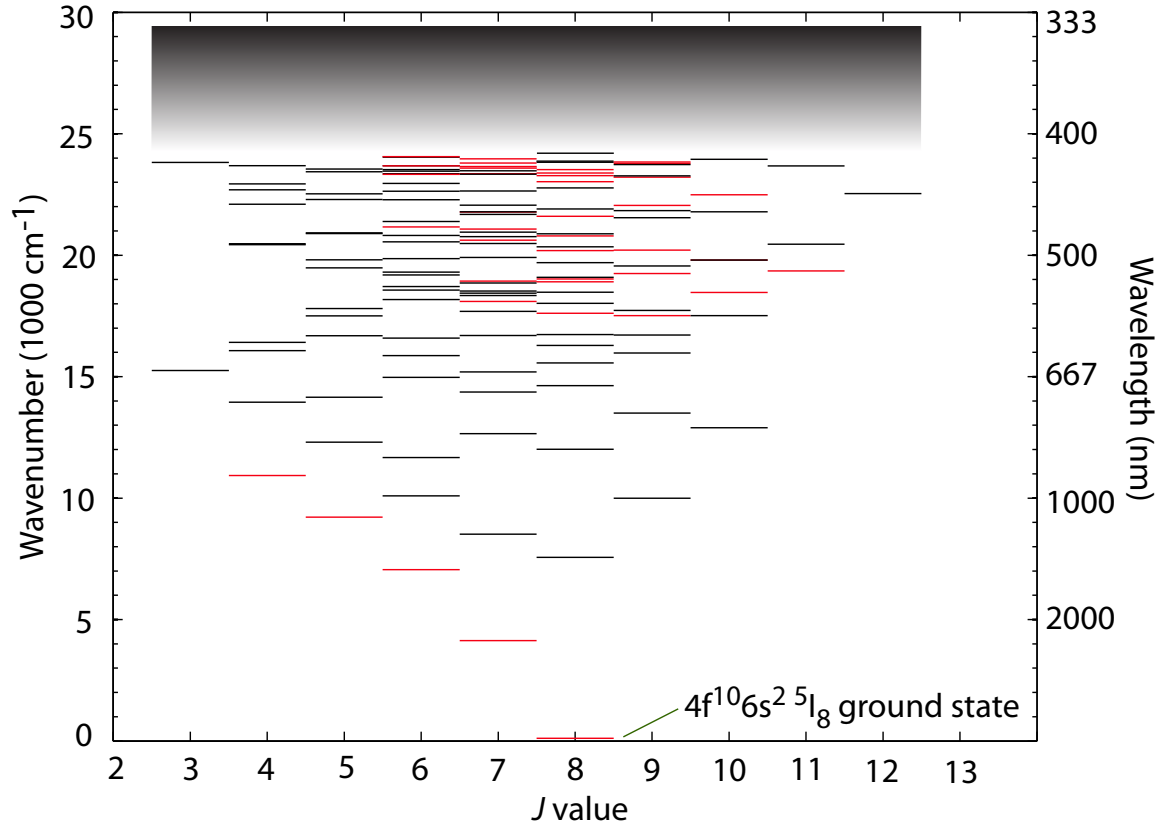


Figure 5.1: Energy level structure of Dy. States are grouped according to total electron angular momentum J . Red and black colors denotes even and odd parities respectively. Levels above 24000 cm^{-1} that are less relevant to our work are replaced by a grey block where shading indicates high density of levels. Data from [77].

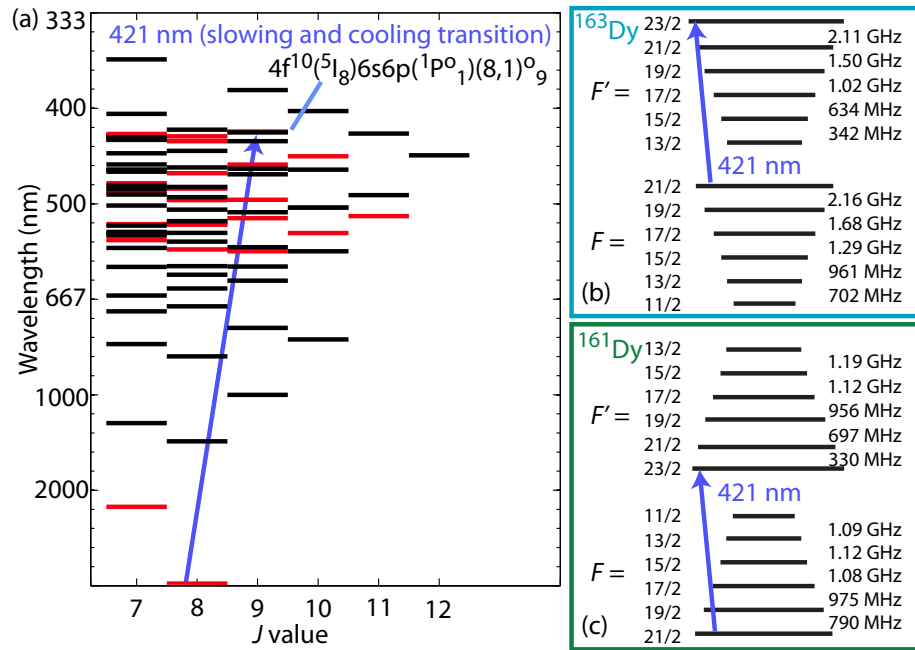


Figure 5.2: Energy levels of Dy relating to 421 nm transition. (a) Dy energy levels [77]. MOT and Zeeman slower employ the strongest laser cooling transition (421 nm) between the even parity (red) ground state and the odd (black) excited state $4f^{10}(^5I_8)6s6p(^1P^o_1)(8,1)^o_9$. (b),(c) Fermion hyperfine structure (not to scale). F, F' are total angular momenta (including nuclear spin) for ground and excited states. Bosonic isotopes have no hyperfine structure.

Table 5.1: Isotopes of Dy.

Isotopes	Abundance ¹	Nuclear spin	Statistics
¹⁶⁴ Dy	0.283	0	boson
¹⁶³ Dy	0.249	5/2	fermion
¹⁶² Dy	0.255	0	boson
¹⁶¹ Dy	0.189	5/2	fermion
¹⁶⁰ Dy	0.023	0	boson
¹⁵⁸ Dy	9.5×10^{-4}	0	boson
¹⁵⁶ Dy	5.6×10^{-4}	0	boson

¹Data from ref. [81].

total electron angular momentum $J' = 9$.⁴ Its natural linewidth is 32.2(8) MHz, which we measured in Ref. [16] and is consistent with that measured in Ref. [77]. This transition is much wider compared to the D2 lines of alkali atoms but similar to the blue-wavelength lines of alkaline earth atoms. This $J = 8 \rightarrow J' = 9$ transition for Dy bosonic isotopes ($F = 21/2 \rightarrow F' = 23/2$ for fermions) can form a quasi-cycling transition for laser cooling, provided the branching ratio from the 421-nm excited state to the low lying states is sufficiently small. We chose this strong transition for transverse cooling, Zeeman slowing, magneto-optical trapping, and imaging.

⁴This belongs the $4f^{10}6s^2 \rightarrow 4f^{10}6s6p$ type of transition which possess most of strong optical lines of Dy I. In this state, 4f electrons are coupled together with total angular momentum $j_1 = 8$, while 6s and 6p electrons are coupled together with total angular momentum $j_2 = 1$. Finally the total angular momentum is formed from JJ coupling ($J' = j_1 + j_2 = 8 + 1$). This coupling scheme is typical for medium to high excited states.

5.3 Repumperless 421 nm MOT

The Dy cooling and trapping apparatus consists of a high temperature oven, transverse cooling stage, Zeeman slower, and MOT chamber. Details of the design can be found in Chapter 2 and [27, 34]. The atomic beam passes through a differential pumping tube before intersecting four 100-mW, 2 cm-long transverse cooling beams detuned -0.2Γ from the 421-nm transition. Transverse cooling enhances the optimized MOT population by a factor of 3–4. The atomic beam then passes through a spin-flip Zeeman slower [34, 69] operating at -24Γ . The power of the Zeeman slowing laser is ~ 1 W; we find a linear increase in MOT population until ~ 1 W. Atoms in the slowed beam are captured by a 6-beam MOT with detuning $\delta = -\Gamma$, 1.1-cm beam waists, and total intensity $I = 36$ mW/cm². A 35 G/cm magnetic quadrupole gradient $\nabla_z B \approx 2\nabla_x B$ produces the maximum MOT population. The 421-nm lasers are derived from a frequency doubled Ti:Sapphire laser that is transfer-cavity locked to a Rb-stabilized diode laser. No repumping beams are used to form the MOT.

We reported the magneto-optical trapping of all abundant isotopes of $^{164-160}\text{Dy}$ in [27] with atom numbers ranging $1-5 \times 10^8$. We also observed MOTs of $^{156,158}\text{Dy}$, with orders of magnitude lower atom numbers due to their low natural abundances.

5.3.1 Recycling mechanism

We systematically studied the dynamics of the 421-nm Dy MOT. Details are addressed in [17, 27]. Perhaps the most interesting feature of this MOT is that it functions without repumpers, in contrast to the MOTs for alkali atoms. We performed MOT recapture experiments to quantitatively understand this mechanism.

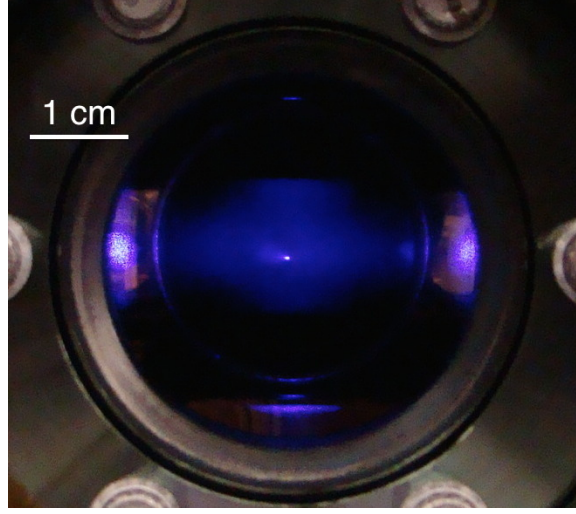


Figure 5.3: Color picture of blue 421-nm MOT. The bright blue dot at center is the trapped cold Dy cloud emitting resonant 421-nm light.

The experiment begins with a steady-state MOT. Subsequently, the MOT beams are quenched and the loading source is shut off. The remaining atoms are trapped only by the spherical quadrupole field. After a certain holding time, the MOT beams are turned back on and the fluorescence from the recaptured MOT is recorded. The evolution of the fluorescence shows several features: it quickly grows to a peak value several times *higher* than that of its progenitor MOT and it decays with a double exponential dependence.

These main features are captured in a minimal model (motivated from [18]) as illustrated in Fig. 5.4. In the 421-nm MOT, the blue cooling laser excites a fraction $f_{\text{ex}} < 1/2$ of the population to the 421-nm level, where it can decay with branching ratio R_1/Γ to the metastable states. Upon decay, a fraction pf_{ex} of the population is captured in the magnetic trap of the MOT at rate $pf_{\text{ex}}R_1$. MOT recycling data, examples of which are shown in Figs. 5.4 (b) and (c), support a dual decay path through the dark metastable states; the population decays through the slow (or fast)

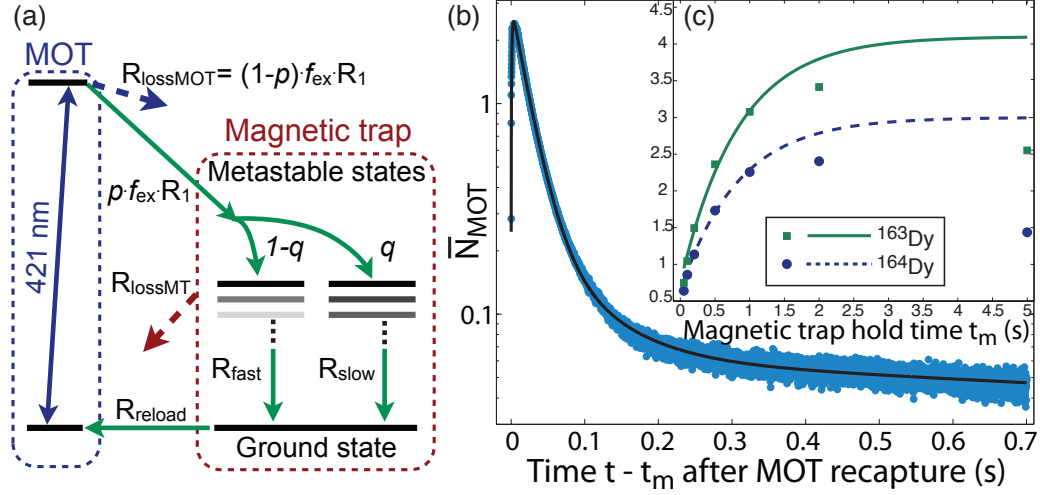


Figure 5.4: Dy 421-nm MOT loading and recycling model. (a) Dy MOT recycling and continuously loaded MT schematic. (b) Population ratio $\bar{N}_{\text{MOT}}$ of recaptured MOT to steady state MOT. Black line is fit using Eqs. ?? to $\bar{N}_{\text{MOT}}$ with Zeeman slower and atomic beam off, and $t_m = 1$ s delay between steady state MOT and recapture. Inset (c) plots the peak recaptured MOT population versus hold time in MT. Lines are fits to $t \leq 1$ s data with exponential time constants $[R_{\text{slow}}^{163}, R_{\text{slow}}^{164}] = [1.5, 2.3] \text{ s}^{-1}$ and extended to later times for comparison. $\bar{N}_{\text{MOT}} < 1$ at early times is due to the initial $N_{\text{fast}} \neq 0$.

branch with probability q (or $1 - q$) at rate R_{slow} (R_{fast}). Population that reaches the ground state reloads the MOT at rate R_{reload} , which depends on MOT parameters.

There are two loss rates from the otherwise closed system (once loading from the Zeeman slower ceases). First, portions of the MOT population can decay to non-magnetically trapped metastable states at rate $R_{\text{lossMOT}} = (1-p)f_{\text{ex}}R_1$. Additionally, metastable and ground state MT populations can be lost due to background and two-body inelastic collisions. Two-body loss is difficult to quantify in the metastable states, and we only investigate ground state MT loss. This is justified because R_{slow} is faster than the ground state component of R_{lossMT} , and we neglect R_{lossMT} in the rate equations.

Based on the model described above, we write down the coupled differential equa-

tions for populations in each channel that best fit the fluorescence curve in Fig. 5.4.

The final results for ^{163}Dy and ^{164}Dy (as examples for fermions and bosons):

$$[R_1^{163}, R_{\text{fast}}^{163}, R_{\text{slow}}^{163}] = [1170(20), 19(2), 1.5(1)] \text{ s}^{-1},$$

$$[R_1^{164}, R_{\text{fast}}^{164}, R_{\text{slow}}^{164}] = [1700(100), 29(1), 2.3(1)] \text{ s}^{-1},$$

and $[p, q] = [0.82(1), 0.73(1)]$.

With the fitting results from MOT recapture experiment, the dynamics of the 421-nm MOT become clear. First, the branching ratio $R_{\text{fast/slow}}/\Gamma < 1 \times 10^{-5}$ is quite small. Thus, the 421-nm transition is almost closed for laser cooling. This small branching ratio might be because these decays between nearby levels correspond to a few μm emissions which suppress the phase space in the final state after decay (Einstein spontaneous decay coefficient $A \sim \omega^3$), and transition matrices between those states and 421-nm excited states have small values. After our experiment, a first principle calculation of those transition strengths was reported which supported the observed low branching ratio [82].

Another feature of the 421-nm MOT is that atoms in dark states are not lost. Because of the luckily small branching ratio, atoms are sufficiently cooled before decaying into dark states. These cold atoms in dark states also have large magnetic moments and are readily trapped by the field gradient of the MOT. When they finally decay into the ground state, they can be recooled by 421-nm MOT beams. This recycling mechanism is not present in a conventional MOT, but can be created with the addition of one or more lasers in other atoms, such as Cr [83].

Since the lifetimes of those dark states are long ($\sim$ second), a large fraction of the atoms are trapped in those metastable states, which effectively serve as a reservoir.

In fact, most of the atoms are trapped in dark states rather than in bright states (Fig. 5.4 (c)), which explains the extra fluorescence in a time delayed recaptured MOT.

As a benchmark, we typically load and trap 5×10^8 atoms (2×10^8 ^{164}Dy atoms in bright states and 3×10^8 in dark states) for ^{164}Dy within 2 s, which is a relatively rapid. We note that achieving half of this number is enough for quantum gas production (see Chapters 9, 10) and is easily maintained on a daily basis.

Steady state bosonic Dy MOT populations form in proportion to natural abundance. However, the MOT population of fermions ^{161}Dy and ^{163}Dy are $1/6\times$ and $5/6\times$ the expected value, respectively. This is likely due to the lack of optical repumping that would otherwise serve to pump atoms out of the dark states hyperfine manifolds ($F = 11/2, 13/2, 15/2, 17/2, 19/2$). We note that for ^{163}Dy , the hyperfine splittings between $F = 21/2 \leftrightarrow F = 19/2$ in ground state and $F' = 21/2 \leftrightarrow F' = 23/2$ in excited state are accidentally near degenerate ($f_{F=21/2 \leftrightarrow F'=23/2} \sim f_{F=19/2 \leftrightarrow F'=21/2}$) (see Fig. 5.2 (b)).⁵ There is also a close degeneracy of the other transitions pairs except the last one. Thus, the cooling laser simultaneously serves as a hyperfine repumper, which explains the relatively larger MOT of ^{163}Dy .

5.3.2 Sub-Doppler cooling

The 421-nm MOT reaches a temperature of ~ 1 mK, which is consistent with the Doppler limit of the 421-nm transition ($T_{\text{Doppler}} = 760 \mu\text{K}$). The Doppler limit

⁵The hyperfine interaction strength in the 421-nm excited state is relatively strong ($\sim \text{GHz}$), compared to the typical hyperfine splittings of tens to hundreds of MHz in the excited states of alkali $D_{1,2}$ lines.

($T_{\text{Doppler}} = \hbar\Gamma/2k_B$) is the theoretical cooling limit. In a semiclassical picture it is due to the residual momentum diffusion of atoms arising from the spontaneous nature in atomic transitions.

Sub-Doppler cooling [84, 85, 86] beyond the Doppler limit is a crucial route in preparing quantum alkali gases. It normally occurs in an optical molasses [69] after MOT capture. Since differential optical light shifts between different Zeeman states is the key ingredient in this mechanism, a small Zeeman shift in low external magnetic fields is desirable. When Landé g -factors are near degenerate for both the ground state and excited state, the effect of magnetic fields is minimized and population wide sub-Doppler cooling may occur in a MOT [87, 88].

For the Dy 421-nm transition, $\Delta g/g_g = 1.7\%$ as compared to $\Delta g/g_g = 34\%$ for ^{87}Rb D_2 line. One naturally expects a similar in situ sub-Doppler cooling effect as above. Experiments show that the core of the Dy MOT are sub-Doppler cooled in one direction or another or all directions, depending on details of MOT operation parameters. The typical temperature is $200\ \mu\text{K}$, 10 times large than Rb and Cs optical molasses sub-Doppler cooled temperatures, but similar to sub-Doppler temperatures in the ^{87}Sr MOT [89]. Such anisotropic sub-Doppler cooling was systematically studied in [28] with a possible explanation based on the theory of velocity-selective resonances in a highly magnetic gas [90]. While the achieved temperature was as low as $\sim 10\ \mu\text{K}$ in some cases, only a small fraction (a few percent) of atoms are efficiently cooled in one direction in this mechanism, which makes sub-Doppler cooling less practically useful for quantum gas production. Details of the study on Dy sub-Doppler cooling were reported in [28].

5.4 Static quadrupole magnetic trap

Ultracold atoms in a spin mixture are expected to undergo inelastic collisions in general. We investigated these collision rates in the few-partial wave regime (at an initial temperature of 200 μK) via absorption imaging of the atoms confined in a static quadrupole magnetic trap (MT) at various delay times t_m , observing a fast decay rate in the spin mixture of Dy. In the many-partial-wave regime, Dy atoms were also observed to have a large inelastic collisional rate in Harvard group [91].

The population loss in the magnetic trap is likely due to background and inelastic collisions of ground state atoms in the MT (Majorana spin flip losses are negligible at these t_m 's and trap densities.). After loading the MOT, we extinguished the MOT, Zeeman slower, and atomic beams while maintaining a constant ∇B . The MT was turned off at t_m , and after 1 ms time of flight an absorption image was integrated for 200 μs . Accounting for gravity $-g\hat{z}$, the images taken in the x - z plane containing the quadrupole axis (along $\hat{z}$) were fit to:

$$p_x(x) \propto \exp(-2|x|\sqrt{1-\bar{g}^2/\bar{x}})(1+2|x|\sqrt{1-\bar{g}^2/\bar{x}}),$$

$$p_z(z) \propto \exp(-2|z|/\bar{z}-2\bar{g}z/\bar{z})(1+2|z|/\bar{z}),$$

where $\bar{g} = mg/(\bar{\mu}\nabla_z B)$, $\bar{\mu}$ is the mean μ of the atomic cloud, and characteristic lengths $[\bar{x}, \bar{z}] = 2k_B T/(\bar{\mu}\nabla_{x,z} B)$ [87]. We extract the mean cloud density n , temperature T , and $\bar{\mu}$ versus t_m , which are plotted in Fig. 5.5 for several MT gradients.

The ^{164}Dy density decay data in Fig. 5.5(a) were fit to the one plus two-body decay equation $\dot{n} = -\gamma n - \beta n^2$, where γ is the loss rate due to background collisions and β is a measure of inelastic two-body losses. For the three trap gradients, the extracted β 's

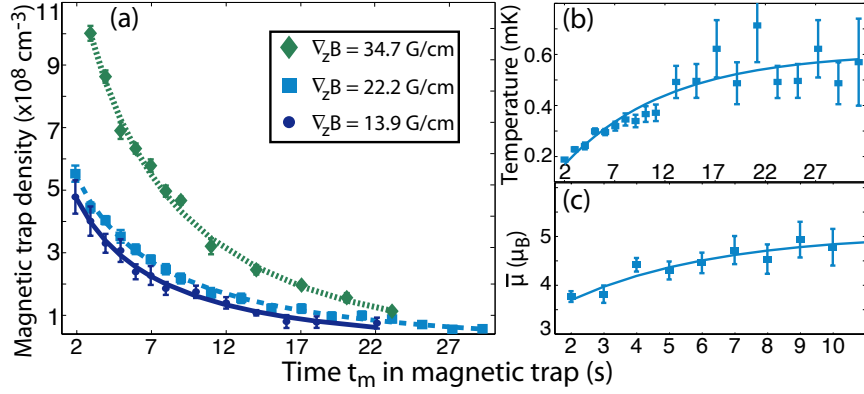


Figure 5.5: Two-body inelastic loss in magnetically trapped spin unpolarized Dy atoms in the few-partial-wave regime. (a) Decay of magnetic trap mean density for three gradients. Density profiles are consistent with a single thermal distribution after $t_m = 2$ s. Data are fit to obtain one (γ) and two-body (β) collisional loss rates. (b) Heating in $\nabla_z B = 22.2$ G/cm magnetic trap. (c) Increase in mean magnetic moment $\bar{\mu}$ for the same trap. An exponential fit gives a $\bar{\mu}(t)$ time constant of $\tau = 0.22(5)$ s $^{-1}$.

and γ 's are consistent with one another within 1σ ; the weighted means are $[\bar{\beta}, \bar{\gamma}] = [2.1(2) \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}, 5.6(3) \times 10^{-2} \text{ s}^{-1}]$. Fitting the data to $\dot{n} = -\beta n^2$ results in a worse χ^2 . Temperature heating data—e.g., Fig. 5.5(b)—are fit to an exponential, resulting at early times in a heating rate of $17(2) \mu\text{K/s}$, which is consistent with heating rates in spin unpolarized Er and Cr MTs [87, 92] and is likely due to spin-relaxation collisions. The MT population is initially distributed among weak-field seeking Zeeman states, and spin exchange collisions tend to polarize the sample toward $m_J = 8$. Indeed, $\bar{\mu}$ increases with time (Fig. 5.5(c)), and $\bar{\mu}(t)$ reaches $8 \mu_B$ within $t_m = 4$ s in the high gradient $\nabla_z B = 34.7$ G/cm trap.

The measured $\bar{\beta}$ is likely due to an unresolved combination of inelastic spin exchange, magnetic dipole-dipole relaxation (MDDR), and anisotropic electrostatic-driven spin relaxation collisions. Compared to $\beta_{\text{Cr}} = 3 \times 10^{-11} \text{ cm}^3/\text{s}$ in $200 \mu\text{K}$ spin-polarized ^{52}Cr [92], $\bar{\beta}$ is close after a correction of the MDDR scaling $\propto D^4 S^{-1}$

(where D is the dipolar length and S the spin quantum number as we shall discuss in Chapter 8). We note that $\bar{\beta}$ falls into a range that seems to be typical for those highly magnetic atoms: with non-maximally spin-polarized Cr ($1.1 \times 10^{-10} \text{ cm}^3/\text{s}$) [92] and with 500 mK inelastic Er and Tm collision rates, 3.0 and $1.1 \times 10^{-10} \text{ cm}^3/\text{s}$, respectively [91]. Subsequent theoretical work [93] considering all loss channels in a magnetically trapped Dy cloud at the experimentally relevant temperature showed a two-body loss rate at $\beta = 0.87 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ which is close to the value from our experiment.⁶

Such a large inelastic collisional rate for Dy (as well as other highly magnetic atoms) in a MT indicates that low-field-seeking states are not suitable for a conventional evaporative cooling in the MT. Evaporative cooling in the absolute ground state ($m = -8(-21/2)$) which is a high-field-seeking state, was therefore performed in an optical dipole force trap, leading to the production of quantum gases of Dy.

