1	Intro	oduction1
	1.1	Motivation2
	1.2	Rydberg Atoms and the Dipole-Dipole Interactions 3
	1.3	Atomic Units 3
	1.4	Dissertation Structure 4
2	Ехре	erimental Setup8
	2.1	Magneto-Optical Trap9
	2.1.:	Principle of the Magneto-Optical Trap9
	2.1.2	2 Saturated Absorption Spectroscopy
	2.1.3	3 Ultra High Vacuum Chamber16
	2.1.4	4 Characterization of the MOT16
	2.2	Lasers and Amplifiers
	2.2.	1 Nd:YAG Lasers
	2.2.2	2 Diode Lasers
	2.2.3	3 Mode-Lock Laser
	2.2.4	4 Chirped Pulse Amplification
	2.2.	5 Regenerative Amplifier
	2.2.0	6 Linear Amplifier24
	2.2.	7 Dye Laser and Dye Amplifier25
	2.3	THz Pulses
	2.4	Detection and Data Collection
	2.4.	1 State-Selective Field Ionization
	2.4.2	2 Synchronization System
	2.4.3	3 Measurement Operation

	2.5 Ma	intenance and Daily Operation	34
	2.5.1	Daily Examination	34
	2.5.2	Operation of Regenerative Amplifier	35
	2.5.3	Operation of the MOT	36
3	Models	in Simulation	41
	3.1 Ryc	lberg Atoms	42
	3.1.1	Modern Picture of Rydberg Atoms	43
	3.1.2	Properties of Rydberg Atoms	45
	3.2 Tw	o-Body Model	46
	3.2.1	Introduction to the Two-Body Model	46
	3.2.2	Nearest Neighbor Distribution	47
	3.2.3	Förster Resonance Energy Transfer	48
	3.3 Dip	ole-Dipole Interaction	49
	3.3.1	Dipole Moment	49
	3.3.2	Dipole-Dipole Interaction in the Classical Picture	50
	3.3.3	Dipole-Dipole Interaction in the Quantum Picture	52
	3.3.4	Dipole-Dipole Coupled System in an Electric Field	55
	3.4 Bla	ckbody Radiation Model	56
	3.4.1	Blackbody Induced Transition	57
	3.4.2	Radiation Model	57
4	Absence	e of Collective Decay in a Cold Rydberg Gas	61
	4.1 Int	roduction	62
	4.2 Exp	perimental Procedure and Results	65
	12 Am	alysis and Discussion	70

	4.4	Conclusion	79
5	Ryd	berg Wavepackets Evolution in A Frozen Gas of DD Coupled Atoms	85
	5.1	Introduction	86
	5.2	Experimental Procedure	88
	5.3	Experimental Results	90
	5.4	Discussion	93
	5.5	Conclusion	97

Figure 2.1: Simplified one dimensional model for MOT [2]
Figure 2.2: Schematic for MOT design. It's a combination of anti-Helmholtz coils and six
counter-propagating laser beams
Figure 2.3: Hyperfine energy structure of 85Rb. Trap laser drives the transition, $5s1/2$ F
= 3 to $5p3/2$ $F = 4$, and the repump laser drives the transition, $5s1/2$ $F = 2$ to $5p3/2$
F = 3.
Figure 2.4: The saturated absorption spectra for the Rb $5s1/2$ to $5p1/2$ hyperfine
transitions and their crossover peaks [3]. Small dark arrows are pointing to the real
resonances. Blue dotted arrow, pointing to the side of the crossover peak, indicates
where the trap laser is actually locked at. Red dotted arrow indicates the position where
the repump laser is locked at
Figure 2.5: Schematic of Nd:YAG lasing [6]. It is a typical four-level lasing scheme 18
Figure 2.6: Layout of Continuum Surelite Nd:YAG laser. It can output beams of 4 different
frequencies [5]
Figure 2.7: Basic layout of the mode-lock laser [12]. The green line is the pump light from
Millennia Vs laser and the red line is the mode-locked beam in the cavity which is
centered at 780-800 nm. The prism pair is used to compensate for group velocity
dispersion (GVD). By tapping the 2 nd prism, we can produce a temporary unstable
beam. Stronger intensity part in this beam will be enhanced, producing the periodic
pulsed output
Figure 2.8: Schematic of chirped pulse amplification system. Seed light is first temporally
stretched using the stretcher. Then the stretched pulse gets amplified. Finally the pulse