⁶We note there is a systematic error in estimating atom density in these experiments. The absorption imaging method suffers from spin dependent sensitivity and will underestimate the atom number in non-stretched states. In retrospect, fluorescence imaging would be more suitable in this experiment. A post-facto correction in spatial density lowers the fitted value of two-body loss rate for the given observed decay curves, which would make the values even more consistent with theoretical predictions.

Chapter 6

From 1 mK to 1 μ K: Narrow-Line Laser Cooling

6.1 General Considerations

Narrow-line magneto-optical traps were developed for the laser cooling and trapping of alkaline earth metals (and atoms with similar electron configurations, e.g. Yb). In those elements, the existence of the intercombination lines (spin singlet to triplet states) associated with very low transition probabilities drastically lowers the Doppler cooling limits from temperatures typically in the sub-mK regime to the μ K to sub- μ K regime. Narrow-line laser cooling plays a very important role in generating quantum alkaline earth gases [94, 95, 96, 97], as well as running neutral atom lattice clocks [98, 99]. Although narrow-line cooling can be understood from Doppler cooling theory, it is worth a detailed description due to its importance in preparing ultracold atomic gases of Dy as well as its unique technical challenges posed by Dy compared

to conventional narrow-line cooling.

The broad 421 nm transition is highly effective for loading and cooling Dy in a MOT, nonetheless its ~ 1 mK Doppler limit is too high for directly loading into an optical dipole trap (~ 100 μ K trap depth). On the other hand, Dy has a rich spectrum of weak optical transitions, and a narrow-line cooling stage after the 421 nm MOT can bridge this temperature gap. Particularly, 1001, 741, 626 and 598 nm lines were considered as possible candidates for several reasons (see Table 6.1 and Fig. 6.1): their linewidths were determined or predicted to be close to or below ~ 100 kHz, implying a Doppler limit of a few μ K; they are the $J \rightarrow J' = J + 1$ type of cycling transitions, where J, J' are the total electronic angular momenta for ground and excited states, respectively; and lastly, there are no E1 (electric dipole) allowed decay channels to states other than the ground state, thus they form an effective two-level system.

The final choice among them stems largely from practical considerations: the availability of reliable laser sources and the ease of maintenance. The 626 and 598 nm transitions fall into the visible regime where dye lasers are typically employed, while near infrared (NIR) solid state semiconductor lasers with single mode and relatively narrow linewidth are readily available for the 1001 and 741 nm transitions. Compared to dye lasers, semiconductor lasers are easier to maintain, safer, and their frequency narrowing is more robust. Ultimately, we decided to use the NIR transition at 741 nm.¹ All four candidate transitions were incompletely documented for the purpose of laser cooling; hence we carried out laser spectroscopy of the 741 nm transition

¹By the time of writing this work, the author is aware that Dy narrow-line cooling at 626 nm has also been demonstrated [100] using sum frequency generation which employing two different NIR fiber lasers.

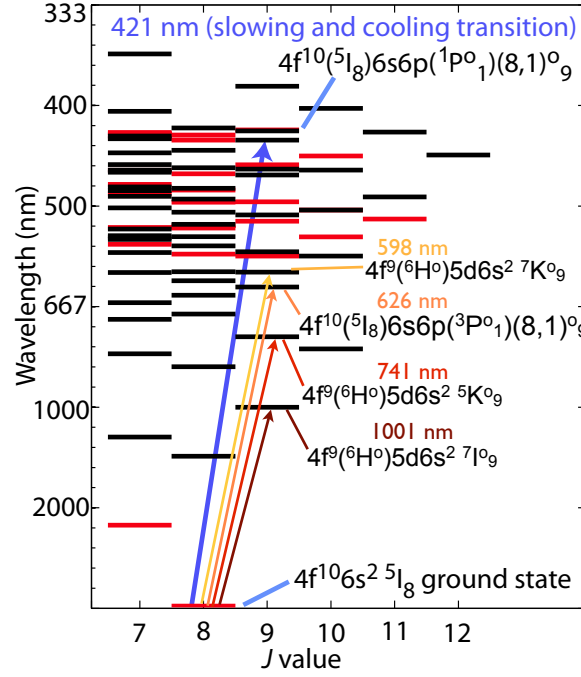


Figure 6.1: Candidates for narrow-line cooling transitions in Dy. Data of levels are from [77]. J is the total electronic angular momentum number. The relevant laser cooling transitions are marked with wavelengths and spectroscopic terms.

before any attempt of laser cooling. This transition turns out to be well suited for laser cooling, and we can cool Dy gases from 1 mK to $\sim 1 \mu$ K, with a generous atom number of tens of millions, for all isotopes. This significantly improves the initial conditions of the evaporative cooling stage that follows.

Table 6.1: Laser cooling parameters for four narrow-line transitions in Dy

Lines	g	$\Delta\nu$	τ	I_{sat}	v_{cap}	T_{Doppler}	$T_{\text{recoil}}^{\text{I}}$
598 nm	1.24	12 kHz ^{II}	13 μ s	7.5 μ W/cm ²	7.3mm/s	290 nK	327 nK
626 nm	1.29	135 kHz ^{III}	1.2 ns	72 μ W/cm ²	8.5cm/s	32 μ K	298 nK
741 nm	1.23	1.78 kHz ^{IV}	89.3 μ s	0.57 μ W/cm ²	1.3mm/s	43 nK	213 nK
1001 nm	1.32	53 Hz ^V	3 ms	6.9 nW/cm ²	0.05mm/s	1.3 nK	116 nK

6.2 Spectroscopy of 741 nm optical transition

The key aspects studied by this spectroscopic work of the 741 nm transition were the resonant frequencies of the different isotopes and the linewidth of the transition. We set up an apparatus to determine the isotope shifts and hyperfine structure, as well as the lifetime of the excited state.

6.2.1 The apparatus for frequency measurement

The spectroscopic measurement for determining the isotope shifts and hyperfine structure employed a crossed excitation laser and atomic beam method [51]. In a UHV chamber [102], thermal Dy atoms effused from a high-temperature oven working at 1275 °C. The atoms were collimated by a long differential pumping tube which formed an atomic beam with a diameter of 5 mm and a diverging (half) angle of ~ 0.02 rad.

^I g is the Landé factor of the excited state, $\Delta\nu$ is the linewidth is, τ is the excited state lifetime, $I_{\text{sat}} \equiv \pi\hbar c\Gamma/3\lambda^3$ is the saturation intensity, $v_{\text{cap}} \equiv \Gamma\lambda/2\pi$ is the Doppler cooling capture velocity, $T_{\text{Doppler}} = \hbar\Gamma/2k_B$ is the Doppler limit, and $T_{\text{recoil}} = \hbar^2k^2/mk_B$ is the single-photon recoil temperature.

^{II}Ref. [101], theory

^{III}Ref. [77], experiment 5% uncertainty

^{IV}Ref. [16], experiment 1% uncertainty

^VRef. [101], theory

The beam entered a chamber with two pairs of optical viewports oriented orthogonally to the atomic beam.

A 5 mW 741 nm laser beam [$1/e^2$ waist (radius) ≈ 2 mm] from an external cavity diode laser (ECDL, Toptica DL Pro) was directed through the UHV chamber via one pair of viewports. This laser had a mode-hop-free tuning range of 20 GHz. Atomic fluorescence on the 741 nm line was collected through an orthogonal viewport by a 2'' AR-coated achromatic lens pair with a magnification of $0.4\times$. This formed an image on the detection area (1-mm diameter) of a femtowatt photodetector (Si PIN, DC gain: 2×10^{10} V/W, bandwidth ≥ 750 Hz). The whole system was carefully shielded from stray light.

The direct output from the detector suffered from low signal-to-noise (SNR) due to the oven's thermal radiation and the multiple scattering of 741 nm light from the windows and the chamber's inner walls. Averaging 512 times and adding an electronic bandpass filter (0.3 Hz to 3 kHz) with a DC amplification of 10 improved the signal-to-noise to a sufficient level without artificially broadening the Doppler-limited resonances. With a laser scanning rate of 7 GHz/s and a 15 MHz residual Doppler-broadening, the ~ 2 ms transit time of the spectral peaks are only somewhat slower than the detector's response time (200 μ s). Nevertheless, all relative peak position data are sufficiently insensitive to this effect to render it negligible for the following analysis.

To measure the full spectrum of isotope shifts and hyperfine states, we scanned the ECDL using a piezoelectric transducer (PZT) that modulates its grating position. However, the free scan of the ECDL suffered from the slight nonlinear scanning of the

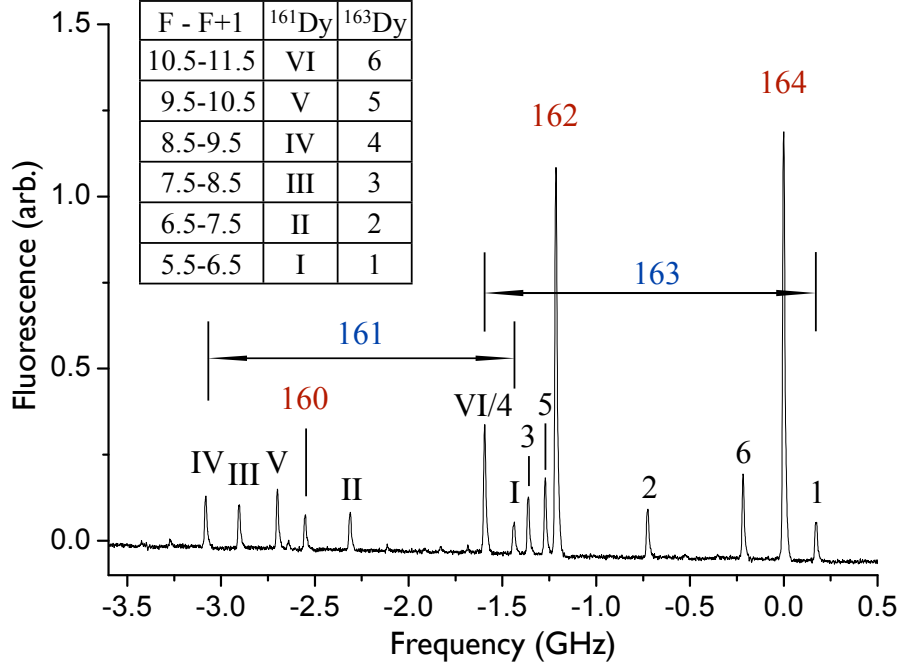


Figure 6.2: Spectrum of 741 nm line for the five most abundant Dy isotopes. Bosonic isotope peaks are marked with mass numbers in red. Different hyper-fine peaks of fermionic isotopes (blue) are identified with the markers defined in the inset table. The 6th peak of ¹⁶¹Dy and the 4th peak of ¹⁶³Dy overlap.

PZT versus drive voltage. To calibrate the frequency scan, we coupled the 741 nm light into a temperature-stabilized confocal cavity with a 750 MHz free-spectral range (FSR) within a scan range > 10 GHz. Simultaneously recording the transmission of the confocal cavity with the fluorescence signal provides a frequency calibration as the ECDL is scanned. Cavity frequency intervals were calibrated by imprinting rf sidebands—which repeat every FSR interval—onto the cavity-coupled 741 nm light with a stable and calibrated rf frequency source. To correct for the nonlinearity of the scan, a calibration was performed by fitting a polynomial to the cavity spectrum. The maximum deviation throughout the 10 GHz scan due to a quadratic term was 3% of the linear term (the cubic term was orders of magnitude smaller). We corrected

the nonlinear effect up to quadratic order.

The calibrated spectrum after 512 averages is shown in Fig. 6.2. These data are sufficient to resolve and identify all $J \rightarrow J + 1$ ($F \rightarrow F + 1$) 741 nm transitions for the bosonic (fermionic) isotopes. Optical pumping was a negligible effect in this spectrum since the $\sim 10 \mu\text{s}$ atoms' transit time was much shorter than the transition lifetime (see below).

6.2.2 Hyperfine structure

The relatively weak interactions between electrons and the atomic nucleus lead to a splitting of the atomic energy levels. This is normally orders of magnitude smaller than the splittings of the fine structure due to spin-orbit coupling effects in atoms. The hyperfine Hamiltonian could be written down in the form:

$$H_{\text{hfs}}/h = A \vec{I} \cdot \vec{J} + B \frac{\frac{3}{2} \vec{I} \cdot \vec{J} (2 \vec{I} \cdot \vec{J} + 1 - I(I+1)J(J+1))}{2I(2I-1)J(2J-1)} + \dots \quad (6.1)$$

where h is the Planck constant, J and I are the electron and nucleus angular momenta, respectively, and A and B are constants to be determined.

Physically, A represents the strength of the magnetic dipole interaction due to the dipole moment of the atomic nucleus in the magnetic field generated by the electrons. For alkali metals it is the dominant term in determining the hyperfine structure. B represents the strength of the electric quadrupole interaction due the quadrupole moment of the atomic nucleus in the electric field gradient generated by the electrons. It is nonzero only when $I, J \geq 1$. For Dy, B is nonnegligible. All higher perturbative terms are ignored.

Table 6.2: Values of hyperfine coefficients A and B (MHz) for the excited state of the 741 nm transition in Dy, and comparison with those of the ground state.

Coeff.	$^{163}\text{Dy}_{\text{th}}^{\text{I}}$	$^{163}\text{Dy}_{\text{expt}}$	$^{161}\text{Dy}_{\text{expt}}$	$163/161_{\text{e}}$	$163/161_{\text{g}}^{\text{II}}$
A	142	142.91(3)	-102.09(2)	-1.3999(4)	-1.40026(4)
B	4000	4105(2)	3883(1)	1.0570(9)	1.05615(90)

The hyperfine peaks of ^{163}Dy and ^{161}Dy are highly mixed in frequency due to the similarity of the ground and excited states' A coefficients. Identification of the isotope and hyperfine transition peaks were determined with guidance from theory in Ref. [101], and a least squares fitting routine extracts the values of A and B (see Table 6.2). Since this is a J to $J + 1$ transition, for the fermions the observed lines are of the $F \rightarrow F + 1$ type; the much weaker $F \rightarrow F$ and $F \rightarrow F - 1$ transitions are barely resolved or even fall below the noise floor in Fig. 6.2. The relative strengths of those resonant peaks are very consistent with calculations involving $3j$ and $6j$ symbols [65]. We note that for the excited states, the ratios of $A^{163}/A^{161} = -1.3999(4)$ and $B_e^{163}/B_e^{161} = 1.0570(9)$ are consistent with the analogous ratios for the ground state, within 0.01%. There is no noticeable hyperfine anomaly for the 741 nm transition [103].

Based on the values of A and B from Table 6.2, the hyperfine structure of the ground and excited state of 741 nm transition is drawn in Fig. 6.3. For fermions, the cycling transitions for laser cooling would be between $F=21/2$ and $F'=23/2$. Notice that the hierarchy of hyperfine manifolds for ^{161}Dy and ^{163}Dy are opposite due to the

^IRef. [101]

^{II}Ref. [104]

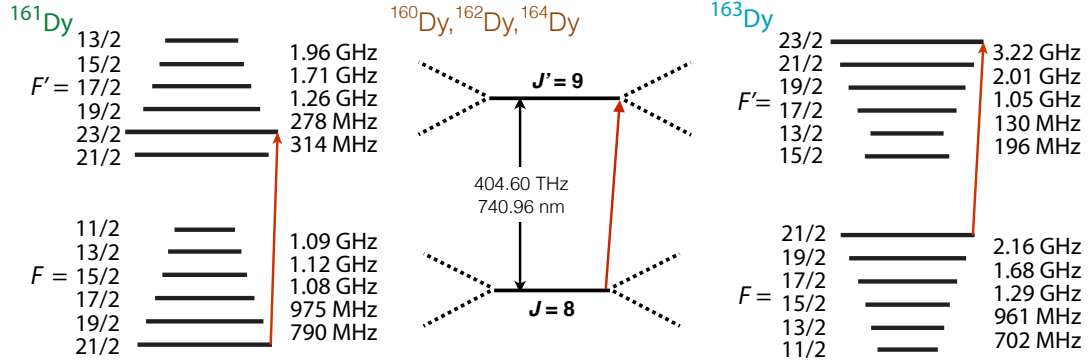


Figure 6.3: Hyperfine structure and 741 nm laser cooling transitions for ^{161}Dy and ^{163}Dy . Middle panel shows the unsplit ground state and excited state of 741 nm transition for ^{160}Dy , ^{162}Dy , ^{164}Dy , and the transition frequency is from [77]; left or right panel shows the hyperfine structure and cooling transitions for ^{161}Dy or ^{163}Dy respectively.

different signs of A for these two isotopes. This leads to different collisional properties of the two isotopes, as we will discuss in Chapter 8. Also, $F'=23/2$ for ^{161}Dy and $F'=13/2$ for ^{163}Dy appear out of the sequential order in each of the manifolds they belong to due to the relatively large values of B .

6.2.3 Isotope shifts

The measured values of isotope shifts (from ^{164}Dy to ^{160}Dy) for the 741 nm transition are listed in Table 6.3 with reference to ^{164}Dy , together with the documented isotope shifts for the 457 nm line. The 457 nm line has a pure electronic configuration which makes it useful as a reference transition for creating the King plot [105] described below. The fermionic isotope shifts are derived from the center-of-gravity of the hyperfine peaks, the positions of which are extracted from hyperfine structure fits. We could not obtain the isotope shift of ^{158}Dy and ^{156}Dy due to their low natural

Table 6.3: Isotope shifts in the 741 nm and 457 nm transitions in Dy.

Isotope shifts	741nm	457 nm ^I
¹⁶⁴ Dy	0	0
¹⁶³ Dy	−915(2) MHz	660(3) MHz
¹⁶² Dy	−1214(3) MHz	971(2) MHz
¹⁶¹ Dy	−2320(5) MHz	1744(3) MHz
¹⁶⁰ Dy	−2552(5) MHz	2020(3) MHz

abundances and the weak strength of the 741 nm transition.

The isotope shift arises due to the differences in the masses and volumes of the nuclei of the isotopes and has the following form: $\delta f_{i,A} = E_A V_i + M_{i,B}$, where $\delta f_{i,A}$ is the isotope shift for the isotope pair i in the transition A. The first term on the right hand side is the contribution from the volume effects, which can be decomposed into the electronic field-shift parameter E_A and volume factors V_i . The mass term $M_{i,A}$ which can be further decomposed into the sum of the normal mass shift (NMS) $M_{nms,i,A}$ and the specific mass shift (SMS) $M_{sms,i,A}$. The NMS is directly proportional to the change of the reduced electron mass between the isotope pair, which can be analytically calculated [105]. The SMS describes the correlation between momenta of electrons and is determined by the intercept of the King plot.

For two different transitions A and B, the equation above can be rewritten as:

$$\delta f_{i,B} = \frac{E_B}{E_A} \delta f_{i,A} + M_{i,B} - \frac{E_B}{E_A} M_{i,A}. \quad (6.2)$$

The new equation indicates that the experimental data fall on a line with a slope of

^IRef. [106]

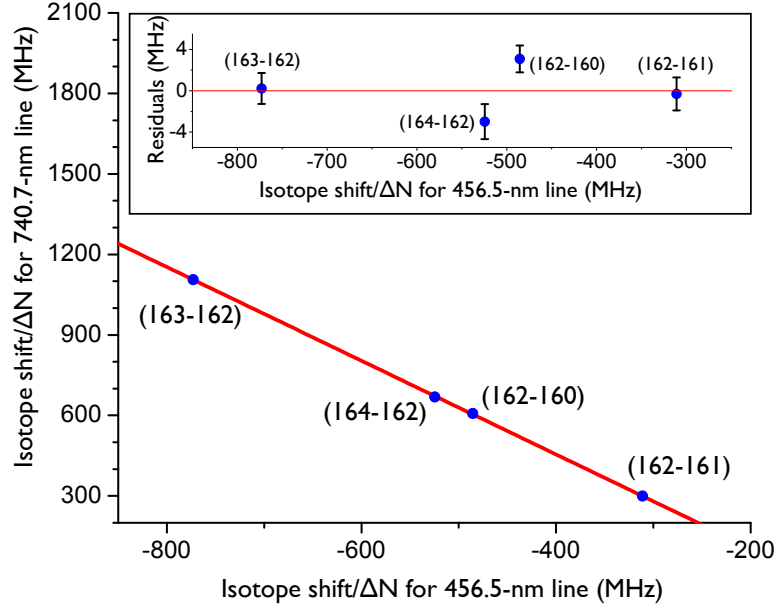


Figure 6.4: King plot of the isotope shifts in the 741 nm line versus isotope shifts in the 457 nm line. ΔN is the mass number difference between the isotope pairs. Inset is the fit residual.

$\frac{E_B}{E_A}$ (King plot). The intercept of the plot provides the relation of mass shifts between A and B transitions. With the independent knowledge of the NMS and SMS for transition A and the NMS for transition B, the SMS for transition B is determined.

We draw the King plot using the documented $4f^{10}6s^2(^5I_8) \rightarrow 4f^{10}6s6p(^7I_8)$ transition at 457 nm as the reference transition [106, 103]. As discussed above, the isotope shift includes the contribution from the mass term, which is related to the change of isotope mass, and the field shift, which is due to the finite possibility of electrons being inside the nucleus. Different electron configurations lead to different field shifts: From the fit of the isotope shifts of the 741 nm transition plotted against those values for 457 nm, the ratio of electronic field-shift parameter $E_{741}/E_{457} = -1.746(9)$ is determined based on the slope of the linear fit. The minus sign indicates the very

different nature of two transitions, i.e., one is of $4f \rightarrow 5d$ type while the other is $6s \rightarrow 6p$.

The NMS for the 457 nm and 741 nm transitions are $\delta\nu_{\text{nms}164-162}^{457} = 27$ MHz and $\delta\nu_{\text{nms}164-162}^{741} = 17$ MHz, respectively [105]. The SMS of the 457 nm transition is 7(8) MHz [106], which allows us to calculate the SMS for the 741 nm line: $\delta\nu_{\text{sms}164-162}^{741} = 563(17)$ MHz, based on the intercept of the King plot. Such a large SMS is known to arise from the following effects: $4f$ electrons deeply buried inside the electron core, strong electron correlations, and $4f$ electrons coupling to each other before coupling to the outer $6s$ electrons [78]. A transition of type $6s \rightarrow 6p$ leaves the inner electrons little changed, while $4f \rightarrow 5d$ transitions lead to large changes in the inner electron correlations. The large experimental value of the 741 nm transition SMS is consistent with the typical values for $4f \rightarrow 5d$ transitions [107].

6.2.4 Lifetime measurement

A direct lifetime measurement based on the fluorescence decay observed with the crossed-beam method is not possible due to large transit time broadening relative to the natural decay time. (The UHV chamber limits our observation path length to a few cm; the 500 m/s beam velocity causes the transition time to be comparable to the transition lifetime.) Therefore, we resorted to measuring the fluorescence scattered from relatively static atoms, i.e., from the ~ 1 mK atoms in the MOT and in the magnetostatic trap (MT). We measured a consistent lifetime with both methods.

In the “MOT” method, we shine a retroreflected 5 mW 741 nm excitation beam of waist 3 mm onto a MOT generated on the 421 nm transition and record the decay

of 741-nm scattered light after the 741 nm beam is extinguished. While the 741 nm excitation beam is on, the system establishes a steady-state population distribution among the 421 nm, 741 nm, and ground states. By switching on and off the 741 nm laser beam with a period of 250 μ s, the atoms initially shelved in the 741 nm state will decay back to the ground state via the spontaneous emission of 741 nm photons. The small $1:10^5$ branching ratio of the 421-nm transition, measured in previous work [108, 102], means that the Dy atom is effectively a three-level system during the 250 μ s decay measurement: decay out of the three-level system from the 421 nm state to the metastable states occurs on a much longer time scale, >2 ms. Solutions to the optical Bloch equations [109] for such a “V” system—two excited states coupled to a ground state via resonant 421 nm and 741 nm light—verify that the decay rate observed is equal to the decay rate of the bare 741 nm state.

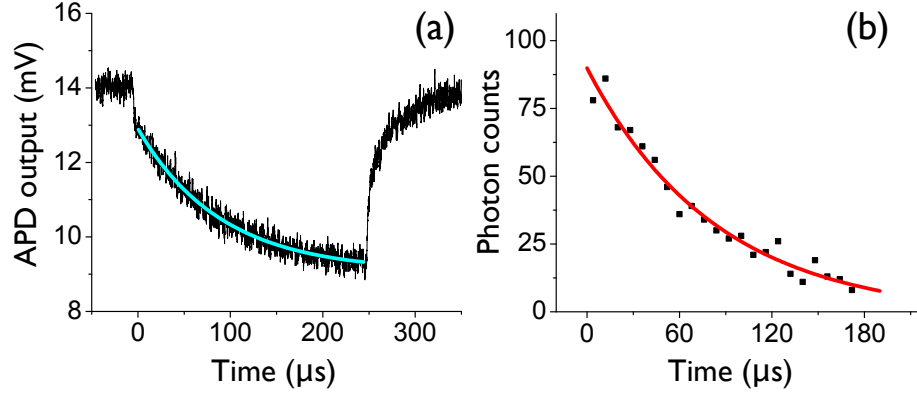


Figure 6.5: Measured decay of 741-nm fluorescence. (a) MOT method: Recorded 741-nm fluorescence signal from a Dy MOT by an APD averaged 10752 times; (b) MT method: Photon counting of scattered 741 nm light from a magnetic trap.

An avalanche photodetector (APD) and collection lenses with a 741 nm narrow bandpass filter are used to detect the weak signal, which we average 10752 times to

Table 6.4: Lifetime of 741 nm excited state

MOT method	MT method	Theory ^I
89.6(7) μ s	84(14) μ s	21 μ s

obtain the data in Fig. 6.5(a). Note that for neither this method nor the following one does the presence of 421 nm light or magnetic field gradients affect the natural decay rate measurement of the closed 741 nm transition.

In the “MT” method, we extinguish the 421-nm MOT light and capture the atoms in the magnetic quadrupole field of the now-extinguished MOT. We wait 5 s to allow the atoms to equilibrate in the MT [108] before shining onto the trap a resonant retroreflected 741 nm beam of 5 mW power and waist 3 mm. A single photon counter with collection lenses and a 741 nm bandpass filter records the very weak flux of 741 nm photons from the MT after the 741 nm excitation beam is extinguished [see Fig. 6.5(b)]. The long experimental run time necessary to measure a single decay limits the obtainable statistics.

Single exponential fits to the data in both methods derive lifetimes that are consistent with each other (see Table 6.4). We note that the values are $4\times$ longer than the theoretical value reported in Ref. [101], and the measurement reported here may be used to refine Dy structure calculations [110]. With such a narrow linewidth—the weighted combination of the lifetime is 89.3(8) μ s, resulting in a linewidth of 1.78(2) kHz—narrow-line cooling on the 741 nm transition is more technically challenging than originally thought.

^IRef. [101]

6.3 The 741 nm MOT

6.3.1 A very narrow line for laser cooling

Narrow-line laser cooling with a 2 kHz-wide optical transition is quite challenging and drastically different than the 421 nm Dy MOT. First, the capture velocity on 741 nm cooling is orders of magnitude smaller than the typical atoms' velocities after 421 nm cooling (see Table 6.5). This velocity mismatch could lead to an almost complete loss of atoms during the transfer to the narrow-line MOT, when employing conventional MOT settings. This is because Doppler cooling is effective only within the capture velocity as well as the fact that the narrow-line transition takes such a long time for a single excitation-decay cycling process (for 741 nm it is 0.2 ms) that the thermal atoms can drift out of the MOT beams before they are efficiently slowed and retrapped in the new MOT.

Second, the maximum optical forces in a 741 nm MOT at infinite light intensity is only 1.9 times the force of gravity, and in practice the optical forces may be equal to or less than gravity. This is in contrast to the conventional optical-force-dominated MOT. For comparison, the mature and highly successful Sr narrow-line MOT [111, 112] works in the regime in which optical forces are weak yet still eight times larger than gravity, but even so gravity's influence in the Sr MOT is far from negligible [112].

Additionally, for continuous cooling, the spatial gradient of the Zeeman shift should be smaller than the Doppler shift due to the deceleration from one absorption-reemission photon process. This is the adiabatic condition [111] which can be expressed as $\mu dB/dz < \hbar k^2 \gamma / 2mv$, where $\mu = 2\pi \times 1.7$ MHz/G for Dy, ∇B is the field

Table 6.5: Differences in laser cooling parameters of 421 nm and 741 nm transitions

Lines	$\Delta\nu$	τ	v_{cap}	$v_{\text{rmsD}}^{\text{I}}$	$F_{\text{OPT}}^{\text{II}}$	F_B^{III}
421 nm	32 MHz	5 ns	14 m/s	0.34 m/s	6×10^4	7
741 nm	1.8 kHz	89 μ s	1.3 mm/s	2.6 mm/s	1.9	1.2

gradient of MOT, m and v are the mass and velocity of atoms, and k and Γ are the wave vector and linewidth of the 741 nm transition. While fulfilling this condition is trivial for a MHz-wide transition, for a kHz-wide transition it is tricky. For Dy at a few G/cm, this criterion is not met at any temperatures hotter than 10 μ K.

For these reasons, an unconventional narrow-line MOT is needed to solve the issues.

6.3.2 "Blue-detuned" MOT

McClelland's group proposed a novel narrow-line MOT scheme that is applicable to strongly magnetic atoms [113]. The conclusion, which was experimentally demonstrated, is somewhat counter-intuitive: the atoms can be cooled in a quadrupole field with a laser frequency tuned to be higher than the natural atomic resonant frequency ω_A (i.e. blue detuned). This differs from normal Doppler cooling, which occurs on the other side of the atomic resonance (i.e., red detuned). The Zeeman shift away from the quadrupole field zero then shifts the transition such that the cooling lasers are effectively red-detuned at this offset position, which now provide a large magnetic

^IThe rms velocities at Doppler limits.

^{II}The max optical forces at the infinity light intensity ($\hbar k \Gamma / 2$) in units of gravity force.

^{III}The magnetic-field-gradient force in units of gravity force according to each typical MOT settings. For 421-MOT the gradient is 20 G/cm and for 741-MOT is 3.5 G/cm.

field gradient force to support against gravity while the optical fields cool.

Consider an atom moving only in the z direction in an inhomogeneous magnetic field $B = B'z$ together with a single circularly polarized laser beam (wave vector k and frequency ω_L detuned from atomic resonant frequency by $\delta = \omega_L - \omega_A$) traveling in the $-z$ direction. In the negative spatial ($z < 0$) regime, a semi-classical force on the atom depends on position z and velocity v in the following way [113]:

$$F(z, v) = F_{\text{conserv}} - F_{\text{opt}} \frac{s}{1 + s + 4(\delta/\Gamma + z/z_0 + kv/\Gamma)^2}, \quad (6.3)$$

where

$$F_{\text{conserv}} = -Mg + g_g m_J \mu_B |B'|, \quad F_{\text{opt}} = \frac{\hbar k \Gamma}{2}, \quad z_0 = \frac{\hbar \Gamma}{\mu' |B'|}, \quad (6.4)$$

g_g is the Lande factor for ground states, $\mu' \approx g_e \mu_B$ for Dy after neglecting the fortuitously tiny differences of Lande factors between ground and excited states, m_J denotes Zeeman state of the atom, s the saturation parameter [69].

A stable trap may be formed [113] that for polarization σ^+ , laser detuning $\delta > 0$, and atoms in $m_J > 0$, as long as

$$0 < \mathcal{F} \equiv F_{\text{conserv}}/F_{\text{opt}} < s/(1 + s) \leq 1. \quad (6.5)$$

This takes the form of a damped harmonic oscillator: $F \approx -\kappa(z - z_{eq}) - \beta v$, near a position $z_{eq} < 0$ and for $v \sim 0$, where $\kappa, \beta > 0$ are the spring constant and damping coefficient, respectively. Indeed, a direct simulation (Fig. 6.6) clearly shows the appearance of a potential dimple with damping forces at low velocities.

The existence of such an attractor in phase space could be understood in the following picture. An atom in a low-field-seeking state ($gm_J > 0$) is attracted towards the magnetic quadrupole field center due to gradient forces which dominate over the

pull of gravity. As the atom moves in the field gradient and the resonant frequency of the 741 nm line is modulated by the Zeeman shifting, the optical force is also modulated. At a certain point the optical force together with gravity balances the gradient force, which turns out to be a stable equilibrium. The optical force pushes the atom to this new equilibrium position $z_{eq} < 0$. At z_{eq} the laser frequency is locally red detuned ($\delta(z) = \omega_L - \omega_a(z) < 0$) and the familiar Doppler damping effect is recovered.

There are several new features for this blue-detuned MOT. The trapped gas is off-center from the quadrupole node and the trapping center is laser-detuning dependent in a linear way. As laser frequency is more blue detuned, the position of the MOT center shifts outwards from the quadrupole field node while trapping frequencies and damping coefficients at the dimple remain unchanged, see Fig. 6.6. This is a handy tool for finely tuning the position of the trapped cloud. At the same time, the accuracy of laser frequency detuning is less essential for cooling than one might worry, considering usual MOT frequency detunings are a few or tens of Γ , which would pose a rather stringent requirement on long-time frequency stability of the laser system.

Another important feature is this type of MOT *does not* require a balance between gravity and the optical force. As long as the gradient force is larger than gravity, the latter does not pose a severe limit on the linewidth of the narrow-line cooling transition. Hence very narrow optical transitions can be used. The only requirement is that the atoms possess a strong enough magnetic moment in low magnetic gradients.

We note that because the magnetic field is non-zero and directionally well-defined throughout the dimple regime, the MOT beam can optically pump the atom among

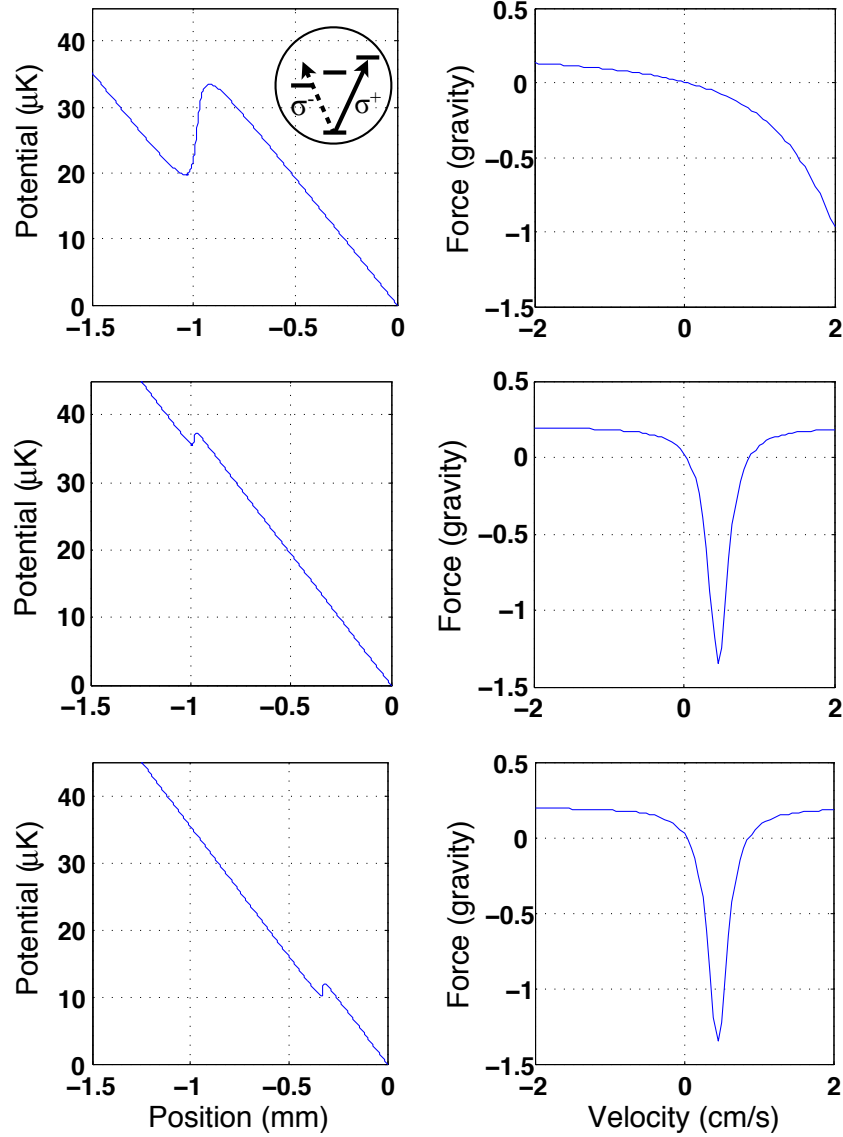


Figure 6.6: Numerical simulation of the 741 nm MOT in trapping and cooling. The left column shows the conservative potential profile an static atom experiences, gravity is along the negative direction; the right column shows the damping force for small velocities at the trapping center, in units of gravity. Upper panel corresponds to $I/I_s = 200$, $\delta = 600$ kHz; middle panel corresponds to $I/I_s = 10$, $\delta = 600$ kHz; lower panel corresponds to $I/I_s = 10$, $\delta = 200$ kHz. Magnetic field gradient $\partial_z B = 3.5$ G/cm for all. The round inset shows Zeeman shifting of excited sublevels (only 3 out of 19 in total shown) favors σ^+ transitions due to laser's "blue detuning".

Zeeman sublevels which leads to a highly spin-polarized MOT. The mechanism is illustrated in figure 6.6, where the near resonant σ^+ beam pumps atoms to the stretched state $m_J = J$. This eliminates the need for an optical pumping stage subsequent to MOT cooling which is normally needed for spin state preparation before evaporative cooling to prevent undesirable heating. This spin purification occurs at the same time that we take the advantage of low Doppler limit in a narrow-line MOT.

From simulations we note that, not surprisingly, this narrow-line MOT has a quite shallow trap depth (Fig.6.6). At high MOT-beam intensity ($s \gg 1$), the trap depth is relatively deep, but the damping coefficient is low. A colder temperature is obtained when the damping coefficient is higher at low MOT-beam intensity ($s \sim 1$) according to the simulation. This is also verified from the analytical expression of MOT temperature: $T = T_D (s/2\mathcal{F}\sqrt{s/\mathcal{F} - 1 - s})$, where $T_D = \hbar\Gamma/2k_B$, $s = I/I_s$ [113].

The model above explains the situation in 1D. In experiments, we need 3D cooling and trapping. Additional laser beams are added in the x,y directions. Note that because the stable trapping and cooling occurs on a constant-field contour surface (see the "smiling face" in Fig. 6.9), which is an arc zone right below the quadrupole field node, the circularly polarized beams in x, y directions are not purely σ^+ when viewed by atoms locally. This means the pumping and cooling efficiencies suffer to some degree and it leads to a temperature difference $T_{x,y} > T_z$.

6.3.3 Experimental results

The narrow-line MOT stage starts from the moment the 421 nm MOT beams are turned off. Dy atoms in low-field-seeking states ($m_J > 0$) are subsequently loaded into the quadrupole magnetic trap (MT). The field gradient is adiabatically reduced to ~ 10 G/cm, which lowers both the temperature of the cloud and the real space density (phase space density is conserved). This facilitates the initial capture for narrow-line cooling and also minimizes the inelastic collisional loss from dipolar relaxation. At the same time, Dy atoms from metastable states decay (see chapter 4) back into the ground state. Empirically, two seconds of collecting in the MT maximizes the total atom number in the 741 nm MOT, which is consistent with the lifetime of those metastable states (for more details refer to the chapter on 421 nm MOT). During this stage, the 741 nm beam's intensity is kept maximized and frequency modulated [111] at 15 kHz with a frequency excursion of up to 1 MHz. Such a large spread of sidebands in frequency greatly enhances the probability of capturing atoms belonging to different velocity groups.

Once the majority of the atoms are successfully trapped in the MT in the ground state, the magnetic field gradient is further reduced from ~ 10 G/cm to 5 G/cm. Narrow-line cooling takes effect simultaneously since the condition in eqn. 6.5 is fulfilled. Even though the capture velocity of the 741 nm MOT is rather small, the atoms' velocity is reduced at the classical turning points of the potential wall in the MT, greatly increasing the capture efficiency of atoms that are otherwise moving too quickly (Fig.6.8). The damping effect in the dimple ensures that the atoms, once captured, will be cooled and re-trapped in the narrow-line MOT. We further enhance

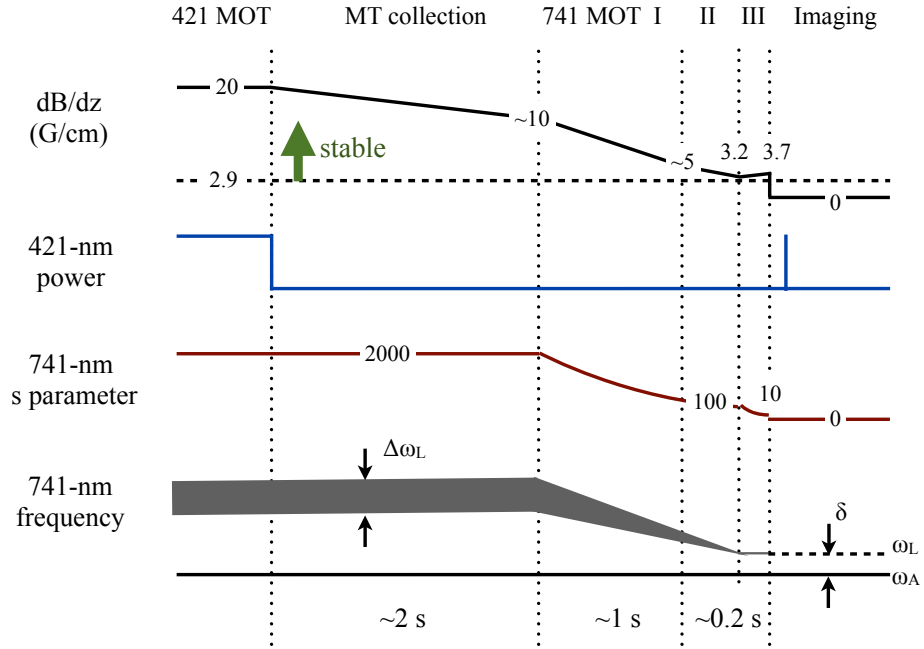


Figure 6.7: Loading sequence of the 741 nm MOT. The stages are MT stages, 741 nm MOT in different substages I, II, III and the (optional) imaging stages. dB/dz is the field gradient (2.9 G/cm is the critical value before Dy atoms fall due to gravity), $s = I/I_s$ is the saturation parameter, $\Delta\omega_L$ is the broadened laser frequency range, and δ is the laser frequency (ω_L) detuning from the unshifted atomic resonant frequency ω_A usually +0.5 — +1 MHz. The plot is not to scale. The numbers should be regarded as a guide while engaging in experimental optimization. Refer to text for details.

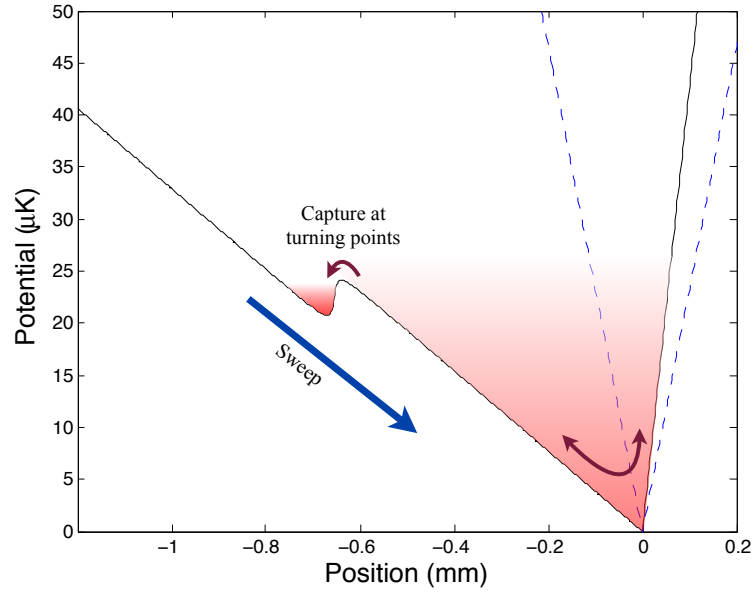


Figure 6.8: Capture mechanism of the 741 nm MOT. The V-shaped solid lines depict trapping potentials due to conservative forces, while dashed lines are for the trapping potential due to magnetic gradient force alone. The magnetic atoms are held by the quadrupole field as illustrated by the bottom arrow. The color scale reflects the in situ atomic density. Near classical turning points, atoms are almost stationary and hence are captured and accumulated in the dimple formed by 741 nm MOT beams. As the MOT frequency detuning sweeps, the dimple collects nearly all atoms along the way towards the quadrupole field null point (depicted as blue arrow pointing downwards).

capture efficiency by moving the dimple towards the MT center, thereby collecting atoms from a large spread of initial velocities as they come to a stop at the classical turning points of the MT potential. This collection scheme removes the tight constraints on capture velocity in the narrow-line cooling scheme.

As a considerable fraction of atoms are loaded into the dimple, we slowly lower the field gradient to slightly above the critical value at which magnetic trapping is stable against gravity, which favors the adiabatic condition.² and lower MOT temperature [113] Meanwhile, we decrease the MOT beam intensity. As the atoms cool, we decrease the spread of frequency modulation sidebands of the MOT beams. The atoms reach the lowest temperature when a field gradient of 3.2 G and laser intensity of $s = 10$ are reached. Finally, we raise the field gradient a little to compress the cloud at a cost of slightly higher temperature, resulting in a higher phase-space density in the MOT [113].

Following the procedures above, the overall loading efficiency from the 421 nm MOT to the 741 nm MOT reaches 50%, while the loss is likely due to inelastic collisions in the MT, permanent loss of atoms in high-field-seeking states before they could be optically pumped into the correct trapping states, and perhaps Majorana loss in the later stages when the quadrupole-field-trapped atoms become very cold.³ The minimum temperature obtained in the z direction reaches $T_z \sim 700$ nK, while in transverse directions $T_{x,y} \sim 1 - 2$ μ K. While there are trade-offs between atom number, density, and temperature, a typical combination of key parameters after

²As we discussed earlier, a lower magnetic gradient decreases R which favors the adiabatic condition for continuous laser cooling

³For comparison, the transfer efficiency in the NIST Er narrow-line MOT, which didn't employ as sophisticated a transfer procedure, was less than 10% [114].

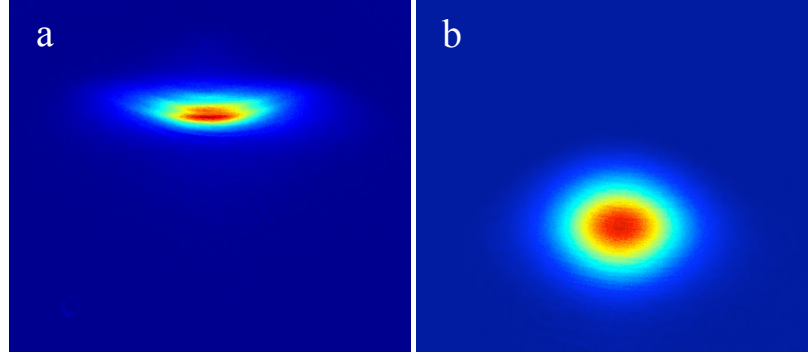


Figure 6.9: Time of flight σ^+ absorption images of the 741 nm MOT. Side view. Field of view ~ 2 cm. The false color scale represents the density of expanded Dy cloud 5 ms (a) and 38 ms (b) after release from trap. The atom number is 1.6×10^8 and temperature is $\sim 10 \mu\text{K}$. Note the "smiling face" shape of cloud at small TOF. It reflects the in situ MOT shape due to the resonant condition described in text.

optimization is: 9×10^7 atoms (^{164}Dy) trapped at $\sim 1 \mu\text{K}$ with a phase-space density close to 1×10^{-4} . The resultant ultracold gases have a spin purity very close to 100% as far as we can tell within the SNR in our Stern-Gerlach experiments.

We note that this 741 nm MOT is repumperless, because it employs an optical transition that is truly closed (see Fig. 6.1). For fermions, the other hyperfine manifolds are dark states. However, the large hyperfine splittings (a few hundreds MHz to GHz) effectively reduced this loss due to the large detuning-to-linewidth ratio.⁴ This is verified from the experimental observation that the rMOT number ratio between bosons and fermions is close to those in bMOT, indicating the absence of this additional loss channel.

⁴The 1.8 kHz linewidth is almost six orders of magnitude smaller than the detunings.

6.3.4 Dy 741 nm MOT cooling limit and ODT loading

We note that the Doppler limit of 741 nm transition is 43 nK and the photon recoil limit is 213 nK (Table 6.1), so this transition is in a regime where the Doppler limit is lower than the recoil limit. Earlier Sr experiments showed that the ultimate temperature limit would be half the recoil limit [19]. This indicates that Dy 741 nm narrow-line MOT may in principle reach a temperature of ~ 100 nK, which has not yet been realized in experiments. Most likely, this is due to the exothermic dipolar relaxations (see Chapter 8) that occur for atoms pumped to the low-field-seeking state $m_J = 8$ or $m_F = 21/2$ in this blue-detuned MOT. The competition between cooling and heating mechanisms determines the final MOT temperature which would be higher the theoretical limit.

We also note that the laser's coherence is crucial to this very-narrow-line MOT. While at University of Illinois, we prepared a 10 μ K MOT [5], which was later proved to be limited by the laser linewidth which was estimated to be in the low tens of kHz range. At Stanford University, the laser system has been upgraded so that the laser linewidth is narrower than the natural linewidth of 741 nm transition, which has allowed us to laser cool Dy to the sub-microKelvin regime.

Atoms in the other stretched state $m_J = -8$, which are free from dipolar relaxation, could in principle be trapped in the 741 nm narrow-line MOT [113]. However, laser detunings and MOT beam polarizations have to be changed accordingly, compared to the current blue detuned mode. Since this state is a high-field-seeking state, atoms would experience an anti-trapping background potential from the quadrupole field gradient and a small trapping dimple formed by the interplay between the mag-

netic field and cooling beams. This combination would produce a less stable MOT, and we have not attempted it.

Though the 741 nm MOT is highly effective in cooling Dy atoms to very low temperatures, the temperature of the atoms in the optical dipole trap immediately after the loading from the narrow-line MOT is limited to 5 μ K, which is considerably hotter.⁵ The two reasons for this are likely: the density in the ODT is higher than that of MOT, leading to a faster dipolar relaxation which isn't inhibited until we perform the ARP transfer to the high-field-seeking state;⁶ the large light shift (hundreds of kHz) perturbs the narrow-line cooling which itself is based on a 2-kHz-wide transition. Indeed, we noticed the polarization of ODT beam influences the loading efficiency considerably: we found linear polarization in a direction parallel to the local magnetic field where MOT is located is favorable for ODT loading. In our configuration, the ODT beam is perpendicular to the axis of quadrupole field and crosses the center of the MOT. A possible explanation could be that other polarizations added m -dependent vector or tensor light shifts perturbing the Zeeman shift in such a way that the blue-detuned MOT cooling mechanism is less efficient. Further work is needed to elucidated the subtle interplay between the narrow-line MOT and the scalar, vector, tensor light shifts due to the high-power ODT beam.