is again compressed to something close to its original duration, but with much greate
(106 – 107 times) energy
Figure 2.9: Schematic of regenerative amplifier. The switch-in pockels cell controls when
a seed pulse is trapped in the resonator and the switch-out pockels cell controls when
the pulse is ejected from the cavity.
Figure 2.10: Schematic of linear amplifier. The beam passes the Nd:YAG pumped gain
medium three times and gets amplified.
Figure 2.11: Schematic for a Hansch dye laser and the 2 nd harmonic generation. The angle
of the tuning grating determines the output frequency
Figure 2.12: Schematic for double cell dye amplifier used in the lab.
Figure 2.13: A typical THz pulse generated in the lab [8]. The duration is about 5 ps and
the peak frequency is 0.2-0.4 THz. This pulse is generated using tilted-pulse-front
pumping optical rectification and the temporal profile of the pulse is characterized
using the electro-optic sampling method. The frequency spectrum is then derived from
the time-domain profile.
Figure 2.14: Schematic of TPFP setup [8].
Figure 2.15: Schematic of the tipping of electron potential. Solid line is the $1/r$ potential
when there is no external field applied to the atom. Dashed line shows the tip o
potential when a filed is applied to the atom. When such a field is strong enough
electrons are able to escape from the trap
Figure 2.16: Schematic of the synchronization system. White circles are inputs and dark
circles are outputs.

Figure 2.17: A typical ionization signal shown on an oscilloscope. The central peak
representing the population of state $32s + 32p$. The measurement program uses a user
defined gate and integrates the area under the peak within the gate
Figure 2.18: Front view of the chamber where the MOT is positioned in. A, B, and C
represent three trap beams. 38
Figure 3.1: Classical view of Rydberg orbits of (a) H and (b) Na. In H the electron orbits
around the proton. In Na it orbits around the +11 nuclear charge and ten inner shell
electrons. In high L states Na behaves nearly identically to H, but in low L states the
Na electron penetrates and polarizes the inner shell electrons of the Na + core [5]. 43
Figure 3.2: Schematic of atom pairs in a MOT. For each atom in the MOT, we only consider
the effect of its nearest neighbor. One atom and its nearest neighbor is considered to
be "a pair of atoms".
Figure 3.3: Schematic for typical FRET. Black cycles represent the initial pair states and
gray cycles the final pair states. (a) is $pp \to ss'$, (b) $ps \to sp$ and (c) $ps' \to s'$
[13]49
Figure 3.4: Schematic of the interaction between two classical dipoles
Figure 3.5: Schematic for a DD system in an electric field
Figure 3.6: Decay model for atoms starting from state 40s as an example. The red dash
lines between two states indicate blackbody stimulated transitions between these two
states. The blue dash curves represent the spontaneous decay
Figure 4.1: (a), (c) Probabilities for finding atoms in $26s + 25p$ (green, fastest decay), $32s$
(red, intermediate decay), and 40s (blue, slowest decay) as a function of detection time
τ for Rydberg densities of ρ ~3×109 cm $-$ 3 (a) and ρ ~1.5×108 cm $-$ 3 (c).Note that
the sum of the 26s and 25p populations is shown since their corresponding features