While the major figures of merit of this blue-detuned MOT—atom number and temperature—are insensitive to laser detuning in a fairly broad range of MHz, the location of the MOT is sensitive to laser detuning. Loading efficiency into the fixed-

⁵Due to the enhancement of the real space density in the ODT, the phase space density suffers only a factor of two in the ODT after the transfer.

⁶The fact that ODT loading takes hundreds of ms prevents us to spin-transfer the atoms to high-field-seeking state $m = -8(-21/2)$ immediately.

position ODT crucially depends on the optimal overlapping between the MOT and ODT beam(s). A MOT position sensitivity of $2\mu\text{m}/\text{kHz}$ is therefore technically challenging when the ODT beam has a small waist and laser frequency is drifting. The upgrade to our optical frequency standard (see Chapter 3) reduces this effect to a level so that it is no longer the bottleneck of instability for our current Dy experiments.

Chapter 7

Optical Dipole Force Trapping

7.1 Dynamic polarizability and dipole force trapping

Atoms are polarizable in an external electric field (dc or ac). The induced electric dipole moment $\langle d \rangle = \alpha E$ of an atom is proportional to the polarizability α at a given electric field strength E .¹ The value of α depends both on atom species and field frequency. We consider a simple model to provide some intuition about the polarizability's order of magnitude [116]. Imagine a static electric field separates an electron and a nucleus apart by one Bohr radius a_0 for a hydrogen atom in ground state. The field strength E is $e/4\pi\epsilon_0 a_0^2$, as set by the Coulomb force. The polarizability of a hydrogen atom is therefore

$$\alpha = \frac{ea_0}{e/4\pi\epsilon_0 a_0^2} = 4\pi\epsilon_0 a_0^3 \quad (7.1)$$

¹We assume here that α is a scalar, which is independent of direction. Tensor terms are usually relatively small for most atoms including Dy [115] Also we limit our discussion to the weak field regime (The situation in an intense field is not considered).

In atomic units (a.u., in which $a_0 = 1$, $1/4\pi\epsilon_0 = 1$), this polarizability is unity. This very crude classical approximation is not bad, as the rigorous quantum treatment gives an exact analytical value of $9/2$ [66]. For the many-electron atom Rb ($Z=37$), the static polarizability of the ground state is about 319 (a.u.) [117].²

In an ac electric field, the induced atomic dipole moment will oscillate with the external field oscillating at angular frequency ω . In a quasi-static driving field, d is in phase with E , so the energy associated with the dipole $H = -\langle d \cdot E \rangle = -\frac{1}{2}\text{Re}(\alpha(\omega))E^2 < 0$ within the complex representation $\alpha(\omega)$. This leads to an attractive dipole force towards the position of the field maximum. This attractive force is the origin of the optical dipole force trap (ODT). At very high frequencies, electrons cannot respond to the external field fast enough, and when the induced dipole lags behind the field by more than 90° , the dipole force becomes repulsive.

The polarizability $\alpha(\omega)$ can be calculated by time-dependent quantum perturbation theory and the associated interaction potential of an induced dipole is [118]:

$$U_{\text{dip}}(\mathbf{r}) = -\frac{3\pi c^2}{2\omega_0^3} \left(\frac{\Gamma}{\omega_0 - \omega} + \frac{\Gamma}{\omega_0 + \omega} \right) I(\mathbf{r}), \quad (7.2)$$

where ω_0 and Γ are the frequency and decay rate of the transition in a two-level system and $I \propto E^2$ is light intensity in the optical frequency regime. Indeed, we see that when ω is smaller than ω_0 , the conservative potential can be used for atom trapping at the spot with maximum light intensity I .

The imaginary part of the complex polarizability $\alpha(\omega)$ leads to a dissipative force

²This is not orders of magnitude off from the naive scaling of electron number based on the hydrogen atom, assuming electrons have no interactions with each other, though such a guess could be quite wrong in predicting the trend, e.g., Dy ($Z=66$) has much smaller DC polarizability than that of Rb—correlations, screening effects among electrons do matter.

as one can see the power that the atom absorbs from the driving field is:

$$P = \langle \dot{d}E \rangle \propto \text{Im}(\alpha)I, \quad (7.3)$$

where $\dot{d}$ is the time derivative of dipole d , from which the photon absorption rate (or photon scattering rate) is determined by [118]:

$$\Gamma_{\text{sc}}(\mathbf{r}) = -\frac{3\pi c^2}{2\hbar\omega_0^3} \left(\frac{\omega}{\omega_0}\right)^3 \left(\frac{\Gamma}{\omega_0 - \omega} + \frac{\Gamma}{\omega_0 + \omega}\right)^2 I(\mathbf{r}) \quad (7.4)$$

Such inelastic scattering can heat up ultracold atoms in an ODT, and therefore it should be reduced as much as possible. From eqn. 7.2 and 7.4, it is clear that by detuning the frequency far away from resonance and increasing the light intensity, the trapping potential depth is maintained while harmful scattering is reduced.

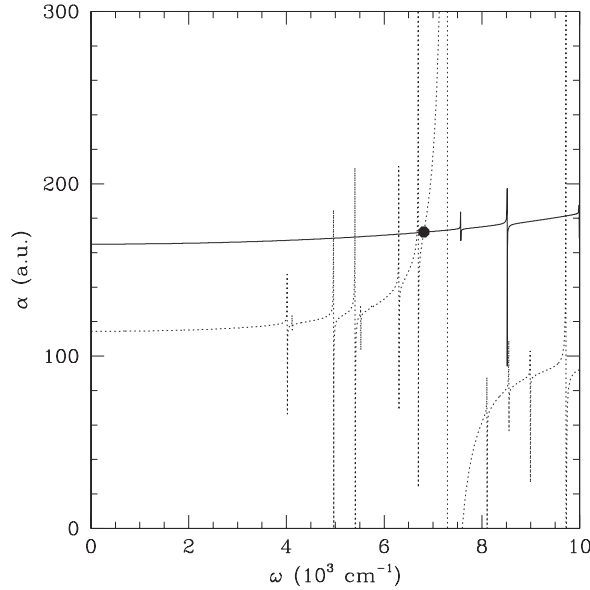


Figure 7.1: Dynamic scalar polarizability α of the ground state (solid) and 741 nm excited state (dotted) of Dy. Two lines cross at "magic" wavelengths (frequencies) [119] of 741 nm transition where light shifts are the same for both states. The plot is from [120].

It is straightforward to extend eqn. 7.2 to multi-level systems by directly summing the contributions of all electric dipole allowed transitions [116, 120]. The relative importance of each transition depends on the frequency detuning as well as transition linewidth as we see in eqn. 7.2. For Dy atoms, strong optical transitions lie in the blue and UV regimes, so optical frequencies in the NIR regime are effectively red-detuned, and the net potential is negative (or equivalently, $\alpha > 0$) for dipole force traps, as is shown in Fig. 7.1. As one can see, the predicted polarizability of Dy from the quasi-static regime up to NIR wavelength of 1 μm is between 100-200 a.u., which is relatively small. By comparison, Rb has a polarizability of 770 a.u. [117] at 1064 nm. Considering the availability of high-power commercial lasers, 1064 nm YAG or Yb doped fiber lasers are natural options³ for an ODT, with a relatively low scattering rate. For the first demonstration of a Dy ODT and the following evaporative cooling in the crossed ODT for generating quantum gases, we use two fiber lasers with outputs of 20 and 30 W respectively. When the Dy machine was upgraded at Stanford University, we switched to two 50-W fiber lasers.

7.2 ODT for Dy at 1064 nm

Experimentally, a focused Gaussian beam conveniently provides a local light maximum at the focus. The trapping potential close to the center can be expanded in the harmonic trap form [118]:

$$U = -U_0[1 - 2(r/w_0)^2 - (z/z_R)^2] \quad (7.5)$$

³The frequency-doubled Nd:VO4 solid state laser with an output at 532 nm is another choice for higher polarizability, but the scattering rate is also considerably higher compared to a 1064 nm laser. We chose to start with 1064 nm for the first experimental attempt.

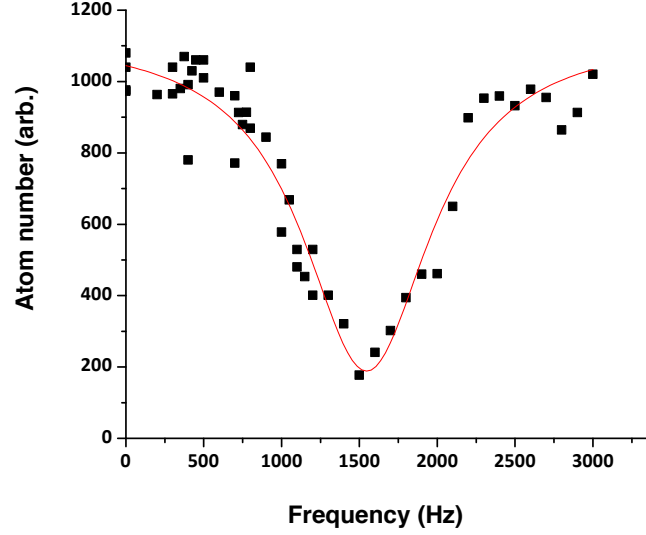


Figure 7.2: A typical parametric heating spectrum in 1064 nm ODT. Red line is a fit to Lorentzian functional form.

where U_0 is the trap depth, r and z the radial and axial displacement from the center, w_0 the waist at focus and $z_R = \pi w_0^2 / \lambda$ the Rayleigh length. Comparing with a harmonic potential $\frac{1}{2}m\omega^2 r^2$, the corresponding trap frequencies along r and z are:

$$\begin{aligned}\omega_r &= \sqrt{\frac{4U_0}{mw_0^2}} \\ \omega_z &= \sqrt{\frac{2U_0}{mz_R^2}}\end{aligned}\tag{7.6}$$

Since trap frequencies are directly related to many important physical quantities in cold atom experiments, we directly determine them from parametric heating experiments. The residual atom number after a fixed holding time in an intensity modulated ODT is recorded as a function of modulation frequency, as shown in Fig. 7.2. Atom loss is maximized at frequencies $\omega = 2\omega_0/n$, where ω_0 is the trap frequency and n is an integer [121]. Theoretical calculations indicate the peak corresponding to the

highest frequency $\omega = 2\omega_0$ is the strongest [121] at a fixed modulation depth, as is shown in experiment (Fig. 7.2). The trap frequencies are usually determined within a few percent relative uncertainties in such an experiment. Because the longitudinal trapping frequency is often too low (~ 10 Hz) in a single-beam ODT, we use a crossed ODT for trapping cold atoms.

From eqn. 7.6, one can deduce the trap depth and the dynamic polarizability given known laser intensities. At University of Illinois, we noticed that the experimentally derived α at 1064 nm is systematically smaller [5] than the theoretical result by Dzuba, *et.al.* based on relativistic configuration interaction method (CI) [120] (Fig. 7.1). At Stanford University, we improved the experimental setup with better beam quality control and measurement precision. Our recent experiment indicates a value of 75 a.u. (with a fairly conservative uncertainty estimation of 20%⁴) at 1064 nm, while Fig. 7.1 from [120] gives $\alpha \sim 180$ a.u. at 1064 nm, and their recent work [122] offers a value of $163 \pm 15\%$ a.u. for the dc polarizability. The cause of this inconsistency is unknown so far, as we have checked all systematic errors, including the laser powers, beam waists, beam alignment and polarization.⁵ Another independent way to estimate the polarizability is based on atomic spectroscopy. Starting with the well-documented spectroscopic data of 39 Dy I transitions ranging from NIR to 286 nm [123],⁶ we

⁴This is mainly limited by the error in determining waists of the ODT at $\sim 20 \mu\text{m}$, which could be improved in future. The experimental uncertainty is far below the difference between experiment and theory.

⁵Any possible vector or tensor polarizabilities are predicted to be at percent level of scalar light shift, as calculated in Er [115], which is also consistent with the fact that we did not observe any noticeable change in trapping frequencies within experimental errors as we varied polarizations of ODT beam.

⁶They are the ones to the ground state. The transition rates were obtained from literature listing measurements of the decay rate of excited states calibrated by branching ratios determined independently, with 5% uncertainty claimed in the literature.

directly calculated the scalar polarizability using the formula in [120]. We obtained values of 167 a.u. at dc, 198 a.u. at 1064 nm and 470 a.u. at 532 nm. These values should be considered as a lower bound of α as it is hard to find all experimental data for high-excited states of Dy including unbounded states when using this method. Preliminary dipole force trapping at 532 nm seems to be weaker than predicted again, as in the case of 1064 nm. We note that the Er ultracold atom experiment found a similar inconsistency in values of α between experiment and theory [122].

Chapter 8

Ultracold Collisions of Highly Magnetic Atoms

8.1 Introduction

Understanding inter-particle interactions in the cold and ultracold regimes [124],¹ are crucial in cold atom experiments. Interactions (through ultracold collisions) influence the trap stability as well as collective excitations of quantum gases [125]; it leads to the rich physics of fermionic pairing [126]; it offers a window for studying the few body physics that shows beautiful universality [127, 128]; and it provides a platform for a wide spectrum of many-body quantum simulation in ultracold atoms, which has become one of the mainstreams of atomic experiment [129]. Last but not least, it is crucially important to evaporative cooling, which is the only general route

¹Following the convention of [124] we distinguish cold and ultracold regimes from the collisional point of view. In the ultracold regime, the degree of angular motion between collisional partners is frozen and only radial motion is maintained (the *s*-wave scattering in partial wave expansion). However, as we shall see, this concept should be modified for collisions due to dipole-dipole interactions.

to prepare large ensembles of quantum gases, so far.

This chapter is devoted to the collisional properties of Dy and highly magnetic atoms in general. We do not intend to list all of the important and interesting phenomena in this topic, rather we present reviews on some key ingredients that are important in experiments of ultracold Dy gases. Some of them are fairly recent and subject to ongoing experimental tests. We begin with a general framework for describing elastic and inelastic collisions, followed by sections describing the unique elastic scattering and inelastic scattering due to the long-ranged dipole-dipole interaction. A possibility to achieve metastable fermionic Dy gases in different spin states or a spin mixture is discussed. Other two-body loss mechanisms are mentioned and three-body loss for Dy is explored in the initial stage of experiments. At the end, we discuss some new features in Feshbach resonances for Dy isotopes.

This chapter is related to our experimental work reported in [17, 29].

8.2 Overview: good or bad collisions

For dilute atomic gases, the inter-particle interactions arise from elastic or inelastic collisions. Elastic collisions do not change the internal states of atoms or the external total kinetic energies of atoms before and after collisions, but may change the thermal distribution of atoms statistically. Inelastic collisions change internal states of atoms, and the associated change in internal energy is released (absorbed) to (from) external kinetic motion. At energy E , the elastic collisional cross section in an open channel:²

²A channel is a collisional state with specified internal atomic/molecular states and external motional states of the atoms undergoing a collision.

α is [130]

$$\sigma_{el,\alpha} = g_\alpha(\pi/k^2) |1 - S_{\alpha,\alpha}|^2, \quad (8.1)$$

and the loss from channel α in terms of inelastic cross section is:

$$\sigma_{inel,\alpha} = g_\alpha(\pi/k^2)[1 - |S_{\alpha,\alpha}|^2], \quad (8.2)$$

where $g_\alpha = 1(2)$ for distinguishable (indistinguishable) particles, $\hbar k$ is relative momentum of two atoms and $S_{\alpha,\alpha} = \exp(2i\eta_\alpha)$ the scattering matrix taking the unitary matrix form. At the low energy limit, the phase shift $\delta_\alpha(E) \rightarrow \text{atan}^{-1}(-ka_\alpha)$ where a_α is the s -wave scattering length, an important length scale [67]. Assuming α is a real number, the S matrix is unitary. We immediately find that the elastic cross section $\sigma_{el,\alpha}$ is bounded and the maximum value possible is determined by k^{-2} . This situation corresponds to the so-called unitarity regime.

To incorporate loss mechanisms, a complex value of the scattering length $\tilde{a}_\alpha = a_\alpha - ib_\alpha$ could be introduced [130], with real numbers a and b . The S matrix is no longer unitary.³ Note that the collisional rate is $K = \sigma v$ where relative velocity $v = \hbar k/\mu$ for atoms with reduced mass μ . Substituting the S matrix with functions of a and b , in the low energy limit ($k \rightarrow 0$) we have the scattering cross sections and

³In quantum mechanics, a unitary transformation is linked to the conservation of total probability of finding matter in all states, and a violation is the manifestation of leaks outside the Hilbert space considered (usually a subspace of the full Hilbert space).

collisional rates:

$$\sigma_{el,\alpha} = 4\pi g_\alpha (a_\alpha^2 + b_\alpha^2) \quad (8.3)$$

$$\sigma_{inel,\alpha} = 4\pi g_\alpha b_\alpha / k \quad (8.4)$$

$$K_{el,\alpha} = [(2h/\mu)g_\alpha (a_\alpha^2 + b_\alpha^2)]k \quad (8.5)$$

$$K_{inel,\alpha} = (2h/\mu)g_\alpha b_\alpha \quad (8.6)$$

This shows that the elastic collisional rate among atoms decreases while the inelastic collisional rate approaches a constant at the low temperature limit.⁴ In some cases, this deteriorating ratio of elastic-to-inelastic collisional rates (experimentalists call this the ratio of good-to-bad collisions) prevents further evaporative cooling beyond some temperature. It is imperative to suppress $\sigma_{inel,\alpha}$ and enhance $\sigma_{el,\alpha}$ by intelligently choosing the right collisional channel α , the suitable atomic isotope species or by taking advantage of Fano-Feshbach resonances in some cases. For Dy atoms, the dipole-dipole interactions change both σ_{el} and σ_{inel} drastically.

8.3 Dipolar elastic scattering

In this section, we limit our discussion to elastic scattering and drop the subscript α . A scattering process in the central force potential can be expanded in different terms based on angular momenta l , as described in textbooks, e.g., [67]. This partial wave expansion is natural in that l and m_l are good quantum numbers under spherically rotational symmetry. Each term contributes to the total cross section

⁴The underlying assumption is that a and b are k independent in $k \rightarrow 0$ limit. It is true for most atoms, but the story becomes complicated for dipolar fermions, as we shall see in later sections of this chapter

independently in the relation:

$$\sigma = \frac{4\pi}{k^2} \sum_{l=0}^{\infty} (2l+1) \sin^2 \delta_l, \quad (8.7)$$

where the phase shifts δ_l vary as k^{2l+1} as $k \rightarrow 0$ for a finite-ranged potential. Lower partial waves become more important and all non- s partial waves vanish (Wigner threshold law). Often the potential varies as $1/r^n$ at interatomic separation r , in which case the scaling above holds for $l < (n-3)/2$, while for higher partial waves the correct scaling is k^{n-2} [116]. As for a van der Waals potential ($n = 6$), d, f, g and all higher partial waves have the same k^4 scaling in phase shifts. The p wave also vanishes at $k \rightarrow 0$, but in at slower rate. However, for dipolar scattering ($n = 3$) all partial waves have phase shifts scaled as k and a very important result arises: $\sigma_l(k \rightarrow 0) \neq 0$.

The conclusion above is understandable semiclassically. For non- s partial waves, atoms undergo rotations with respect to each other and centrifugal barriers prevent them from approaching each other. With lower and lower collisional energies, atoms stay farther away outside the centrifugal barriers and probe smaller portions of the tail of the $-C_6/r^6$ van der Waals potential, where C_6 is a constant. For a relatively slowly decaying $-C_3/r^3$ potential where C_3 is a constant, ultracold atoms can still probe considerable potential in the regions that extend far outside the centrifugal barriers, leading to a less sensitive scaling of phase shift on E (or k). Because of the non-vanishing higher partial waves at the collisional threshold ($E \rightarrow 0$), the differential cross section is not isotropic any more, rather, it takes the form of $\sim [\log(|\vec{k}' - \vec{k}|b)]^2$ where $\hbar\vec{k}, \hbar\vec{k}'$ are the input and output relative momenta and b is some truncation parameter to avoid a divergence of $1/r^3$ at short distances [131]. It arises solely due

to the long-ranged nature of the interaction potential, and no underlying symmetry is implied. The anisotropic dipole-dipole interaction $(1 - \cos^2\theta)/2r^3$ belongs to this class of collision, with an additional feature of being a non-central force potential. As one can show, the Fourier form of the dipole-dipole interaction, which is essentially the scattering amplitude in the Born approximation [67], is $\tilde{U}_{dd}(\vec{K} = \vec{k}' - \vec{k}) = C \times (\cos^2\alpha - 1/3)$ where α is the angle between $\vec{k}'$ and $\vec{k}$ [15] and C is some combination of physical constants. This clearly shows the angular dependence in the scattering which one expects. Additionally, the scattering is independent of the modulus of $\vec{k}$ which means the anisotropy in scattering persists at all energy scales ($E \propto k^2$). Such a behavior implies some kind of scaling invariance in distance ($1/k$) which may be regarded as another manifestation of long-ranged nature from scattering point of view.⁵

The dipolar interaction breaks the spherical rotational symmetry, and hence the l partial waves are not independent. Coupling between different partial waves leads to coupled differential equations in the scattering problem, and concepts of phase shifts, scattering matrix (usually in a scalar form) and scattering amplitude are now matrices, not scalars. Solving coupled-channel Schrödinger equations [132] for a given scattering potential determines the matrices $\delta_{l,l'}^m$ or $S_{l,l'}^m$ that completely fix the scattering problem. We note that between aligned dipole moments, the dipole-dipole

⁵The term long-ranged and short-ranged interactions are sometimes confusing in different contexts. Relative to the electron exchange interaction taking place at atomic-scale distances which is responsible for chemical bondings, the van der Waals interaction is a long-ranged interaction. Compared to dipolar interaction, van der Waals interaction is however short-ranged, since it decays relatively faster. One generally accepted criterion is to see the divergence (convergence) of the space integral of the potential considered. According to this, dipolar interaction is long-ranged interaction in 3D and short-ranged in lower dimensions, while 3D is the critical dimension. This is discussed in [15]. We limit our discussion to 3D throughout this chapter.

interaction is $V_{dd} \propto (3\cos^2\theta - 1)/2 \sim P_{2,0}(\cos\theta)$, where θ is the angle between inter-atomic axis and the spin-aligned direction, and only channels with $l' - l = 0, \pm 2$ are coupled; hence the matrix is sparse.

Some dimensional analysis would be helpful in estimating the cross section. Recall that for the van der Waals potential $-C_6/r^6$, the characteristic length is $R_{\text{vdW}} = (MC_6/\hbar^2)^{1/4}$ and correspondingly a characteristic energy is $E_{\text{vdW}} = \hbar^2/2MR_{\text{vdW}}^2$ [130]. While the s -wave scattering length a can vary widely due to unknown boundary conditions (short-range physics dependent) in the scattering problem, R_{vdW} sets the general magnitude of $|a|$ (on the order of 10 nm in general, rather than e.g. 10 μm). E_{vdW} sets the scattering energy below which s partial wave becomes dominant. In a similar way, for the dipolar interaction C_{dd}/r^3 ,⁶ the characteristic length⁷ $D = M\mu^2/\hbar^2$ sets the scale of the scattering cross section D^2 below the energy scale $E_D = \mu^2/D^3 = \hbar^6/M^3\mu^4$. Because of its importance, D has a name called dipolar length in literature. By adding contributions from all partial waves term by term,⁸ numerical calculations within the Born approximation [132] show that all odd (even) non- s partial waves⁹ contribute to total cross sections of 3.351 (1.117) D^2 for distinguishable particles. The Born approximation fails for s -wave scattering which is sensitive to short-ranged physics. Therefore phenomenologically a term of $4\pi a_s^2$ is added to the total cross section of distinguishable bosons, where the value of a_s is to

⁶ $C_{dd} = \mu^2$ or d^2 in Gaussian units for magnetic or electric dipoles. Also we neglect angular-dependent part for now

⁷To avoid confusion, we reserve μ for magnetic dipole moment and M for reduced mass. There are variations in the prefactor of D , e.g., [132] uses 1, [15] uses 1/3, [133] uses 2, and we use the definition of [132].

⁸The convergence is slow, though higher order partial waves clearly contributes less and less.

⁹This should be understood as coupled partial waves. Because of parity, odd couples to odd and even couples to even. The two classes do not mix each other.

be determined.¹⁰ It should be understood that our discussion above is limited to the threshold scattering. Beyond the characteristic energy the total dipolar cross section decreases as D/k , predicted by the Eikonal approximation and verified numerically in [132]

For Dy, $D = 196 a_0$ (equivalent to that of an electric dipole of 0.1 Debye) and $E_D = 55 \mu K$. The large value of E_D means that throughout the whole conventional ultracold regime ($E < E_{\text{vdW}}$), dipolar scattering is universal for Dy. Particularly for identical fermions, which normally forms a collisionless ideal gas in the ultracold regime, this relatively large value of cross section is $\sigma_D = 6.7D^2 = 7.2 \times 10^{-12} \text{cm}^2$ (as a benchmark, for ^{87}Rb , $\sigma_s = 7.1 \times 10^{-12} \text{cm}^2$) which provides a decent elastic scattering rate at typical atom densities in ultracold atom experiments. Another prediction is that σ_D is independent of species (as called "universal"), so ^{161}Dy and ^{163}Dy would have essentially the same scattering properties, if we neglect the tiny differences in D due to mass dependence. For Dy bosonic isotopes, however, the total cross section is still species dependent, because the s-wave scattering term is not known a priori. When $a_s = 58a_0$, the contribution from s-wave scattering is equal to that from high partial waves terms (i.e., those partials waves greater than s) due to dipolar scattering ($2.2D^2$). If $a_s(B)$ is tuned to be zero via Fano-Feshbach tuning [130], the residual part is solely due to the universal dipolar scattering, which is still considerable. In other words, Dy bosons would not behave as an ideal gas at zero scattering length, as alkali or alkaline earth atoms do. As we shall see, this is what we found in our Feshbach experiments.

¹⁰To be exact it is the term corresponding to T_{00} where the T matrix is defined as $T_{l,l'} = i(S_{l,l'} - 1)$.

8.4 Dipolar inelastic scattering

The earlier discussion treats the particles as structureless magnetic dipoles aligned in the z direction during the dipolar scattering; therefore it is intrinsically elastic scattering. If the magnetic dipole has its origin in atomic spin in the familiar form of $\mu = g_s \mu_B \mathbf{s}$, where g_s is the Landé g factor of spin state s and μ_B is Bohr magneton, then generally the spins $\mathbf{s}$ may change orientation after a dipolar scattering, which leads to a change in quantum number m_s along the z direction (the quantization field's direction in the lab frame). Such an internal state change is the outcome of an inelastic scattering process which is called dipolar relaxation, and is very important experimentally.

The general form of the dipolar interaction between two spins $\mathbf{s}_1$ and $\mathbf{s}_2$ is:

$$U_{dd} = \frac{\mu_0(g_s \mu_B)}{4\pi r^3} [\mathbf{s}_1 \cdot \mathbf{s}_2 - 3(\mathbf{s}_1 \cdot \hat{\mathbf{r}})(\mathbf{s}_2 \cdot \hat{\mathbf{r}})] \quad (8.8)$$

where $\hat{\mathbf{r}}$ is the real space unit vector pointing from one atom to the other atom.

Expanding in the spherical basis, we have [116]

$$U_{dd} \propto \sum_{m=-2}^2 Y_{l=2,m}^*(\hat{\mathbf{r}}) \Sigma_{2,m} \quad (8.9)$$

where Y_{lm} is a spherical harmonic function and

$$\Sigma_{2,0} = -\sqrt{\frac{3}{2}}(s_{1z}s_{2z} - \mathbf{s}_1 \cdot \mathbf{s}_2/3) \quad (8.10)$$

$$\Sigma_{2,\pm 1} = \pm \frac{1}{2}(s_{1z}s_{2\pm} + s_{1\pm}s_{2z}) \quad (8.11)$$

$$\Sigma_{2,\pm 2} = -\frac{1}{2}s_{1\pm}s_{2\pm} \quad (8.12)$$

where $s_{\pm}$ are ladder operators to raise or lower the spin state m_s . From the $\Sigma_{2,m=0,\pm1,\pm2}$ terms we can see that dipolar scattering can conserve the total spin's projection, or flip one spin by $\Delta m_s = 1$ or flip two spins simultaneously. The net change in total spin projection is limited to no more than $\Delta m_s = 2$ and this spin angular momentum is transferred to the orbital angular momentum's projection m_l as indicated in eqn. 8.9. Therefore the z component of the total angular momenta of collisional pair is conserved. To restate this clearly, the selection rules for dipolar scattering that connects two channels $|m_{s1}, m_{s2}, l, m_l\rangle$ and $|m'_{s1}, m'_{s2}, l', m'_l\rangle$ are:

$$l' - l = 0, \pm 2; \quad (8.13)$$

$$m_{s1} + m_{s2} + m_l = m'_{s1} + m'_{s2} + m'_l \quad (8.14)$$

$$\max |m'_l - m_l| = 2 \quad (8.15)$$

This transfer of an internal spin degree of freedom to an external orbital degree of freedom resembles the Einstein-de Hass effect. The increase in orbital angular momenta of atoms in this mechanism is recently proposed to create vorticities for simulating quantum Hall effects [134]. Also, note that Zeeman energy is released or absorbed when the spin state changes in an external magnetic field, leading to the exotic demagnetization cooling technique [134].

To determine the influence of dipolar relaxation, the cross section of dipolar scattering for identical atoms in the same internal state is calculated in the Born approximation [92]:

$$\begin{aligned} \sigma = & \left(\frac{m}{4\pi\hbar^2}\right)^2 \frac{1}{kk'} \left[\int |\tilde{U}_{dd}(\mathbf{k} - \mathbf{k}')|^2 \delta(|\mathbf{k}''| - k') d\mathbf{k}'' \right. \\ & \left. \pm \int \tilde{U}_{dd}(\mathbf{k} - \mathbf{k}') \tilde{U}_{dd}^*(-\mathbf{k} - \mathbf{k}') \delta(|\mathbf{k}''| - k') d\mathbf{k}'' \right], \end{aligned} \quad (8.16)$$

where m is the atomic mass¹¹ $\mathbf{k}$ and $\mathbf{k}'$ are the initial and final relative wave vectors, $\tilde{U}_{dd}$ is the Fourier transform of dipolar interaction U_{dd} , and the sign $\pm$ relates to bosons (fermions) due to differing exchange symmetry.

The stretched state $m_s = S$ ($S = J = 8$ for Dy bosons and $S = F = 21/2$ for Dy fermions) are particularly interesting since they are collisionally stable against spin exchange processes. After dipolar relaxation, both atoms in initial stretched spin states can undergo 0, 1, or 2 spin-flips, and the corresponding total cross sections averaged over $\hat{\mathbf{k}}, \hat{\mathbf{k}}'$ are [92]:

$$\begin{aligned}\sigma_0 &= \frac{16\pi}{45} S^4 \left(\frac{\mu_0 (g_s \mu_B)^2 m}{4\pi \hbar} \right)^2 [1 \pm h(1)] \\ &= \begin{cases} \frac{32\pi}{45} D^2 & \text{for boson (+)} \\ \frac{32\pi}{15} D^2 & \text{for fermion (-)} \end{cases}\end{aligned}\quad (8.17)$$

and

$$\begin{aligned}\sigma_1 &= \frac{8\pi}{15} S^3 \left(\frac{\mu_0 (g_s \mu_B)^2 m}{4\pi \hbar} \right)^2 [1 \pm h(1)] \frac{k'}{k} \\ &= \left[\frac{32\pi}{15S} D^2 \right] \left\{ [1 \pm h(k'/k)] \left(\frac{k'}{k} \right) \right\}\end{aligned}\quad (8.18)$$

$$\begin{aligned}\sigma_2 &= \frac{8\pi}{15} S^2 \left(\frac{\mu_0 (g_s \mu_B)^2 m}{4\pi \hbar} \right)^2 [1 \pm h(1)] \frac{k'}{k} \\ &= \left[\frac{32\pi}{15S^2} D^2 \right] \left\{ [1 \pm h(k'/k)] \left(\frac{k'}{k} \right) \right\}\end{aligned}\quad (8.19)$$

where

$$h(x) = -\frac{1}{2} - \frac{3}{8} \frac{(1-x^2)^2}{x(1+x^2)} \ln \left[\frac{(1-x)^2}{(1+x)^2} \right] \quad (8.20)$$

is a monotonic function defined in $[1, +\infty)$ with $h(1) = -0.5$ which asymptotically approaches $1 - 4/x^2$ as $x \rightarrow +\infty$ (Fig. 8.1). The 1, 2 spin-flip cross sections are scaled

¹¹From now on, m and M are distinguished as the mass and reduced mass.

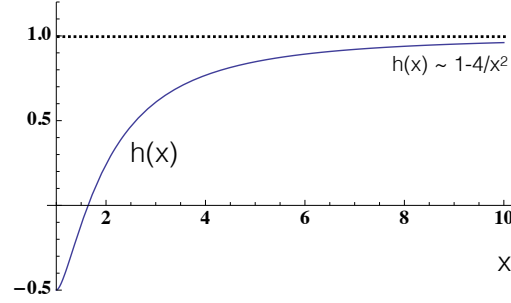


Figure 8.1: Function $h(x)$ in inelastic dipolar scattering. It is plotted in $[1, +\infty)$.

down by factors of $S \sim 10$ and $S^2 \sim 100$ compared to the zero spin-flip cross section respectively, indicating the single spin-flip process is the dominant loss mechanism and double spin-flip process is neglected in the following discussions. In eqns 8.18, 8.18, the last term $\frac{k'}{k}$ is the normal kinematics factor associated with inelastic scattering [116]. The middle $[1 \pm h(k'/k)]$ however, differentiates the behaviors of identical dipolar bosons and fermions drastically. In an external magnetic field, the ratio of output and input wave vectors in a dipolar inelastic collision is:

$$\frac{k'}{k} = \sqrt{1 + \frac{2m\Delta E_B}{\hbar^2 k^2}} \rightarrow +\infty, \quad (8.21)$$

as $k \rightarrow 0$ and/or Zeeman energy released $\Delta E_B \rightarrow +\infty$. In this limit, the two-body dipolar relaxation event rates are:

$$\mathcal{K}(E)_{1,B} = \sigma_{1,B} v_{rel} = \left[\frac{32\pi}{15S} D^2 \right] \left(2 \frac{k'}{k} \right) \left(\frac{\hbar k}{M} \right) \sim \sqrt{B} \text{ for boson (+)} \quad (8.22)$$

$$\mathcal{K}(E)_{1,F} = \sigma_{1,F} v_{rel} = \left[\frac{32\pi}{15S} D^2 \right] \left(4 \frac{k}{k'} \right) \left(\frac{\hbar k}{M} \right) \sim \frac{E}{\sqrt{B}} \text{ for fermion (-)} \quad (8.23)$$

In our experiment, the released Zeeman energy is higher than the trap depth and every collisional event leads to two atom loss, therefore the atom loss rate in a trapped cloud with temperature T should be $\beta = 2\langle \mathcal{K} \rangle_T$ (thermal average). The value of β influences

the density equation $d^2n/dt^2 = -\beta n^2 - \Gamma n$, where Γ is related to the life time of the cold atoms in trap. Both β and Γ can be determined experimentally. Under normal experimental conditions of temperatures and magnetic fields, $\mathcal{K}_{1,F} \ll \mathcal{K}_{1,B}$ which should be experimentally observable.

8.5 Other two-body loss and three-body loss

Unlike alkali or alkaline atoms, Dy is a non- s -state atom with total orbital angular momentum $L = 6$ in the ground state. Therefore the electron cloud is not spherically symmetric. The dispersion potential takes the forms of anisotropic electrostatic first-order quadrupole-quadrupole and second order dipole-dipole interactions (the van der Waals interaction) at long range, besides the anisotropic magnetic dipole-dipole interaction. The more complete potential curve including the centrifugal barrier takes the form of [93]:

$$V(R) \sim C_3/R^3 + C_5/R^5 - \frac{1}{R^6}(C_6^{iso} + C_{6,2}^{aniso} + C_{6,4}^{aniso}) + \frac{\hbar^2 l(l+1)}{2MR^2} \quad (8.24)$$

where $C_3 \sim 0.00266$ (*a.u.*) is the coefficient for the magnetic dipole-dipole interaction we discussed earlier, $C_5 = 6Q_{zz} * Q_{zz} = 1.6 \times 10^{-4}$ (*a.u.*) is rather small for quadrupole-quadrupole interaction which is deduced from quadrupole tensor $\mathbf{Q}$ [93] and hence negligible,¹² and the three terms in parenthesis are for van der Waals interaction. C_6^{iso} corresponds to the isotropic major part, $C_{6,2}^{aniso}$ and $C_{6,4}^{aniso}$ are the dipole-dipole and

¹² C_5 is small because in the long range, dipolar interaction dominates, while in the short range, van der Waals interaction dominates—there is no regime where C_5/r^5 term would be dominant. Also, C_3 and C_5 are given to offer some idea of magnitudes, with the angular dependence not explicitly expressed. One needs to keep in mind there are tensor terms in explicit forms.

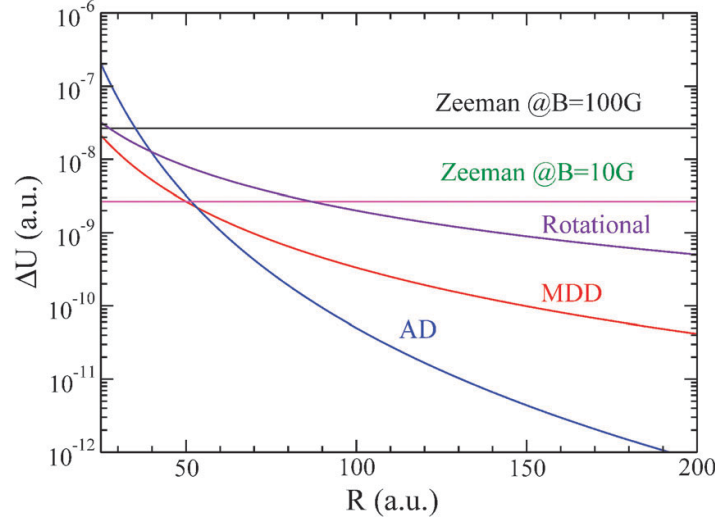


Figure 8.2: Level splitting due to the dominant interaction forces in atomic units as a function of interatomic separation. In atomic units the Zeeman splitting is $g_j B/2$, the splitting between $l = 0$ and 2 rotational levels is given by $6/(2m_r R^2)$, the splitting due to the magnetic dipole-dipole (MDD) interaction is $2\alpha^2 J(g_J/2)^2/R^3$, where α is the fine structure constant, and the anisotropic dispersion (AD) interaction is $\Delta C_6/R^6$, where $\Delta C_6 = 25$ a.u. Here m_r is the reduced mass in units of the electron mass. The plot is from [93].

quadrupole-quadrupole-like anisotropic coefficients. The $C_{6,2}^{aniso}$ and $C_{6,4}^{aniso}$ terms can lead to change in the total spin projection $M_{tot} = M_1 + M_1$ by 2 and 4 units for two Dy atoms in the $J = 8, M = 8$ state. In general, the major anisotropic coefficient is two orders of magnitude smaller than the isotropic coefficient ($C_6^{iso} \sim 1800$ a.u.) [93]. The size of C_6 corresponds to a mean scattering length scale of $\bar{a}_s = 77a_0$ [130] for Dy. At distances shorter than $\sim 100a_0$, the van der Waals interaction dominates over the magnetic dipolar interaction. Measurement of s -wave scattering lengths for the Dy bosonic isotopes would be carried out in the near future.

In an external magnetic field, spin states are separated by Zeeman splittings, and spin flips can happen when the energy difference in spin states due to

the anisotropy in the interatomic interaction matches the Zeeman splitting. Those anisotropies in dispersion curves have different distance dependence, as one see in Fig. 8.2. The competition can lead to different loss mechanisms at different magnetic field strengths. From the plot, dipolar interaction induced loss is expected to be dominant at low magnetic fields and low temperatures. Further experimental and theoretical research is needed to elucidate the collisional properties of ultracold Dy atoms, particularly in those spin states important for quantum simulation.

If we relax the constraint to the stretched states ($m_J = \pm 8$ for boson or $m_F = \pm 21/2$ for fermion) but still within the lowest Zeeman state manifold, there would be an additional inelastic process called spin exchange. Unlike dipolar relaxation which does not conserve the total spin of two colliding atoms, spin exchange redistributes the spins while conserving the total spin projection in z direction:

$$m_{s,1} + m_{s,2} = m'_{s,1} + m'_{s,2} = M_{\text{tot}} \quad (8.25)$$

It arises from the fact that two atoms with spins $\mathbf{s}_1$ and $\mathbf{s}_2$ can collide with different total spin S (with possible values of $|s_1 - s_2|, \dots, s_1 + s_2$), and each of S corresponds to a different molecular potential¹³ that leads to different scattering phase shifts. The existence of the off-diagonal terms in the interaction term of the Hamiltonian in the original basis $|s_1, m_{s1}; s_2, m_{s2}\rangle$ leads to the coupling between different spin states $|s, m_s\rangle$, while the total spin projection M_{tot} is still conserved.

A similar situation happens for spin-1/2 alkali atoms, where singlet and triplet electron potentials are very different and spin exchange processes are fast (rate $\propto |a_t - a_s|^2$). A notable exception is the ^{87}Rb where singlet (a_s) and triplet (a_t) s-wave

¹³In fact, for Dy, it is both s and Ω dependent, where Ω is the total spin project along the internucleus axis.

scattering lengths are almost degenerate accidentally [135]. In alkali atoms, spin exchange cross section is typically around $10^{-11} - 10^{-12} \text{ cm}^2$ based on the scale of scattering lengths, compared to that of magnetic dipolar relaxation¹⁴ at $\sim 10^{-15} \text{ cm}^2$. Because of the high magnetic dipole moment, the dipolar relaxation of Dy is $\sim 10^3$ - 10^4 times faster than alkali atoms and therefore is at the same magnitude as spin exchange process. In the Dy magnetic trapping for a spin mixture in low-field-seeking states $m_J > 0$, we observed a fast loss rate (see Chapter 5) at $200 \text{ } \mu\text{K}$ (few-partial-wave regime) that is consistent to a cross section of 10^{-11} cm^2 [17]. Fast spin exchange process likely contribute as well as dipolar relaxation. The spin exchange process differentiates the initial spin states of the two colliding atoms into a one with higher magnetic moment (higher m_J) and the other with lower magnetic moment (lower m_J). If the lower m_J drops below zero the atom becomes anti-trapped and is permanently lost. This cleaning work leads to higher mean value of m_j in the spin mixture and indeed the mean magnetic dipole moment $\bar{\mu} \propto \bar{m}_J$ of spin mixture was observed to be increasing in the magnetic trap [17]. This cleaning job would stop once the cloud is maximally spin polarized, but the additional exothermic dipolar relaxation would keep lowering the mean value of m_J from stretched state $m_J = J$, hence the cleaning and polarizing work from spin exchange would not stop. Since this two-body process is density dependent, the loss rate is highest at the center of the trap where the atoms with kinetic energies less than the average value of the thermal gas are removed with priority. This so-called "anit-evaporative cooling" phenomenon leads to a hotter clod

¹⁴The order of magnitude of the magnetic dipolar inelastic cross section is set by $\sigma_{inel} \sim S^{-1} \mu^4 (m/m_e)^2 r_0^2$ where S the atomic spin, m and m_e are atomic and electron mass and r_0 Bohr radius. For heavy atoms like Rb and Cs, the relativistic spin-orbit interaction modifies the dipolar relaxation rate based on this simple argument [116] There is also a discussion on order of magnitude of the ratio of spin exchange rate and dipolar relaxation [116].

which was also observed in our experiment [17].

For fermionic Dy isotopes, there are 6 hyperfine manifolds in the ground state with about 10-20 Zeeman sublevels in each manifold. The 741 nm MOT, working on the $F = 21/2 \rightarrow F' = 23/2$ cycling transition, pumps ^{161}Dy atoms in the maximally stretched Zeeman state $m_F = F = 21/2$ and the $F = 21/2$ manifold turns out to be the lowest hyperfine manifold. But for ^{163}Dy , the hyperfine hierarchy is opposite and the $F = 21/2$ hyperfine state is higher than other hyperfine manifold with GHz energy splitting.¹⁵ The severe hyperfine-changing inelastic collisions reduce the ODT trapped ultracold ^{163}Dy atom number by one order of magnitude after a holding time of only 1 second, at an initial density $\sim 10^{12} \text{ cm}^{-3}$, even though the atoms were prepared in the dipolar-relaxation-stable state $m_F = -F = -21/2$. Further work on spin state transfer to the absolute ground state is needed for effectively evaporative cooling ^{163}Dy .

An atom in the absolute ground state, which is free from two-body losses in general, is subject to three-body loss. The three-body loss arises when two atoms collide with each other while there is a third atom nearby. The first two atoms form molecular bound states below collision threshold and extra energy is released in form of kinetic energy that distributes between the newly formed molecule and the third atom. In this process the third atom acquires both energy and momentum to fulfill conservation laws. Since the molecular bound energy is normally orders of magnitude larger than the trap depth, all three initial atoms are removed from the trap. In the density regime of 10^{14} cm^{-3} , the lifetime of the Dy atoms in the absolute ground state is mainly determined by the three-body loss rate. Preliminary ^{164}Dy lifetime

¹⁵The hierarchy order of ^{163}Dy is similar to those of most of alkali atoms.

measurement in the $m = -8$ state is consistent with a three-body loss coefficient L in the low $10^{-29} \text{ cm}^6 \text{ s}^{-1}$ regime, when away from Fano-Feshbach resonances.¹⁶ Further detailed work is underway.

8.6 Homonuclear magnetic Feshbach resonances of Dy: a preliminary study

Fano-Feshbach resonance is a resonance phenomenon in the scattering cross section of cold atoms found by tuning the external static magnetic field or optical field. It arises when the energy of a bounded molecular state is degenerate with the energy of the entrance channel of two colliding atoms in the ultracold regime. It has been a powerful tool in tuning the interatomic interactions, enabling many quantum simulation experiments [130, 129]. Therefore it is very important to study the Feshbach resonances of Dy isotopes for the purpose of quantum simulation with this atom. Our preliminary study of homonuclear magnetic Feshbach resonances in the isotopes of ^{164}Dy , ^{162}Dy , ^{161}Dy and ^{160}Dy in low magnetic fields was reported in [29].

The origin of Feshbach resonances in Dy bosonic isotopes is somewhat special. For s -state alkali atoms with hyperfine structures, the isotropic electrostatic potential is diagonal in the partial wave quantum number l, m_l but generally is not diagonal to the atomic quantum states F, m_F of the entrance channel. Consequent couplings between different channels offer the possibility of strong and well isolated Feshbach resonances. For highly magnetic Dy (similarly for Er and other highly magnetic

¹⁶We follow the notation convention that the three-body loss coefficient is L for condensates and $6L$ for thermal gases due to the bunching effect [136].

lanthanides) atoms in the ground state with orbital angular momentum $L = 6$, strong anisotropy in the magnetic dipolar interaction and electrostatic dispersion potential mix different partial waves strongly [137, 26]. A large amount of rotational bound states associated with different rotational orbital ls can be coupled to the entrance channel; consequently a high-density Feshbach resonances emerge, even though there is no hyperfine structure in bosonic isotopes. Theory work [137] found that the emergence, widths and positions of those resonances are heavily influenced by the number of partial waves included in the first-principle coupled-channel calculation, which exemplifies the complicated nature of this problem.

Experimental work was carried out in optically trapped ultracold Dy isotopes at 100s of nK. High-resolution atom loss spectra were recorded by very finely scanning the magnetic field in the range of 0 – 6 G and observing the residual atom number after certain holding times. Experimental details can be found in [29]. A high density of Feshbach resonances in the form of loss peaks were unveiled, e.g., as shown in Fig. 8.3 for the isotope ^{164}Dy . The typical resonance density is 3 per Gauss, two order of magnitude higher than in alkali atoms. All of them have widths far below 1 G and most of them are on the order of 10 mG or even narrower. Another feature is that the number of Feshbach resonances increases $\sim 60\%$ as the temperature goes up from ~ 400 nK to ~ 800 nK, which remains deeply in the conventional ultracold regime, where, all non- s -partial waves are quenched and only s -wave entrance channel is important [130]. This may be understood in the following way: long-ranged scattering processes involve a large number of partial waves even at the zero temperature limit, though the weight of low- l partial waves are dominant [26]. At higher temperatures,

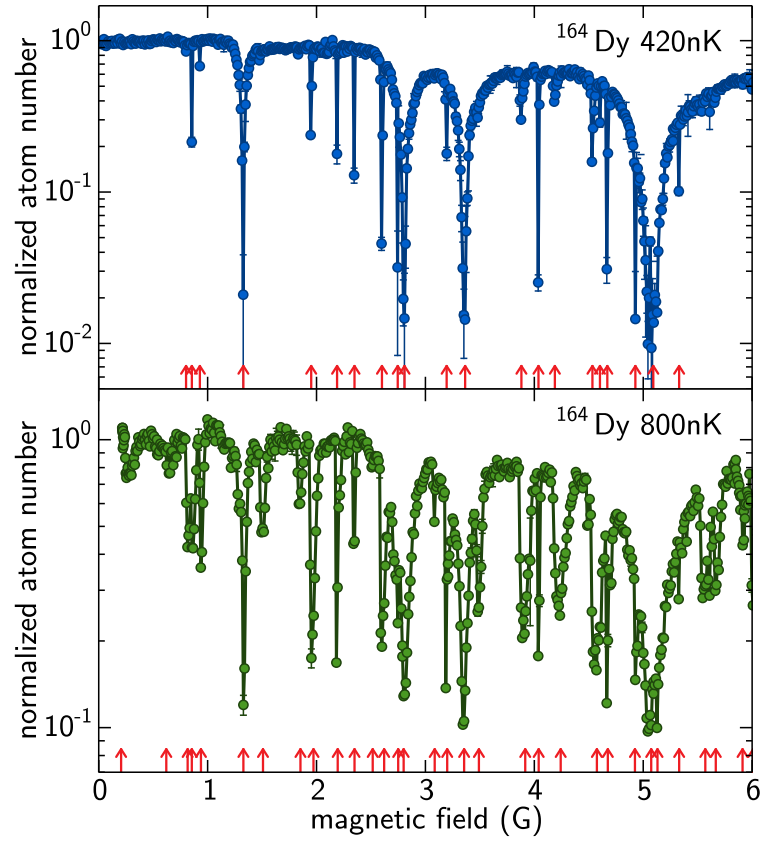


Figure 8.3: Atom-loss spectrum for bosonic ^{164}Dy in magnetic fields at 0 – 6 G. The top panel displays data taken with an ~ 420 nK sample which shows a large number of Fano-Feshbach resonances. The lower panel shows a measurement of the same isotope at ~ 800 nK with several resonances emerging and broadening. The arrows indicate the positions of the resonances.

more partial waves with higher- l become important and consequently their associated bound states have more statistical weight when they become resonant with the energy of entrance channel, resulting in the emergence of new Feshbach resonances experimentally observable and the broadening of others.