could not be adequately separated in the field-ionization signal. Vertical bars show the experimental data with uncertainties, and the solid curves are calculated as described in the text. Measurements and calculations for the 40s decay extend to $500 \, \mu s$ where the remaining population is negligible. (b), (d) Probabilities for finding atoms in 26p (green, fastest rise and decay), 32p (red, intermediate rise and decay), and 40p (blue, slowest rise and decay) levels as a function of detection time τ . The states are populated by blackbody redistribution from the initial 26s, 32s, and 40s levels, respectively. The data were measured simultaneously with those shown in (a) and (c). Vertical bars show the experimental data with uncertainties, and the solid curves are calculated as described in the text. The measured p-state probabilities are normalized to the calculations as described in the text. The calculations have no free parameters and consider only the effects of spontaneous emission and blackbody radiation on isolated atoms.

Figure 4.3: Measured 32p3/2 |mj| = 1/2 (bold line) and |mj| = 3/2 (thin line) excitation probabilities as a function of Rydberg laser frequency in zero applied field. The two data curves are obtained simultaneously in the same laser frequency scan. The small feature on the left (right) of the main |mj| = 1/2 (3/2) peak is the result of imperfect discrimination of the |mj| - 1/2 and 3/2 components via SSFI. The additional peak on

the right of the main feature in each trace is due to the trap-laser dressing of the $5p3/2$
and 5s levels. Its frequency shift from the main peak reflects the Autler-Townes
splitting of the $5p3/2$ initial state76
Figure 4.4: Difference (i.e., splitting) in the transition energies for exciting $32p3/2 mj $ =
1/2,3/2 from $5p3/2$ as a function of applied electric field. Filled circles are
measurements and the solid curve is the result of a numerical Stark map calculation
assuming orthogonal "offset" and "residual" electric field components due to the MCP
of 2.8 and 1.5 V/cm, respectively. The inset shows a magnified view of the portion of
the main figure within the dashed window
Figure 5.1: Measured population in the combined 32s+31p states as a function of the delay
Δt between two THz pulses. The left panels show data collected at low Rydberg
density, $\rho \sim 3 \times 108$ cm -3 , for (a) short ($\Delta t \approx 0$) and (b) long ($\Delta t \approx 15$ ns) delays,
respectively. The right panels show data collected at high Rydberg density, ρ ~ 2 \times
$109 \text{ cm} - 3$, for c) short ($\Delta t \approx 0$) and d) long ($\Delta t \approx 15 \text{ns}$) delays, respectively. The
decrease in oscillation amplitude at high density and long delays is apparent91
Figure 5.2: Fast Fourier transform (FFT) of the delay-dependent populations shown in
Figure 5.1. 92
Figure 5.3: Measured (filled circles) and simulated (solid curve) decay ratio, η , as a function
of Rydberg density93
Figure 5.4: Schematic energy level diagram for the eigenstates of a pair of two level atoms.
The diagrams on the left and right sides of the figure depict the situation at large and
small interatomic spacing, R, respectively95

1 Introduction

Rydberg atoms are coupled by long-range dipole-dipole interactions [1]. They attract a lot of attention for their important role in studying fundamental problems as well as potential applications in quantum control in few- and many-body systems, quantum information processing, quantum calculation, etc. [2-10]. The research discussed in this dissertation is focusing on the influence of the DD interactions on the electron dynamics of cold Rydberg atoms. Three projects are presented in this dissertation, including both introduction of experimental setup and simulation results, respectively. Common experimental setups and background knowledge are also provided.

1.1 Motivation

One of my lab's main efforts is to explore and manipulate quantum dynamics in atomic and molecular systems. A series of projects have been or are implemented to accomplish this effort. These projects help people to understand fundamental science in atomic physics. More than that, they may contribute to potential practical applications from designing many-body systems that simulate model condensed matter systems, to quantum information storage and processing, to the development of new radiation sources and detectors.