Other bosonic isotopes ^{162}Dy and ^{160}Dy show similar Feshbach spectrum qualitatively. For the fermionic isotope ^{161}Dy , the density of Feshbach resonances in $0 - 6$ G reaches 11 per Gauss, presumably due to the extra nuclear spin degree. No Gauss-wide resonance exists in this low-field regime, though in principle wide resonances should exist at some larger field for Dy fermions. Similar high-density Feshbach resonances were found recently in Er bosonic and fermionic isotopes, and the statistics in the experimental data are of the Wigner-Dyson form [138].

For weakly dipolar alkali-metal atoms, the van der Waals interaction-dominated scattering cross section σ_s exhibits a zero-crossing to the side of each Fano-Feshbach resonance peak associated with a vanishing scattering length a_s [130]. At this magnetic field, the weakly dipolar atomic gas is effectively non-interacting, thus inhibiting rethermalization ($\Gamma_{RT} \propto \sigma_s \rightarrow 0$) after a sudden, non-adiabatic change in the trapping conditions. This effect has been observed as a temperature peak versus magnetic field when a_s vanishes in the weakly dipolar systems of alkali-metal atoms [139].¹⁷

In contrast, the magnetic field dependence of the temperature, post-trap change, in the more strongly dipolar Dy gases studied here likely differs from this standard picture. Indeed, we do not observe a temperature peak near the Fano-Feshbach resonances. The large magnetic field-independent cross section of Dy, calculated to

¹⁷This effect has also been observed with Er [140] whose van der Waals interaction strength has been measured to be larger than the dipolar interaction strength away from resonances.

be $\sigma_{\text{DDI}} = 2.4 \times 10^{-12} \text{ cm}^{-2}$ [26], is expected to be of comparable magnitude to the cross section arising from the van der Waals interaction [93]. Though further experimental investigation is required, we argue that this dipolar-interaction-induced elastic cross section remains considerable near the resonances, thus minimizing the σ_s contribution to the total cross section σ_t , which, in turn, does not significantly alter the rethermalization rate.

Chapter 9

Bose-Einstein Condensation of an f-electron Atomic Gas

9.1 Introduction

The interplay between emergent electronic or spatial crystallinity, magnetism, and superfluidity is central to some of the most interesting materials of late, e.g., cuprate and iron-based superconductors, strontium ruthenates, and solid helium [2, 141, 142, 143]. Quantum degenerate gases possessing strong dipole-dipole interactions (DDI) [15] are thought to provide access to strongly correlated quantum phases involving quantum magnetism, spontaneous spatial symmetry breaking, and exotic superfluidity. Supersolid [144, 145, 146, 147] and quantum liquid crystal [7, 148, 149, 150] phases may be accessible using strongly dipolar Bose and Fermi gases; with the realization of such quantum gases, we may shed light on characteristics of these phases difficult to observe in their condensed matter settings [142, 143, 2].

We report the first realization of a strongly dipolar quantum gas at low field, a Bose-Einstein condensate (BEC) of ^{164}Dy , which is an atom with unsurpassed dipole moment $d = 10\mu_B$, where μ_B is the Bohr magneton. For comparison, Rb and Cr's moment equals $1\mu_B$ and $6\mu_B$, respectively. The Dy BEC is the first quantum gas of a highly complex, open *f*-shell lanthanide, and opens a new frontier for exploring scattering behavior involving DDI-induced universality [151, 26, 152, 153], electrostatic anisotropy [154, 93], and an open *f*-shell submerged under closed *s*-shells. These complex atomic properties evidently do not prevent Bose-condensation, and exploring such complex collisional physics will greatly aid attempts to understand collisional behaviors of ultracold polar molecules, whose Bose-condensation has yet to be achieved.

Unlike the ^{52}Cr dipolar BEC [4], we show that the ^{164}Dy BEC reaches the strongly dipolar regime $\epsilon_{dd} = \mu_0 \mu^2 m / 12\pi \hbar^2 a_s > 1$ without careful minimization of the *s*-wave scattering length a_s using a high-magnetic field Feshbach resonance.¹ The use of Feshbach resonances can lead to detrimental three-body loss [23] and precludes the study of ultracold dipolar physics near zero field. Moreover, ultracold dysprosium suffers no chemical reactions like certain polar molecules [155].

Short-wavelength optical lattices confining Dy are capable of exploring uncharted strongly correlated phases beyond the familiar Mott insulator at half-filling. Namely, density waves of various filling factors and lattice supersolids may now be accessible [15, 144, 145, 146, 156] without multilayer lattice enhancement [157]. Moreover, creation of unconventional and anisotropic superfluids [158, 159, 160] as well as explo-

¹For a zero-field (as yet unmeasured) a_s equal to ^{52}Cr 's $a_s \approx 100a_0$, $\epsilon_{dd}^{\text{Dy}} = 1.3$ while $\epsilon_{dd}^{\text{Cr}} = 0.15$. (a_0 is the Bohr radius.) $\epsilon_{dd}^{\text{Cr}} \approx 1$ has been achieved using a 589 G Feshbach resonance [23].

rations of 1D strongly correlated gases [161, 162], trap instabilities [15], spin textures and topological defects [163, 164, 165, 166], roton modes [167, 168], and emergent structure in layered dipolar gases [169, 170, 171] are among the phenomena now within the realm of experimental possibility.

9.2 Production of BEC of ^{164}Dy

The cooling procedure follows aspects of the Sr, Ca, and Yb BEC experiments [172, 173, 96, 97], and we only provide essential experimental details before briefly discussing some of the ^{164}Dy BEC properties. The BEC of ^{164}Dy , natural abundance 28.2%, is formed by forced evaporative cooling in a crossed optical dipole trap (cODT). The cODT is loaded with ultracold atoms from an unusual, blue-detuned narrow-line magneto-optical trap (MOT) [113] formed on the 1.8-kHz 741 nm line [16], the demonstration of which for Dy we report for the first time. This red-wavelength MOT (rMOT) is loaded by a repumperless blue-wavelength MOT (bMOT) formed on the broad, 421 nm transition. A Zeeman slower and transverse cooling stage, both at 421 nm, slow and collimate an atomic beam from a 1250 °C oven. Details regarding the bMOT are provided in Refs. [17, 102], and a subsequent publication will thoroughly describe the Dy rMOT and cODT.

Figure 9.1 depicts the relevant Dy energy level diagram with cooling and trapping lasers as well as a timing diagram for the experiment. The bMOT captures 5×10^8 ^{164}Dy atoms, 70% of which, in steady-state, are ~ 1 mK metastable atoms concurrently confined within the bMOT's magnetic quadrupole trap. We operate the bMOT at 1.4×10^8 total atoms for this work. Of these, 7×10^7 are captured in the rMOT—

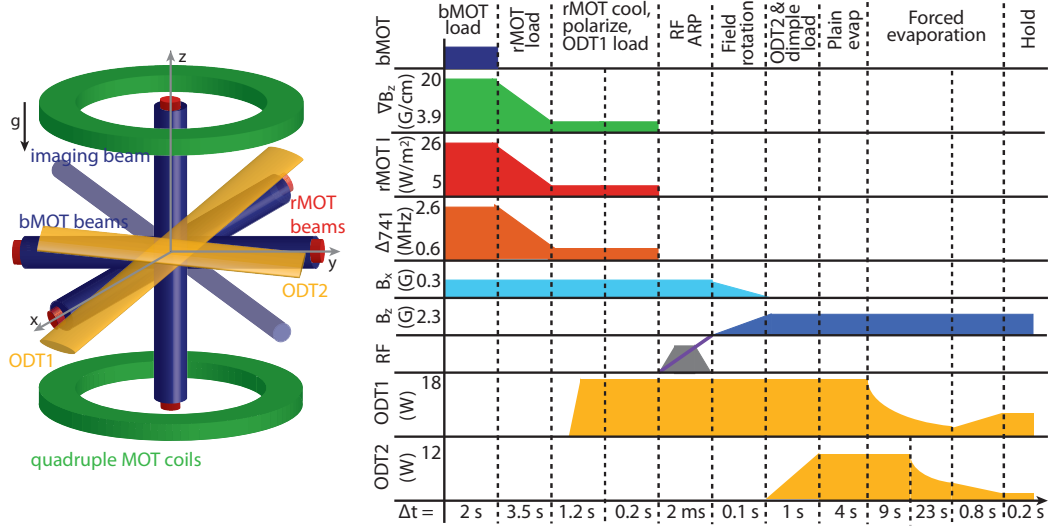


Figure 9.1: Schematics of BEC production setup and control sequence.

whose beams are overlapped with the bMOT's—during the loading phase lasting 3.5 s. Concurrently, metastable Dy decays to the ground state with a time constant of 2.3 s^{-1} [108].

The rMOT differs from most narrow-line MOTs, e.g., Ref. [19], in that the cooling laser is blue-detuned from resonance. For heavy, highly magnetic atoms possessing a narrow optical transition, a spin-polarized MOT may form below the center of the quadrupole field such that gravitational, magnetic, and optical forces balance each other. This technique, first employed for an 8-kHz-wide line in Er ($\mu = 7\mu_B$) [113], creates an ultracold, dense gas at a position determined by the laser detuning such that the magnetic field Zeeman-shifts the otherwise red-detuned maximally weak-field-seeking state (MWS) to the blue of the laser frequency. The cloud shifts position in the magnetic gradient as a function of laser detuning, but the laser linewidth need not be as narrow as the transition, as often the case for narrow-line MOTs.

The Er experiment captured 10-20% of the 10^5 atoms loaded into the Er bMOT [114] by fixing the rMOT laser intensity and detuning while ramping down the quadrupole field before recompression. We improve the capture efficiency of this technique by sweeping the laser's blue-detuning from $\Delta_{741} = +2.6$ MHz to $+0.6$ MHz while reducing the laser intensity (quadrupole gradient) from $I = 26$ W/m² ($\nabla B_z = 20$ G/cm) to 4.8 W/m² (3.9 G/cm).² We capture 50% of the total bMOT-cooled population by following this procedure. We hold this configuration for 1.2 s to cool to 12 μ K and spin-purify the gas in $m_J = 8$, as determined by subsequent Stern-Gerlach (SG) measurements. While ultracold, the gas does not reach the line's 84-nK Doppler limit; similarly, the Er rMOT's 2 μ K temperature was substantially larger than its 190-nK Doppler limit [113].

Reference [113] suggests that the seemingly hotter temperature arises due to momentum kicks from imperfectly extinguishing the quadrupole field before time-of-flight (TOF) measurements. However, these hotter temperatures may also arise from dipolar relaxation heating. Indeed, the plain evaporation of Dy after optical dipole trap loading implies the gas is no colder than 10 μ K when in the rMOT. Inelastic dipolar relaxation rates scale strongly with dipole moment and are expected to be quite rapid for Dy and Er.³ We speculate that the gas heats by releasing Zeeman energy as the MWS state relaxes to lower-energy Zeeman states, while the rMOT lasers re-cool and repump to the MWS state. An equilibrium in temperature and spin polarization is reached for a given ∇B_z , Δ_{741} , and I , and future numerical simulations

²The bMOT is unaffected by the rMOT lasers due to the ratio of transitions strengths at 421 nm and 741 nm by 18,000.

³Indeed, we observe in a 0.3 G field a 25-fold reduction of ODT trap lifetime in the MWS versus the maximally strong-field seeking state.

will investigate this process.

We employ two horizontal ODTs, ODT1 and ODT2, to form the cODT at a position beyond the edge of the rMOT. Intersecting the ODTs with the rMOT during loading repels the gas due to a ~ 10 -kHz Stark shift of the 741 nm line, which repositions the atoms in the quadrupole field to compensate the change in effective rMOT detuning. The optimal loading strategy thus involves setting the 18 W ODT1's propagation height roughly $r = 1$ mm below the rMOT center and fine-tuning the transfer efficiency with the 741 nm laser frequency while preserving spin-polarization and minimizing gas temperature.

The transfer efficiency from the rMOT to ODT1, as well as the initial gas temperature in the ODT1, is very sensitive to the value of r . During the rMOT cooling stage, 20-kHz drifts of the 741 nm laser frequency (linewidth 20 kHz) shift the cloud in z enough to decrease transfer efficiency. A Rb saturation-absorption reference laser stabilizes a transfer cavity to which the 741 nm laser is locked; together this provides sufficient frequency stability for BEC production.

Following this procedure, we obtain up to 4×10^6 atoms in ODT1 at a temperature of 10 μ K, which is $15 \times$ colder than the trap depth of ODT1. We experimentally determine the polarizability of Dy at 1064 nm to be 116 a.u. (atomic units) by measuring the ODT1 trap frequencies. This value is 65% smaller than recently calculated [120] and will be used to improve magic wavelength estimates for future Dy optical lattices.

ODT1 is derived from a 30-W 1064 nm fiber laser, while ODT2 is derived from a 25-W diode-pumped Nd:YVO4 1064 nm laser. Beams from both are intensity-controlled by acousto-optic modulators (AOMs), and each beam is shaped into a

cylindrical waist (e^{-2} radius) focused at the cODT center. The ODT1 has a waist of 30 (60) μm and trap frequency of 1 (0.5) kHz in z (ρ), where ρ lies in the x - y plane. ODT2 has a waist of 22 (70) μm and trap frequency of 1.2 (0.4) kHz in z (ρ).

After extinguishing the rMOT lasers and quadrupole field, we immediately spin-flip the atoms from the MWS-state to the maximally strong-field seeking (MSS) ground state to prevent heating from dipolar relaxation. No optical pumping is needed because, as mentioned above, the rMOT—unlike a traditional MOT—provides a MWS-state spin-polarized gas [113]. RF adiabatic rapid passage (ARP) flips the spin within 2 ms from $m_J = 8$ to $m_J = -8$ by using a $B_x = 0.3$ G magnetic field and a swept-frequency RF-driven coil along z . Stern-Gerlach measurements indicate that ARP produces a nearly pure, absolute ground state gas in $m_J = -8$, and we use such ARP sweeps and SG measurements to calibrate the applied magnetic fields and to zero the ambient magnetic field to <20 mG. The 0.3 G B_x field ($\Delta m = 1$ Zeeman energy equal to 25 μK) is then rotated to point along z and increased to $B_z = 2.3$ G (190 μK) to suppress thermally driven spin transfer to $m_J > -8$ states.

Within 4 s of loading Dy from ODT1 into the 340 μK cODT, formed by ramping ODT2 to 12 W in 1 s, plain evaporation both reduces the atom number to 1×10^5 at the cODT center and decreases the temperature to 2 μK . The trap frequencies are $[f_x, f_y, f_z] = [380, 500, 1570]$ Hz before the forced evaporation sequence begins. Power in the ODTs are reduced using the functional form $P_f = P_i / (1 + t/\tau)^\beta$ [174] using experimentally optimized parameters $\beta_1 = 2$ and $\tau = 15$ s for ODT1. After a 9 s delay, ODT2's power is reduced for 23 s using $\beta_2 = 1.3$ and $\tau = 15$ s. To recompress the trap and obtain an oblate shape— $f_z/f_\rho > 1$, with $f_\rho = \sqrt{f_x f_y}$ —both

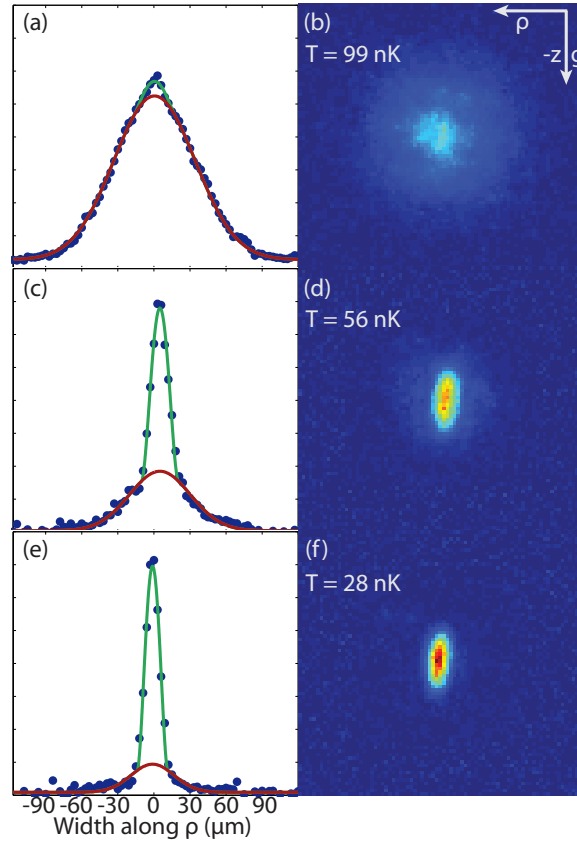


Figure 9.2: TOF profiles of the spin-purified Dy gas for three evaporation time-constants, with $\tau = 15$ s in (e) and (f). (a,c,e) Data at centers are fit to a parabolic profile (green), which underestimates condensate fraction, whereas the distributions' wings are fit to a Gaussian profile (red). (b,d,f) Absorption images of the emerging BEC. (b) The transition temperature is 99(5) nK, with condensate fraction 14.4(3)%; (b) 44(2)% condensate fraction at 56(3) nK; (c) a BEC of condensate fraction 73(4)% and $1.5(2) \times 10^4$ atoms forms at 28(2) nK with density 10^{14} cm^{-3} .

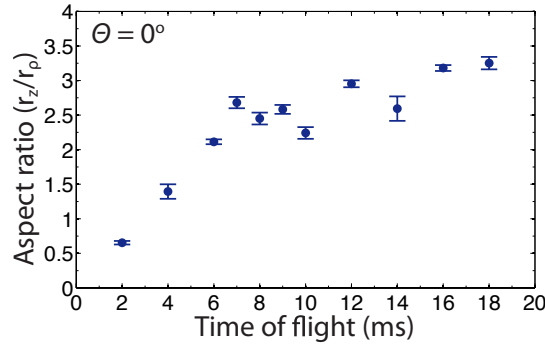


Figure 9.3: Anisotropic expansion profile versus time after trap release. r_z and r_ρ are the dimensions of the parabolic profile fit to the BEC.

beams are linearly ramped in 0.8 s to their final powers. For the data presented here, final ODT powers yield the trap frequencies $[f_x, f_y, f_z] = [205, 195, 760]$ Hz, which provide a ratio $f_z/f_\rho = 3.8$.⁴ The BEC is held for 0.2 s, the first 0.1 s of which the B_z field may be rotated into the ρ -plane. Time-of-flight measurements are made by rapidly extinguishing the cODT and, after 1 ms, rotating the field into the imaging axis [see Fig. 9.1]. Absorption images are obtained using resonant 421 nm light and an exposure of 60 μ s, much less than the decay time to metastable states [108, 27].

The emergence of the Dy BEC below $T = 100$ nK is shown in Fig. 9.2. A dense parabolic peak emerges from a Gaussian background signaling the formation of a condensate with high purity below $T = 30$ nK. The DDI preserves a parabolic condensate profile [175], but the criterion for the Thomas-Fermi (TF) limit is modified for a dipolar BEC. Approximate expressions for the dipolar TF limit in the spherical and highly elongated prolate and oblate BEC regimes have been obtained [176]. Although our moderately oblate BEC fails to fall into these regimes, we may use the oblate BEC analysis in Ref. [176] to estimate that the Dy BEC satisfies the dipolar

⁴Trap frequencies, measured via parametric heating, are uncertain to 5%.

TF limit for “effective” a_s ’s⁵ in the vicinity of the recently estimated van der Waals length $\bar{a} = 76a_0$ [93].

We calculate that an initial phase space density for BEC is reached at 120(20) nK for a trap with our mean frequency 310 Hz and atom number 7×10^4 . Applying corrections [4] for finite size effects and mean field energy (using $a_s = 100a_0$), we estimate a $T_c = 100(20)$ nK, which is consistent with our measurement of 99(5) nK. The nearly pure BEC in panels (e) and (f) contains 1.5×10^4 atoms at a density of 10^{14} cm^{-3} . Further evidence of ^{164}Dy ’s Bose-condensation is provided in Fig. 9.3, which shows the characteristic aspect ratio inversion of an interacting BEC released from an anisotropic trap. These data are obtained with a small, $B_z = 2.3$ G field parallel to the oblate trap’s axis along z . BECs also form with 2.3 G fields rotated to an angle either $\theta = 45^\circ$ or 60° with respect to the z -axis. However, no BEC forms in a 90° field ($B_z = 0$, $B_\rho = 2.3$ G), and a thermal cloud is obtained regardless of evaporation parameters.

9.3 Strongly dipolar BEC

Strongly dipolar BECs—defined as $\epsilon_{dd} > 1$ —have been shown [177, 178, 179, 180, 181, 15] to be unstable for harmonic traps with $\gamma = f_{\parallel}/f_{\perp} < 1$ but metastable for traps with $\gamma = f_{\parallel}/f_{\perp} > 1$, where $f_{\parallel}$ is the trap frequency parallel to the polarizing field, and $f_{\perp}$ is the geometric mean of the trap frequencies perpendicular to the field. The attractive part of the DDI acts to elongate the cloud along the field direction, which if pointed along the trap’s major axis leads to an instability much akin to that

⁵ ^{164}Dy has 153 Born-Oppenheimer potentials [93] which can contribute to the effective a_s .

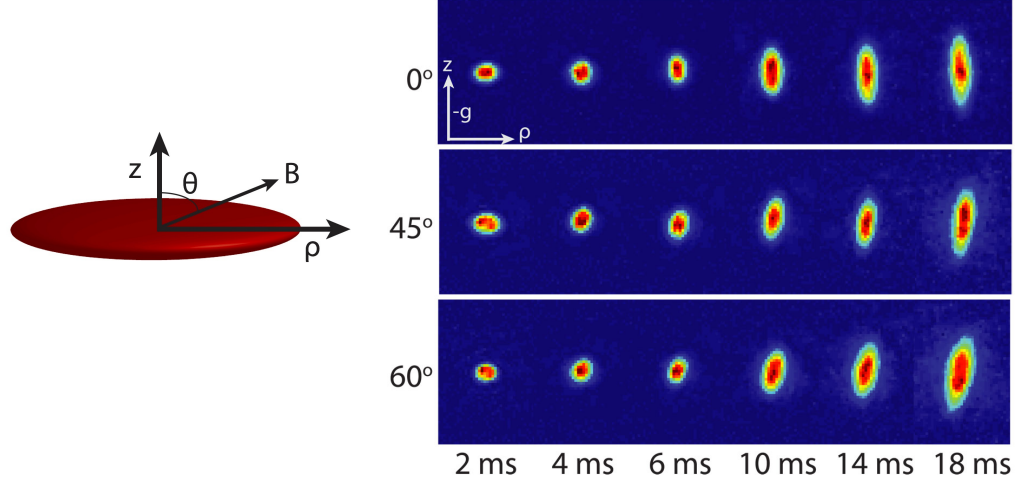


Figure 9.4: Rotated anisotropic expansion profile versus time after trap release. Left panel: Schematic of the oblate trap and magnetic field orientation. Right panel: Images of the expanding condensate after trap release with different orientations of the magnetic field. The condensate rotates by $7(1)^\circ$ [$9.4(6)^\circ$] with respect to the $\theta = 0^\circ$ expansion orientation for $\theta = 45^\circ$ [$\theta = 60^\circ$]. No BEC forms for $\theta = 90^\circ$.

of non-dipolar BECs with large negative a_s [15]. In contrast, aligning the field along the minor axis acts to suppress this elongation, minimizing the attractive contribution from the DDI to the mean field energy and stabilizing the gas. Weakly dipolar BECs—wherein $\epsilon_{dd} < 1$ —are stable regardless of the value of γ , because the repulsive contact interaction dominates the DDI [15].

While we observe a BEC in our oblate trap for $\theta = 0^\circ$ ($\gamma = 3.8$), the failure to observe a BEC in the oblate trap with $\theta = 90^\circ$ ($\gamma = 0.51$) is strongly suggestive of a BEC in the strongly dipolar regime.⁶ Moreover, the tilt of the expanding condensate toward the polarizing field (Fig. 9.4) is indicative of a strongly dipolar condensate

⁶For the field orientation $\theta = 0^\circ$ (90°), γ is defined using $f_{\parallel} = f_z$ ($f_{\parallel} = f_\rho$) and $f_{\perp} = f_\rho$ ($f_{\perp} = \sqrt{f_\rho f_z}$). Most references analyze BEC stability for traps cylindrically symmetric about the polarization axis ($\theta = 0^\circ$). However, an oblate trap with our aspect ratio and $\theta = 90^\circ$ was treated in Ref. [182] with the result that $\epsilon_{dd} = 1$ remains the instability boundary.

and was not observed in the weakly dipolar Cr system [183]. Gross-Pitaevskii equation simulations of the BEC stability boundary and tilt angle versus θ , along with simulations of the TOF aspect ratio evolution, should provide a first measurement of ^{164}Dy 's effective a_s [184].

Chapter 10

Degenerate Dipolar Fermi Gases of Dysprosium

10.1 Introduction

The interplay between crystallinity and superfluidity is of great fundamental and technological interest in condensed matter settings. In particular, electronic quantum liquid crystallinity arises in the non-Fermi liquid, pseudogap regime neighboring a cuprate’s unconventional superconducting phase [2]. While the techniques of ultracold atomic physics and quantum optics have enabled explorations of the strongly correlated, many-body physics inherent in, e.g., the Hubbard model [129], lacking has been the ability to create a quantum degenerate Fermi gas with interparticle interactions—such as the strong dipole-dipole interaction [15]—capable of inducing analogs to electronic quantum liquid crystals. We report the first quantum degenerate dipolar Fermi gas, the realization of which opens a new frontier for exploring strongly

correlated physics and, in particular, the quantum melting of smectics in the pristine environment provided by the ultracold atomic physics setting [1].

A quantum degenerate Fermi gas of the most magnetic atom ^{161}Dy is produced by laser cooling to $10\ \mu\text{K}$ before sympathetically cooling with ultracold, bosonic ^{162}Dy . The temperature of the spin-polarized ^{161}Dy is a factor $T/T_F = 0.2$ below the Fermi temperature $T_F = 300\ \text{nK}$. The co-trapped ^{162}Dy concomitantly cools to approximately T_c for Bose-Einstein condensation, thus realizing a novel, nearly quantum degenerate dipolar Bose-Fermi gas mixture.