Work done by previous graduates Xiangdong Zhang [11] and Mary Kutteruf [12] have pushed the effort a big step. The project "Probing Time-Dependent Electron Interactions in Double Rydberg Wavepackets" done by Xiangdong Zhang helps us understand more about electron interactions within individual atoms. The project "Coherence in Rydberg Atoms: Measurement and Control" done by Mary Kutteruf extended the exploration to the interactions between electric fields and wavepackets in an ensemble of Rydberg atoms. My project is an extent of their work but also a relatively independent research, which is to explore the interactions between Rydberg atoms within an ensemble. Another current graduate student Brian Richards is working on utilizing such interactions. Our projects are like puzzle chunks, working together to make the whole picture more complete.

My project to explore the influence of dipole-dipole interactions on wavepackets of Rydberg atoms has been divided into three sub-projects. First one is to explore the role of DD interactions in suppressing generation of collective decay in an ensemble of cold Rydberg atoms (Chapter 5). The second one is to explore the influence on wavepackets' evolution by DD interactions (Chapter 6). And the final one is to explore the coherence transfer via DD interactions (Chapter 7). Those three sub-projects help us understand better about what the DD interactions do in an ensemble of cold Rydberg atoms.

1.2 Rydberg Atoms and the Dipole-Dipole Interactions

Rydberg atoms are atoms in which an electron is excited to a state with a high principal quantum number. Usually this number is larger than 10. They are good systems for studying atom dynamics. The details about Rydberg atoms and their properties can be found in references such as [1], and are described in Chapter 3.

Dipole-Dipole interactions can influence neighboring Rydberg atoms. Classically the interaction is the result of the electric forces between electrons and the positively charged ion cores to which they are bound. When coupled by dipole-dipole interactions, Rydberg atoms should not be considered as individuals but rather a system. Details of dipole-dipole interactions, both from classical view and quantum physics prospective, can be found in Chapter 3.

1.3 Atomic Units

Atomic units (au or a.u.) are commonly used in atomic physics research. For convenience, we define:

$$\hbar = m_e = e = 4\pi\varepsilon_0 = 1 \tag{1.1}$$

where \hbar is Planck's constant divided by 2π , m_e is the mass of the electron, -e is the charge. We obtain the conversion factors between a.u. and SI units as shown in Table 1.1.

Quantity	Value in atomic units	Value in SI units
Length	1	5.2917721092(17)×10 ⁻¹¹ m
Energy	1	4.35974417(75)×10 ⁻¹⁸ J
Time	1	$2.418884326505(16) \times 10^{-17} \text{ s}$
Velocity	1	2.1876912633(73)×10 ⁶ m·s ⁻¹
Force	1	8.2387225(14)×10 ⁻⁸ N

Temperature	1	3.1577464(55)×10 ⁵ K
Pressure	1	2.9421912(19) ×10 ¹³ Pa
Electric field	1	5.14220652(11)×10 ¹¹ V·m ⁻¹
Electric potential	1	2.721138505(60)×10 ¹ V
Electric dipole moment	1	8.47835326(19)×10 ⁻³⁰ C·m
Magnetic field	1	2.35×10 ⁵ T

Table 1.1: Atomic units to SI units conversion factors.

1.4 Dissertation Structure

Subsequent chapters describe the experimental approach, numerical simulations and several independent projects. Each project contains both experimental description and simulation description. Additional details and common aspects of all experiments are in chapters 2 and 3.

Chapter 2 provides the information about experimental setups. It introduces the apparatus commonly used in the experiments, as well as the daily operation. More details about some instruments could be found in their respective manuals and the dissertations from previous students who worked in this lab.

Chapter 3 talks about physics concepts commonly involved in the experiments, and their mathematical expression in simulations. It is not practical to put every simulation code line used in this dissertation, but by following the models described in Chapter 3, one could reconstruct the simulations in a fairly straightforward way.

Chapter 4 describes a search for the collective decay in cold Rydberg gases. We found no evidence for superradiance, despite the results reported by other groups. Our analysis suggests that, due to dipole-dipole dephasing, our null result is the expected one.