Quantum soft phases are states of quantum matter intermediate between canonical states of order and disorder, and may be considered the counterparts of liquid crystalline and glassy states in classical (soft) condensed matter physics. Such phases tend to arise under competition between short and long-range interactions and often result in the non-Fermi liquid, strongly correlated behavior manifest in some of the most interesting electronic materials of late: high- T_c cuprate superconductors, strontium ruthenates, 2D electron gases, and iron-based superconductors [3]. Recent theory suggests the long-range, anisotropic dipole-dipole interaction (DDI) among atoms in a degenerate Fermi gas, confined in an harmonic trap or optical lattice, may also induce transitions to states beyond the now-familiar insulating, metallic, and superfluid. Namely, phases that break rotational, translational, or point group symmetries may emerge in a manner akin to those found in classical liquid crystals, e.g., the nematic and smectic [2].

Degenerate gases of highly magnetic fermionic atoms, such as ^{161}Dy , may shed light on QLC physics without unwanted solid state material complexity, disorder, and

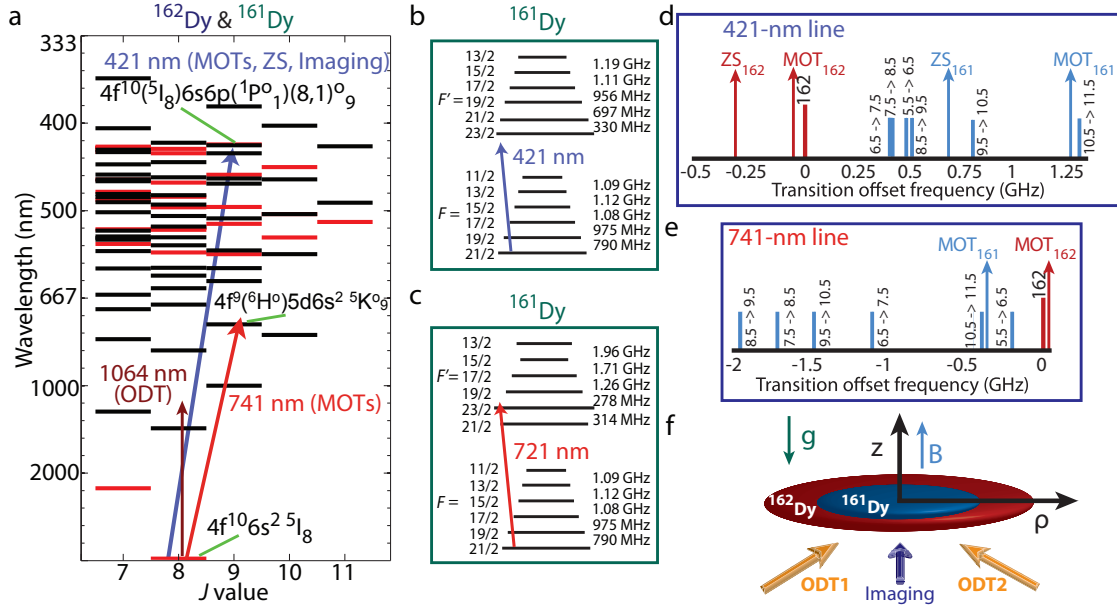


Figure 10.1: Dy transitions and laser cooling and trapping scheme. **a**, Electronic energy level structure for bosonic ^{162}Dy (nuclear spin $I = 0$) and fermionic ^{161}Dy ($I = 5/2$), including laser cooling and trapping transitions. **b**, Additional ground and excited-state hyperfine structure exists for ^{161}Dy ($F = I + J$, where $J = 8$ is the total electronic angular momentum and primes denote the excited states). Shown is the 32-MHz-wide transition at 421 nm used for the transverse cooling, Zeeman slower, capture MOT, and imaging beams. **c**, Blue-detuned, narrow-line (1.8 kHz-wide) MOT cooling transition at 741 nm. **d**, Spectra of 421 nm ^{162}Dy and ^{161}Dy (hyperfine) transitions including relative detunings of each MOT and Zeeman slower (ZS) laser. **e**, Transition and detuning spectra for MOT on 741 nm line. **f**, Sketch of dual species crossed optical dipole trap (ODT) aspect ratio along with magnetic field B and gravity g orientation. The imaging (421 nm) beam and the orthogonal ODT (1064 nm) beams are in the $\hat{\rho}$ -plane.

dynamical lattice distortions. Uniaxial (meta-nematic) [185] and biaxial nematic [7] distortions of the Fermi surface of a harmonically trapped gas in the presence of a polarizing field may be observable as well as meta-nematic and smectic phases in 2D anisotropic optical lattices [148, 149, 150]. An exciting prospect lies in the possibility of achieving spontaneous magnetization in dipolar systems coupled with nematic order [8, 186]. Additionally, DDI-induced pairing of fermions may lead to supersolidity [147] and bond order solids [187].

However, obtaining a quantum degenerate dipolar Fermi gas has been a difficult, unrealized experimental challenge. The highly magnetic fermionic atoms ^{53}Cr (6 Bohr magnetons μ_B) and ^{167}Er ($7 \mu_B$) have yet to be cooled below $10 \mu\text{K}$ [188, 87]. The fermionic polar molecule $^{40}\text{K}^{87}\text{Rb}$ (0.57 Debye) has been cooled to near degeneracy ($T/T_F = 1.4$) [189] and loaded into a long-lived lattice while partially polarized (0.2 D) [190], but complexities arising from ultracold chemistry have hampered additional evaporative cooling [189]. In contrast, magnetic fermionic atoms do not undergo chemical reactions and are immune to inelastic dipolar collisions when spin polarized in high magnetic fields [5, 191].

The strong, r^{-3} character of the DDI arises in ground state polar molecules through a polarizing electric field that mixes opposite parity states. This electric field breaks rotational symmetry; consequently, observing the full range of true (non-meta) quantum nematic and fluctuating smectic phases, and their often unusual topological defects, is not possible in systems of fermionic polar molecules, especially in three dimensions. By contrast, highly magnetic atoms exhibit the DDI interaction even in the absence of a polarizing field. Moreover, the magnetic DDI can be tuned from positive

to negative [192], which may be important for simulating dense nuclear matter.

Dysprosium's isotopes ^{161}Dy and ^{163}Dy are the most magnetic fermionic atoms. With a dipole moment of $\mu = 10 \mu_B$, ^{161}Dy provides a DDI length $l_{\text{DDI}} = \mu_0 \mu^2 m / 4\pi \hbar^2$ that is factors of [400, 8, 2] larger than that of [^{40}K , ^{53}Cr , ^{167}Er]. With respect to fully saturated $^{40}\text{K}^{87}\text{Rb}$ (0.57 D), ^{161}Dy is $30\times$ less dipolar for equal densities, but within a factor of 2 if confined in a lattice of less than half the periodicity. Lattices of wavelength 400–500 nm may be possible with Dy, whereas for molecules, photon scattering from rovibronic states at these wavelengths may reduce gas lifetimes. Indeed, ^{161}Dy confined in a 450 nm lattice would provide a DDI strength more than $3\times$ larger than $^{40}\text{K}^{87}\text{Rb}$ confined in a 1064 nm lattice, if the electric dipole moment is unsaturated (0.2 D) to maintain collisional stability [190]. We estimate that the DDI strength of Dy confined in short-wavelength lattices would be sufficient to observe some of the exotic many-body physics recently proposed [193, 147, 187, 194].

Until recently, the laser cooling of Dy posed an insurmountable challenge due to its complex internal structure and the limited practicability of building repumping lasers: ~ 140 metastable states exist between the ground state and the broadest laser cooling transition at 421 nm (see Fig. 10.1a). Moreover, an open f -shell submerged underneath closed s -shells, combined with a large magnetic moment and electrostatic anisotropy from the $L = 6$ orbital angular momentum, pose challenges to molecular coupled-channel calculations [93] which could otherwise guide early experiments. Even more daunting is the additional hyperfine structure of the fermions, shown in Fig. 10.1b–c, which splits each level into six.

Despite this vast energy level state-space, a repumperless magneto-optical (MOT)

technique was able to individually laser cool and trap of all five high-abundance isotopes (three bosons, two fermions) to 1 mK using a single laser [108] (see Methods for details). Moreover, the complex homonuclear molecular potentials—involving 153 Born-Oppenheimer surfaces—and the associated multitude of scattering lengths did not inhibit the efficient Bose-condensation of spin-polarized ^{164}Dy through forced evaporative cooling [5].

However, quantum degeneracy of identical Fermi gases is often more difficult to achieve than Bose-condensation because s -wave collisions are forbidden due to the requirement that the total wave function for two identical fermions be anti-symmetric with respect to particle exchange. Rethermalization from elastic collisions cease below the threshold for p -wave collisions (at typically 10–100 μK), and efficient evaporative cooling can no longer be maintained. Co-trapping mixtures of particles—either as different spin states of the same atom or as mixtures of isotopes or elements—reintroduces s -wave collisions, providing a finite elastic cross-section for scattering even down to low temperatures. The mixture needs to be stable against inelastic collisions which could add heat or induce trap loss. Evaporating ^{161}Dy in a mixed state of two spins, as proved efficient for ^{40}K , [195] would lead to large dipolar relaxation-induced heating even in the presence of small, mG-level fields because the inelastic, single spin-flip cross-section $\sigma_1 = \sigma\zeta(k_f/k_i)$ scales strongly with dipole moment [92]:

$$\sigma = \frac{8\pi}{15} F_1 F_2^2 \left(\frac{\mu_0 g_1 g_2 \mu_B^2 m}{4\pi\hbar^2} \right)^2 \frac{k_f}{k_i},$$

where F_1 is the spin of the atom whose spin flips ($F_1 = F_2$ for identical particles), g_i are g -factors for atom i , m is mass, and k_i and k_f are the initial and final momenta. For ^{161}Dy (^{162}Dy), $F = 21/2$ ($F = J = 8$) and g_F (g_J) = 0.95 (1.24). The function

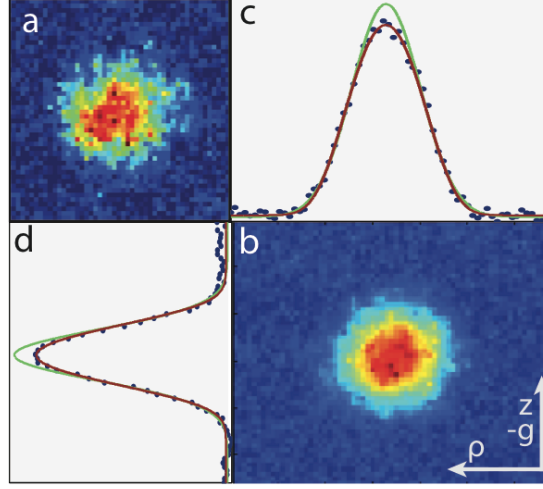


Figure 10.2: **Images of the Dy degenerate Fermi gas.** **a**, Single shot time-of-flight absorption image at $t = 6$ ms. **b**, Average of six images. Density integrations versus $\hat{\rho}$ (**c**) and $\hat{z}$ (**d**). The green curve is a gaussian fit to the data's wings (radius $\sigma = 20 \mu\text{m}$), while the red curve is a fit to a Thomas-Fermi distribution. Data are consistent with a Thomas-Fermi distribution of $T/T_F = 0.21(5)$. The Fermi velocity and temperature are $5.6(2)$ mm/s and $306(20)$ nK, respectively, and the gas temperature is $64(16)$ nK. The degenerate Fermi gas contains $6.0(6) \times 10^3$ atoms at peak density $4(1) \times 10^{13} \text{ cm}^{-3}$.

$\zeta(k_f/k_i) = [1 + \epsilon h(k_f/k_i)]$, where $\epsilon = \pm 1, 0$ and $h(x)$ is defined in Methods, accounts for the contributions of even, odd, or all partial waves to the scattering process.

10.2 Production of dipolar fermi gas of Dy

We choose, therefore, to seek a degenerate dipolar Fermi gas with Dy by sympathetically cooling ^{161}Dy with the boson ^{162}Dy while both are spin-polarized in their strong-magnetic-field seeking ground states: $|F, m_F\rangle = |21/2, -21/2\rangle$ for ^{161}Dy and $|J, m_J\rangle = |8, -8\rangle$ for ^{162}Dy . See Fig. 10.1a–c for energy level schemes. Preparation of this ultracold Bose-Fermi mixture—the first such mixture for strongly dipolar species—builds on our single-species technique [5] for Bose-condensing ^{164}Dy and re-

lies on the laser cooling and trapping of two isotopes before loading both into an optical dipole trap (ODT) for forced evaporative cooling. We sketch here the experimental procedure; further details are provided in the Methods.

Isotopes ^{161}Dy and ^{162}Dy are collected sequentially in a repumperless MOT operating on the 421 nm transition [108] (Fig. 10.1d), with final MOT populations of $N = 2 \times 10^7$ and 4×10^7 , respectively. Next, simultaneous narrow-line, blue-detuned MOTs [113, 5] cool both isotopes to 10 μK via the 741 nm transition (Fig. 10.1e) for 5 s to allow any remaining metastable atoms to decay to the ground state. The blue-detuned MOTs also serve to spin polarize [113, 5] both isotopes to their maximally high-field-seeking (metastable) states $m_F = +F$ ($m_J = +J$) for ^{161}Dy (^{162}Dy).

The blue-detuned MOTs of the two isotopes can be spatially separated due to the dependence of the MOTs' positions on laser detuning [113, 5]. This allows the isotopes to be sequentially loaded into the 1064 nm ODT1 in Fig. 10.1f, which is aligned above the ^{161}Dy MOT but below the ^{162}Dy MOT. First ^{162}Dy and then ^{161}Dy is loaded into ODT1 by shifting the quadrupole center with a vertical bias field. All 741 nm light is extinguished before the spin of both isotopes are rotated via radiofrequency (RF) adiabatic rapid passage (ARP) into their absolute ground states $m_F = -F$ ($m_J = -J$) for ^{161}Dy (^{162}Dy). The ODT1 populations of ^{161}Dy and ^{162}Dy are both initially 1×10^6 before plain evaporation cools the gases to 1–2 μK within 1 s. A 0.9 G field is applied close to the trap axis of symmetry $\hat{z}$ throughout plain and forced evaporation. This provides a $\Delta m = 1$ Zeeman shift equivalent to 50 (70) μK for ^{161}Dy (^{162}Dy). Because this is much larger than the temperatures of the gases, the field serves to maintain spin polarization while stabilizing the strongly dipolar ^{162}Dy

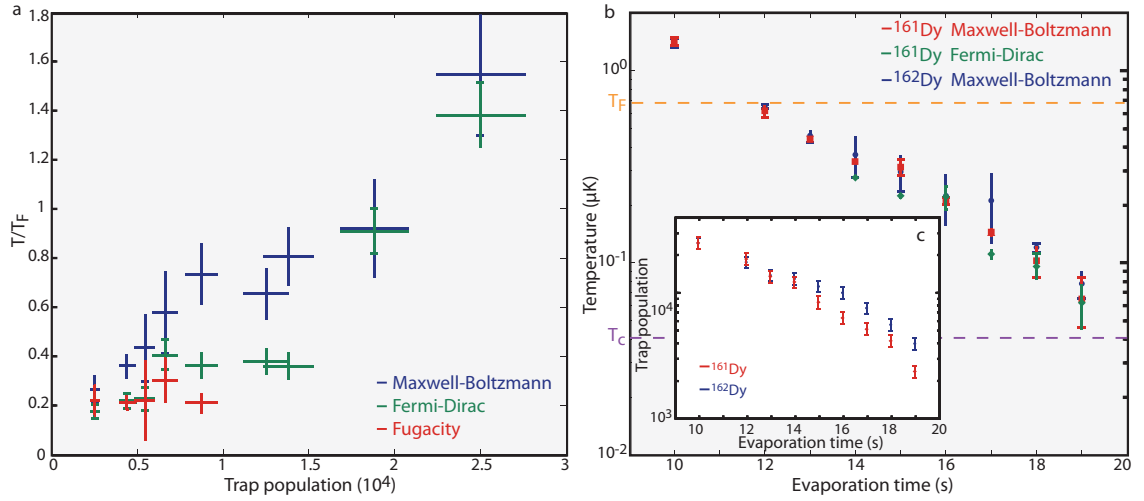


Figure 10.3: **Temperature and population of ^{161}Dy and ^{162}Dy mixture.** **a**, Three measures (see text) of ^{161}Dy $\hat{\rho}$ temperature-to-Fermi temperature T_F as trap population decreases due to forced evaporation of the spin-polarized Bose-Fermi mixture. **b**, The dipolar Bose-Fermi mixture remains in thermal contact throughout the evaporation sequence as measured by fits of TOF densities to gaussian or TF distributions. The orange dashed line demarcates the boundary below which the temperature of ^{161}Dy is lower than T_F . Likewise, the purple dashed line demarcates the temperature below which T_c for ^{162}Dy Bose degeneracy would be reached given the trap frequencies and population at 19 s. **c**, Trap populations of the spin-polarized Bose-Fermi mixture versus evaporation time.

Bose gas against collapse as its phase-space density increases [5].

Magnetic Stern-Gerlach measurements and observations of fluorescence versus polarization are consistent with an RF ARP sequence that achieves a high degree of spin purity for each isotope. Remnant population in metastable Zeeman substates quickly decays to the absolute ground state via dipolar relaxation regardless of collision partner at a rate of $\Gamma \propto \sigma_1 n \bar{v} = 1\text{--}10 \text{ s}^{-1}$, where n is the atomic density and $\bar{v}$ is the relative velocity during the plain evaporation stage. (Since $g_F F = g_J J$, collisions between Bose-Bose, Bose-Fermi, and Fermi-Fermi pairs result in Γ 's of similar magnitude as long as $k_i \approx k_f$ since $h(x \rightarrow 1) \rightarrow 0$. This condition is fulfilled during plain evaporation due to a low ratio of Zeeman-to-kinetic energy. For example, inelastic dipolar $^{161}\text{Dy}\text{--}^{161}\text{Dy}$ collisions ($\epsilon = -1$) proceed at rate $\Gamma = 1\text{--}5 \text{ s}^{-1}$ even in the absence of ^{162}Dy .) Thus, a (two-body) collisionally stable mixture of identical bosons and identical fermions is prepared within the 1 s between spin rotation and forced evaporation.

Subsequently crossing ODT1 with ODT2 forms an oblate trap with frequencies $[f_x, f_y, f_z] = [500, 580, 1800] \text{ Hz}$. Ramping down the optical power lowers the trap depth and evaporates spin-polarized ^{161}Dy to quantum degeneracy; with a 19 s evaporation, the final trap has frequencies $[180, 200, 720] \text{ Hz}$ and $\bar{\omega}/2\pi$ is defined as their geometric mean. Figure 10.2 shows the density profile of ultracold ^{161}Dy , which is more consistent with a Thomas-Fermi (TF) distribution arising from Fermi-Dirac statistics than a gaussian arising from a classical, Maxwell-Boltzmann (MB) distribution.

Figure 10.3 shows a collection of such fits as a function of trap population and

evaporation time. The T/T_F data are extracted from density profiles using three methods: 1) The high momentum wings are fit to a MB distribution with $k_B T_F = \hbar\bar{\omega}(6N)^{1/3}$ extracted from measured trap parameters and population (Fig. 10.3c); 2) similar to (1), but using a TF distribution; 3) fitting the fugacity to directly extract T/T_F . The last method is known to be inaccurate at higher temperatures, while the gaussian fit tends to overestimate the temperature below $T/T_F = 1$. [195] The evaporation does not yet seem to reach a plateau in cooling efficiency at 19 s; poor imaging signal-to-noise hampers measurements at longer evaporation times.

Data in Fig. 10.3b show that thermal equilibrium between the bosons and fermions is maintained throughout the evaporation, and Bose-condensation of ^{162}Dy within the mixture is nearly reached for an evaporation of 19 s. We estimate the corresponding critical temperature $T_c \approx 40$ nK of co-trapped ^{162}Dy by scaling the measured $T_c \approx 120$ nK of singly trapped ^{162}Dy with the cube root of their relative trap populations. (^{162}Dy has been Bose-condensed in the absence of ^{161}Dy ; manuscript in preparation.) The nearly doubly degenerate dipolar Bose-Fermi mixture may lead to interesting dipolar and many-body physics once cooling efficiency improves.

While interacting BECs invert their anisotropic aspect ratio upon time-of-flight expansion, anisotropic degenerate Fermi gases tend to a spherical shape. As the DDI strength increases, the degenerate Fermi gas will expand into a prolate ellipsoid oriented along the magnetization direction regardless of trap aspect ratio. Furthermore, the gas may become unstable when the quantity $\epsilon_{dd} = \mu_0\mu^2(m^3\bar{\omega}/16\pi^2\hbar^5)^{1/2} > 1$. [185] At the lowest attained T/T_F , $\epsilon_{dd} = 0.2$ for ^{161}Dy , and the ratio is l_{DDI}/l_F is 0.05. This DDI strength should lead to Fermi surface distortions (as yet unmeasured) at the

percent level [185]. Both ratios could be enhanced $\sim 3\times$ by increasing trap frequency using a more spherical confinement—while maintaining stability of the dipolar Bose gas—and by increasing trap population. Additionally, the efficiency of evaporation—from gaussian fits to the data, $\gamma = 3 \frac{d(\ln T/T_F)}{d(\ln N)} \approx 2.3$ —may be improved by optimally tuning the magnetic bias field near one of the presumably large number of Feshbach resonances.

10.3 Onset of Fermi degeneracy of single-component Dy gas via dipolar scattering

Surprisingly, we achieve the forced evaporative cooling of spin-polarized ^{161}Dy *without* ^{162}Dy to $T/T_F = 0.7$ at $T_F = 500$ nK. As mentioned above, achieving quantum degeneracy with spin-polarized identical fermions alone is usually not possible due to suppression of elastic scattering below the p -wave threshold $50 \mu\text{K}$. [93] That such a low temperature ratio is achieved may be a novel consequence of the highly dipolar nature of this gas: namely, that a significant elastic cross-section persists to low temperatures due to the as yet unobserved phenomenon of universal dipolar scattering [26]. The associated scattering rate is expected to scale as $m^{3/2}\mu^4$ regardless of the details of the short-range molecular potential. The predicted fermionic Dy universal dipolar cross-section, $7.2 \times 10^{-12} \text{ cm}^2$, is nearly equal to ^{87}Rb 's s -wave cross-section and could provide sufficient rethermalization for the evaporative cooling we observe. While future measurements will quantify spin purity, Fermi statistics does not inhibit identical fermions from spin purifying via dipolar relaxation once

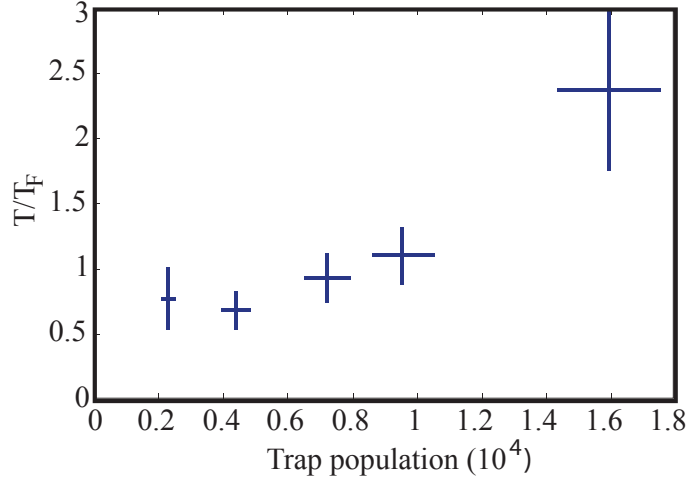


Figure 10.4: Evaporative cooling of single component dipolar fermions.

the RF ARP sequence populates the majority of the atoms in the absolute ground state. We will present these data along with supporting measurements of collisional cross-sections and scattering lengths in a future manuscript.

The efficacy of experimental proposals [1] for studying the quantum melting of QLCs—important for better understanding the relationship of incipient electronic stripe order to unconventional superconductivity—may now be investigated using degenerate ^{161}Dy in optical lattices wherein l_{DDI}/l_F can be greatly enhanced [193]. Looking beyond QLC physics, the large spin $F = 21/2$ of the novel degenerate dipolar Fermi gas presented here opens avenues to explore exotic spinor physics as well as physics associated with strong spin-orbit coupling.

10.4 Methods

Repumperless MOT. The 421 nm Zeeman slower laser is detuned -650 MHz from resonance, and a 421 nm MOT collects 2×10^7 ^{161}Dy atoms in 4 s with the aid of a pre-slower transverse cooling stage. The MOT operates without repumpers despite many decay chan-

nels because the highly magnetic, metastable Dy remains trapped in the magnetic quadrupole trap for a sufficiently long time to decay to the ground state [108]. All beams derived from the 421 nm laser are then shifted down in frequency by 1.18 GHz and a ^{162}Dy MOT collects 4×10^7 atoms in 100 ms. This scheme balances the need for preservation of ^{161}Dy population against collisional decay [108] with the trapping of sufficient ^{162}Dy for subsequent sympathetic cooling. Reversing this procedure results in fewer atoms due to heating of ^{162}Dy in the magnetic trap by light scattered from the ^{161}Dy Zeeman slower beam.

Narrow-line MOT. The narrow-line MOT lasers are blue-detuned 0.6 MHz from the 2-kHz wide 741 nm transition. The MOTs form below the quadrupole center such that the atomic transition is Zeeman-shifted to the blue of the laser. The MOT positions are determined by the balance of optical, magnetic, and gravitational forces [113, 5], and because the laser detunings for ^{161}Dy and ^{162}Dy can be adjusted independently, the two clouds can be easily separated.

ODT, RF ARP and crossed ODT. ODT1 has a waist of 30 (60) μm and ODT2 has a waist of 20 (60) μm in $\hat{z}$ ($\hat{\rho}$). ODT1 is ramped on in 100 ms after 5 s of narrow-line cooling. After loading ODT1—120 (800) ms for ^{162}Dy (^{161}Dy)—a 4.3 G field is applied while a 20-ms RF sequence flips the spin of both isotopes to the absolute ground state. Dipolar relaxation further spin purifies the gas, and within the cross section σ_1 , $h(x) = -\frac{1}{2} - \frac{3}{8} \frac{(1-x^2)^2}{x(1+x^2)} \log \left(\frac{(1-x)^2}{(1+x)^2} \right)$ [see Ref. [92]]. After spin purification, ODT2 is turned on, forming a crossed ODT with depth 300 μK . ODT1 and ODT2 have initial powers of 18 W and 12 W. The background limited lifetime exceeds 180 s.

Forced evaporation scheme. The two beams of the crossed ODT are ramped down according to the functional form $P(t) = P_0/(1 + t/\tau)^\beta$ using experimentally determined

parameters $\tau = 6$ s and $\beta = 1.5$ for both beams. Forced evaporation with ODT2 begins 5 s after ODT1. The beam powers are held at the final values for 400 ms after evaporation to allow for equilibration.

Temperature fitting. The TOF images are fit to both gaussian and TF profiles, the latter of the form [195]

$$\left[\frac{A}{Li_2(-\zeta)} \right] Li_2 \left(-\zeta e^{\frac{-(y-y_0)^2}{2\sigma_y^2}} e^{\frac{-(x-x_0)^2}{2\sigma_x^2}} \right) + c, \quad (10.1)$$

with fitting parameters A , ζ , y_0 , x_0 , σ_y , σ_x , and c . The fitting is scaled by the constant $Li_2(-\zeta)$ such that parameter A corresponds exactly to the image peak OD. Because the high-velocity component of the cloud is less sensitive to signatures of quantum degeneracy, only the wings of the expanded cloud are used for the gaussian TOF fits. From the TF fits, the temperature is determined from size of the cloud, $\sigma_{TF,i}^2 = (k_b T / m \omega_i^2) [1 + (\omega_i t)^2]$, where ω_i is the trap frequency in $\hat{z}$ or $\hat{\rho}$ and t is the expansion time. The cloud fugacity ζ is also determined by the TF fits and provides a direct measure of $T/T_F = [-6Li_3(-\zeta)]^{-1/3}$, where Li_3 is the third-order polylogarithm function.

Appendix A

^{161}Dy Ground State Hyperfine Structure in Magnetic Fields

^{161}Dy ground state has 102 Zeeman sublevels grouped in 6 hyperfine manifolds ($F=21/2$ to $11/2$, $J=8$, $I=5/2$) in the low magnetic limit. In a finite magnetic field, the full Hamiltonian read:

$$\begin{aligned} H/h = & A\vec{I} \cdot \vec{J} + B \frac{\frac{3}{2}\vec{I} \cdot \vec{J}(2\vec{I} \cdot \vec{J} + 1 - I(I+1)J(J+1))}{2I(2I-1)J(2J-1)} \\ & + g_J\mu_B m_J B/h + g_I\mu_N m_I B/h, \end{aligned} \quad (\text{A.1})$$

where $A = -116.231$ MHz and $B = 1091.577$ MHz are hyperfine coefficients (see Chapter 6), g_J, g_I are electronic and nuclear Landé g factors, μ_B, μ_N are Bohr and nuclear magnetons [64]. Eigenvalues of each Zeeman sublevel are obtained through diagonalization of this Hamiltonian at magnetic field strength B . Numerical calculations are presented in Fig. A.3 and Fig. A.3 for lowest hyperfine manifold and full manifolds respectively. Unlike alkali atoms ($J=1/2$) for which the Breit-Rabi formula provides the analytical eigenvalues of zeeman states in the

intermediate field regime [196], Dy has sub-blocks with sizes of 6×6 in the full Hamiltonian in general and no analytical solutions are available. However, we could fit numerical values to polynomials once the Hamiltonian has been diagonalized. Empirically we found a polynomial fit up to cubic term offers approximate analytical formulae with residual errors less than 0.4 MHz within 100 Gauss (Fig. A.3).

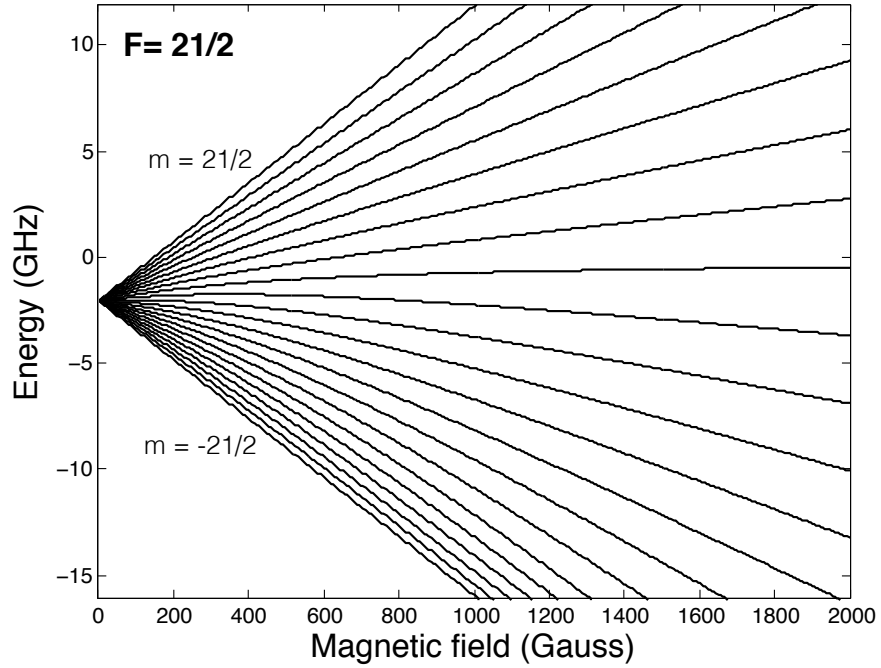
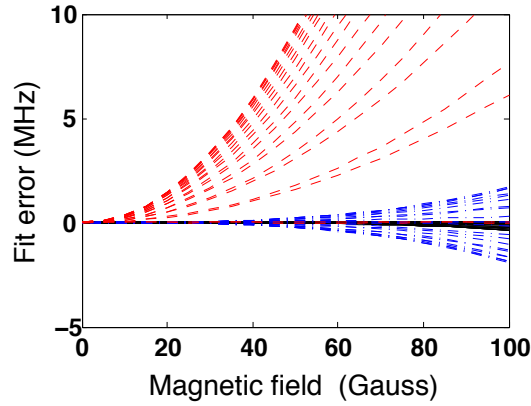


Figure A.1: ^{161}Dy $4f^{10}6s^2\ ^5\text{I}_8$ (ground state) $F = 21/2$ hyperfine manifold in external magnetic field. It is the lowest hyperfine manifold with 22 Zeeman sub-levels. Note that the stretch states $m_F = -21/2$ (lowest) and $m_F = 21/2$ (highest) have no quadratic Zeeman shifts since there is no mixing with other F manifolds in ground state.

Table A.1: Polynomial fit to Zeeman shifts of ^{161}Dy $F=21/2$ ground state up to cubic term.

m_F	C3 ($\times 10^{-6}$ MHz G $^{-3}$)	C2 ($\times 10^{-3}$ MHz G $^{-2}$)	C1 (MHz G $^{-1}$)
-21/2	0	0	-13.902
-19/2	-0.799	-0.693	-12.578
-17/2	-1.357	-1.317	-11.254
-15/2	-1.701	-1.871	-9.930
-13/2	-1.855	-2.356	-8.606
-11/2	-1.845	-2.773	-7.282
-9/2	-1.697	-3.119	-5.958
-7/2	-1.435	-3.396	-4.634
-5/2	-1.086	-3.604	-3.210
-3/2	-0.674	-3.742	-1.986
-1/2	-0.224	-3.812	-0.662
1/2	0.238	-3.812	-0.662
3/2	0.687	-3.742	-1.986
5/2	1.098	-3.604	-3.210
7/2	1.446	-3.396	-4.634
9/2	1.705	-3.119	-5.958
11/2	1.851	-2.773	-7.282
13/2	1.858	-2.356	-8.606
15/2	1.701	-1.871	-9.930
17/2	1.356	-1.317	-11.254
19/2	0.797	-0.693	-12.578
21/2	0	0	13.902

Figure A.2: Fit errors for Zeeman shifts in each sublevels of $F = 21/2$. Dash line (red) is Linear fit error; dash-dot line (blue) is quadratic fit error; solid line (black) is cubic fit error.

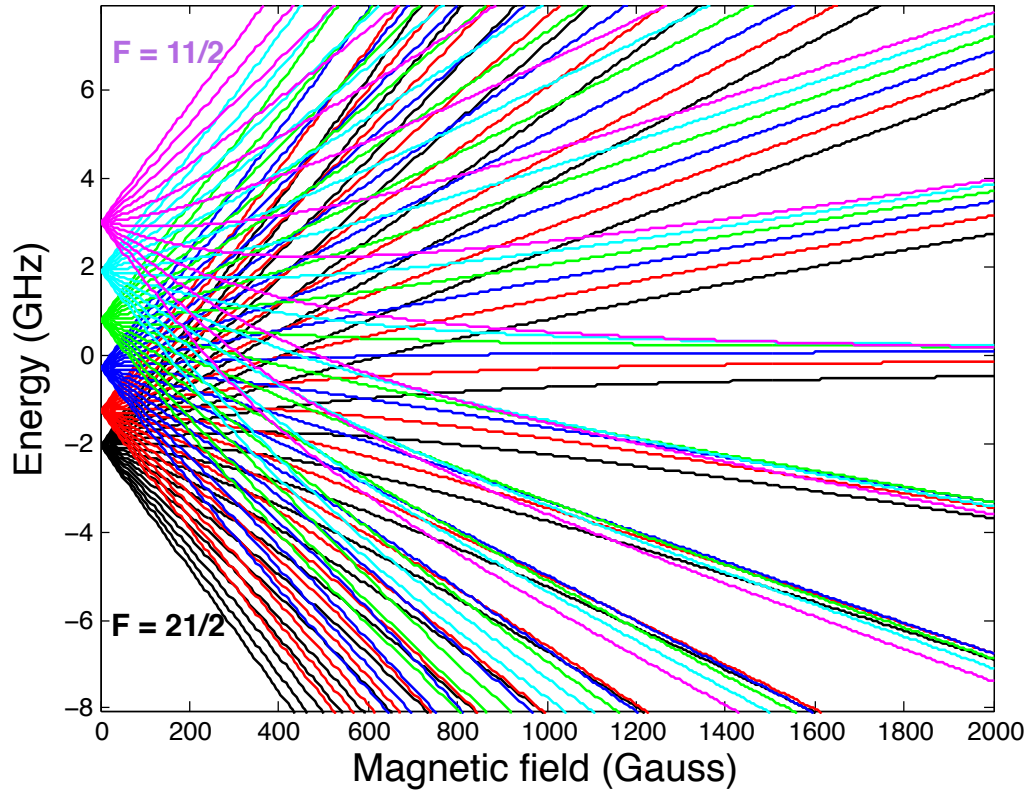


Figure A.3: ^{161}Dy $4f^{10}6s^2\ ^5\text{I}_8$ (ground state) hyperfine structure in external magnetic field. 102 Zeeman sub-levels are grouped in six hyperfine manifolds: $F = 21/2$ (black), $19/2$ (red), $17/2$ (blue), $15/2$ (green), $13/2$ (cyan), $11/2$ (magenta) in an energy-increasing order at zero field limit. In stronger fields (Paschen-Back regime), $m_J = -8, -7, \dots, 8$ increasingly become better quantum numbers.

Appendix B

Optical Absorption of Atoms with $J \rightarrow J + 1$ Transition

The 421 nm imaging transition of Dy is of the $J \rightarrow J + 1$ type transition. For this type of transition, the stretch state $|J, m = J\rangle$ is driven to the excited state $|J' = J + 1, m = J + 1\rangle$ under σ^+ polarized excitation with full oscillator strength [105]. It means it can be treated effectively as two-level system. Mathematically, the absorption of σ^+ light for $|J, m = J\rangle$ is:

$$\begin{aligned}\sigma &\propto W_{e \leftarrow g} \\ &\propto |\langle J' = 9, m' = 9 | d_{q=1} | J = 8, m = 8 \rangle|^2 \\ &= \frac{|\langle J' = 9 || \mathbf{d} || J = 8 \rangle|^2}{2J' + 1} |\langle J, m = J, 1, q = 1 | J', m' = J' \rangle|^2 \\ &= \frac{|\langle J' = 9 || \mathbf{d} || J = 8 \rangle|^2}{2J' + 1}\end{aligned}\tag{B.1}$$

where $W_{e \leftarrow g}$ is the transition rate, $d_{q=1}$ the dipole operator in spherical basis with $q = 1$, $\langle \dots || \mathbf{d} || \dots \rangle$ the reduced matrix, and $\langle J, m, 1, q | J', m' \rangle$ the Clebsch-Gordan coefficient (CG). The Wigner-Eckart theorem is used in the derivation and the normalization convention follows [65, 105].

Now we want to study the absorption by atoms in an arbitrary spin state m under the illumination of isotropic *unpolarized* light. As we shall see, it is independent of m . We write the absorption of state m as following:

$$\begin{aligned}
\sigma^*(m) &\propto W_{e \leftarrow g} \\
&\propto \frac{1}{3} \sum_{m', q} |\langle J', m' | d_q | J, m \rangle|^2 \\
&= \frac{1}{3} \sum_{m', q} \frac{|\langle J' = 9 || \mathbf{d} || J = 8 \rangle|^2}{2J' + 1} |\langle J, m, 1, q | J', m' \rangle|^2 \\
&= \frac{1}{3} \sum_{m', q} \frac{|\langle J' = 9 || \mathbf{d} || J = 8 \rangle|^2}{2J' + 1} \frac{2J' + 1}{2J + 1} |\langle J', m', 1, -q | J, m \rangle|^2 \\
&= \frac{1}{3} \frac{2J' + 1}{2J + 1} \frac{|\langle J' = 9 || \mathbf{d} || J = 8 \rangle|^2}{2J' + 1}
\end{aligned} \tag{B.2}$$

where in forth step the relation of CGs is used [105] and in the fifth step the sum rule of CGs is used.

We could see that $\sigma^*(m)$ is independent of m hence we denote $\sigma^* = \sigma^*(m)$. Comparing eqn. B.1 and eqn. B.2, we find that:

$$\sigma^* = \frac{1}{3} \frac{2J' + 1}{2J + 1} \sigma \tag{B.3}$$

This relation holds no matter whether the ensemble is in arbitrary spin polarized state or a spin mixture, as long as the illumination is isotropic and unpolarized.

Notice that the saturation intensity is $I_s = \frac{\hbar\omega A_{21}}{2\sigma}$ [64], where A_{21} is the Einstein spontaneous emission coefficient. We find that driving arbitrary spin states with isotropic unpolarized light is $\frac{3(2J+1)}{2J'+1}$ times harder as compared to a two-level system or an effective cycling closed transition. If we were to introduce the effective saturation intensity I_s^* based on the same spirit, we have:

$$I_s^* = \frac{3(2J + 1)}{2J' + 1} I_s = \beta I_s \tag{B.4}$$

, where the two level saturation intensity I_s can be calculated using $I_s = \pi \hbar c \Gamma / 3 \lambda^3 \tau$ [69], as long as linewidth of the transition is known. For 421 nm blue transition of Dy, $\beta = 2.68$ for bosonic isotopes ($J = 8$) and $\beta = 2.75$ for fermionic isotopes ($F = 21/2$). Within experimental error, $\beta = 2.7$ is a good choice for all Dy isotopes. This is the direct result of the high spin limit:

$$\lim_{J \rightarrow \infty, J' = J+1} \frac{3(2J+1)}{2J'+1} = 3. \quad (\text{B.5})$$

The above conclusion begins to deviate if: (i) light is not isotropic and unpolarized which hampers the direct use of sum rule of CG coefficients, as well as induces optical pumping among different Zeeman states; (ii) the external magnetic field leads to Zeeman shifts and different detunings among $\sigma^\pm, \pi$ transitions, and it breaks the proportionality between transition strengths and squares of CG coefficients; (iii) the electron transition is not closed and the branching ratio is less than unity. Corresponding corrections could be added accordingly, but no simple relation is available in general.

Appendix C

Linewidth Determination of the 421 nm Transition

Quantitative understanding of the population, dynamics, and cooling mechanisms [28] of the Dy MOT requires the accurate knowledge of the 421 nm transition's linewidth. To ensure the use of the correct value of the 421 nm transition linewidth in laser cooling calculations, we remeasured this linewidth using the crossed-beam method described earlier, though with a 421 nm beam derived from a frequency-doubled Ti:Sapphire laser.

To uniformly and stably scan the laser frequency, we employ the transfer cavity technique to lock the laser to a spectroscopic reference [27]. The optical transfer cavity is doubly resonant at 780 nm and 842 nm. The cavity itself is stabilized by locking a 780 nm ECDL to a hyperfine transition of the 780 nm D2 line in Rb before locking a resonance of the cavity to the stabilized ECDL. The Ti:Sapphire laser which generates the 842 nm beam is then locked to this cavity. In order to scan the Ti:Sapphire laser's frequency while the cavity remains locked to Rb, an electro-optical modulator driven by a microwave source generates tunable GHz-frequency sidebands on the 842 nm laser beam. By locking the sideband to the

cavity, the carrier frequency can be stably scanned via tuning the microwave source. The 421 nm laser beam is obtained from a resonant LBO doubler. In the experiment, the laser frequency is scanned 400 MHz with a period of 1 s to ensure that the bandwidth of the PIN photodetector does not artificially broaden the transition. The fluorescence was collected via a pair of 2" achromatic doublets mounted outside an AR-coated UHV viewport. The electronic detector output was recorded on a fast digital oscilloscope and averaged 64 times.

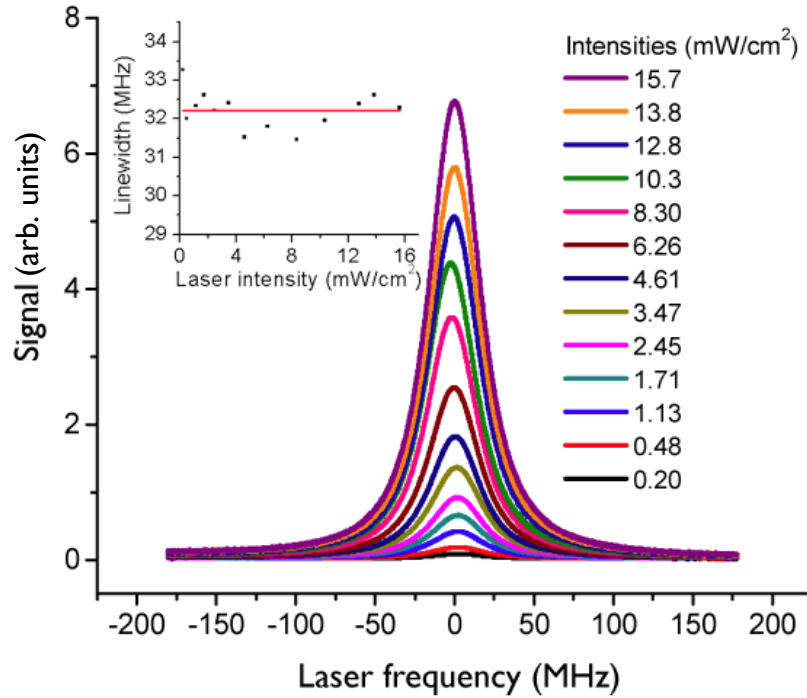


Figure C.1: Crossed-beam spectroscopy of the 421 nm transition. The photodetector signal is recorded as a function of 421 nm laser frequency, referenced to the line peak. The legend lists the laser intensities used in each measurement (highest laser intensity corresponds to highest signal). Each curve is averaged 64 times. Inset: Extrapolation of homogeneous linewidths to value at zero intensity.

The fluorescence versus frequency is shown in Fig. C.1. The profile has the typical Voigt form due to the residual Doppler broadening of the atomic beam. Curves corresponding to different laser beam intensities possess differing linewidths due to power broadening. A

Table C.1: 421 nm transition linewidth error budget.

Source	Uncertainty (MHz)
Extrapolation to zero intensity	0.13
Drift of laser during scans ¹	0.7
Laser linewidth	0.1
Rise time of detector	0.4
Zeeman shift (1st order)	+0.5 ²
Total	+0.9 -0.8

global Voigt fit allows a deconvolution of the Doppler width from the transition linewidth by assuming a single Gaussian Doppler width and by accounting for the power broadening from the laser. The fitted value for the Doppler broadening is 14.8(6) MHz, which is consistent with the estimation of the residual Doppler broadening based on the geometry of the collimation tube and oven orifice.

At low intensities, the power broadening is linear as a function of laser intensity. A linear fit to the extracted linewidths provides the natural linewidth at the zero-intensity limit [197]. The extrapolated value for the natural linewidth of the 421 nm transition is 32.2($^{+0.8}_{-0.9}$) MHz. The uncertainty in the linewidth measurement arises from the error sources listed in Table C.1. Among the errors, the largest source is the laser frequency drift from the imperfect resonator cavity lock.³ Unlike for Er [197], this measurement result is consistent with that listed in the standard tables, 33.1(17) MHz [77, 198].

¹Scan nonlinearity is negligible in transfer cavity technique.

²Residual magnetic field will broaden the linewidth depending on details of the probabilities in $\sigma^{\pm}, \pi$ excitations and an estimation is made accordingly which is always positive in linewidth correction.

³The transfer cavity lock precision has been improved by almost an order of magnitude since move to Stanford University, after this spectroscopic work.

Appendix D

Frequency Reference of Rb at 778 nm

In developing the optical frequency standard, we considered some atomic transitions with a narrower linewidth because this is advantageous for precisely determining the center frequency for a given SNR, which is normally limited in practice. We investigated the Rb 778 nm $5S_{1/2}-5D_{5/2}$ two-photon transition [199, 200], which has a natural linewidth of 300 kHz and is 20 times narrower than the Rb 780 nm D2 line. We obtained a preliminary spectrum of ^{85}Rb corresponding to one hyperfine manifold in the ground state, with a measured linewidth of 1.4 MHz. The residual broadening is likely be due to Zeeman shifts in an unshielded earth field, the frequency jitter of the free-running probe laser, as well as the transient broadening typically present in a probe beam with a finite width which is consistent with our estimation in the setup [199].

While this 778 nm Rb line is a narrow transition suitable for frequency determination, the detection setup is relatively more complicated than the modulation transfer spectroscopy technique.¹ Careful implementation of the 780 nm MTS lock provided a line splitting ca-

¹To achieve a good SNR, one needs to heat up the Rb vapor cell (90-100°C) for higher atomic densities, employ a high power ECDL (50-100 mW) or a cavity for power enhancement, use very careful optical filtering of 778 nm scattered lights ($\text{OD}>7$) and a sensitive detection of emitted 420 nm fluorescence via a photomultiplier.

pability of 1000 which provided a precision in frequency determination good enough for all current experiments.²

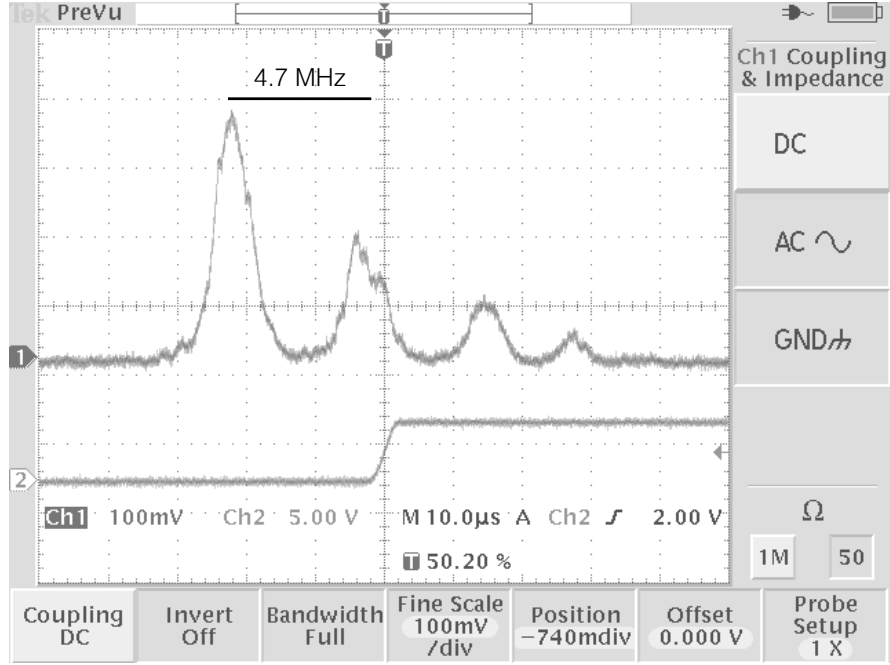


Figure D.1: Rb 778 nm two-photon spectroscopy. Fluorescence record of the spectrum of the $^{85}\text{Rb } 5S_{1/2}-5D_{5/2}$ ($F = 3 \rightarrow F' = 5$ to 1 from left to right). The peak for $F'=1$ is below noise background.

²The 778 nm spectroscopy may be employed for future improvement. The transfer between 780 nm D2 line and 778 nm two-photon spectroscopy is relatively easier since most of optics and laser source can be recycled, while to employ an Iodine frequency standard in the visible optical regime also provides excellent frequency stability [201], an extra laser source and the optics associated would need to be purchased.

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