Chapter 5 describes an exploration of Rydberg wavepacket evolution in dipole-dipole coupled atoms. Our results show, through experiment and simulation, that in dipole-dipole coupled atoms, the wavepackets are not evolving independently, and variations in the coupling strength between pairs of atoms lead to macroscopic dephasing of the electronic wavepacket motion.

Chapter 6 describes a study of coherence transfer between wavepackets on different atoms via resonant dipole-dipole interactions. We observe evidence for the development of wavepacket motion in one set of atoms driven by wavepacket evolution in neighboring atoms. We confirm the coherence transfer by measuring the relative phase between the initial and induced wavepacket oscillations, suggesting the creation of entangled atom pair states with a dynamically evolving Rydberg wavepacket on one, and only one, atom in each pair. Simulations support our interpretation.

Chapter 7 summarizes the work done in this dissertation and briefly discusses the work in the future.

Bibliography

- [1] Thomas F. Gallagher. Rydberg Atoms. Cambridge University Press (1994).
- [2] M. D. Lukin, M. Fleischhauer, R. Cote, L. M. Duan, D. Jaksch, J. I. Cirac, and P. Zoller, Phys. Rev. Lett. 87, 037901 (2001).
- [3] D. Tong, S. M. Farooqi, J. Stanojevic, S. Krishnan, Y. P. Zhang, R. Cote, E. E. Eyler, and P. L. Gould, Phys. Rev. Lett. 93, 063001 (2004).
- [4] F. Robicheaux and J. V. Hernandez, Phys. Rev. A 72, 063403 (2005).
- [5] T. Cubel Liebisch, A. Reinhard, P. R. Berman, and G. Raithel, Phys. Rev. Lett. 95, 253002 (2005).
- [6] T. Vogt, M. Viteau, J. Zhao, A. Chotia, D. Comparat, and P. Pillet, Phys. Rev. Lett. 97, 083003 (2006).
- [7] E. Urban, T. A. Johnson, T. Henage, L. Isenhower, D. D. Yavuz, T. G. Walker, and M. Saffman, Nat. Phys. 5, 110 (2009).
- [8] Alpha Gaëtan, Yevhen Miroshnychenko, Tatjana Wilk, Amodsen Chotia, Matthieu Viteau, Daniel Comparat, Pierre Pillet, Antoine Browaeys, and Philippe Grangier, Nat. Phys. 5, 115 (2009).
- [9] T. Wilk, A. Gaetan, C. Evellin, J. Wolters, Y. Miroshnychenko, P. Grangier, and A. Browaeys, Phys. Rev. Lett. 104, 010502 (2010).
- [10] D. Jaksch, J.I. Cirac, P. Zoller, S.L. Rolston, R. Cote, and M.D. Lukin, Phys. Rev. Lett. 85, 2208 (2000).
- [11] Xiangdong Zhang, Probing Time-Dependent Electron Interactions in Double Rydberg Wavepackets, PhD thesis, University of Virginia (2008).
- [12] Mary Kutteruf, Coherence in Rydberg Atoms: Measurement and Control, PhD thesis, University of Virginia (2010).

- [13] A. Dalgarno, Rydberg States of Atoms and Molecules, eds. R. F. Stebbings and F.B.Dunning, Cambridge University Press (1983).
- [14] H. A. Bethe and E.A. Salpeter, Quantum Mechanics of One and Two Electron Atoms, Academic Press (1957).

2 Experimental Setup

This chapter contains the general experimental setup for the research discussed in this dissertation. It introduces the apparatus and procedure for state excitation, laser cooling, pulse amplification, THz pulse generation, data collection, etc. It also provides procedures for maintenance and daily operation. All experiments are performed on Newport RS 3000 optical tables to reduce mechanical vibrations, in a temperature controlled room to reduce external thermal fluctuations. Other than specifically noted, the repetition rate of all experiments is 15 Hz. Before beginning experiments in the lab, participants must have taken the laboratory safety training.