1 Introduction

The electronic and nuclear motion degrees of freedom in Rydberg atoms are coupled by long-range dipole-dipole interactions [1]. This coupling is responsible for rich few- and many-body quantum dynamics and its coherent manipulation can enable potential applications in the quantum control of few- and many-body systems, quantum information processing, quantum computing, etc. [2-10]. The research discussed in this dissertation focuses on the influence of dipole-dipole (DD) interactions on electron dynamics within cold Rydberg atoms. This is experimentally challenging due to the long separation of time and distance scales associated with the electronic and nuclear motion, respectively. As summarized below, the result of three sub-projects are described in this dissertation. Details of the common experimental setups, computational approaches, and background knowledge are also provided.

1.1 Motivation

Our group's primary research involves the exploration and manipulation of quantum dynamics in atomic and molecular systems. This dissertation describes a series of sub-projects designed to further this effort. Our results provide new insight into the fundamental problems in atomic physics. More than that, they may contribute to 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.

Recent work done by previous graduates Xiangdong Zhang [11] and Mary Kutteruf [12] provided much of the foundation and motivation for the work presented here. The project "Probing Time-Dependent Electron Interactions in Double Rydberg Wavepackets" done by Xiangdong Zhang helps us understand more about time-dependent electron-electron interactions within individual atoms. The project "Coherence in Rydberg Atoms: Measurement and Control" done by Mary Kutteruf explored the use electric fields to control and measure coherence in electronic wavepackets and between coupled atoms in Rydberg ensembles. My project is an extension of their work but is the first to explicitly examine the impact of dipole-dipole interactions between atoms on electron wavepacket dynamics within them. Another current graduate student Brian Richards is working on utilizing controlled DD interactions to manipulate the position correlation function of cold trapped atoms. Our projects are like puzzle chunks, working together to develop new capabilities and make the scientific picture more complete.

My project exploring the influence of dipole-dipole interactions on Rydberg wavepackets has been divided into three sub-projects. The first one explores the role of DD interactions in suppressing collective decay ("superradiance") in an ensemble of cold Rydberg atoms (Chapter 4). The second one characterizes the role of DD interactions in dephasing the macroscopic coherence of an ensemble of Rydberg wavepackets (Chapter 5). The final one examines the transfer of wavepacket coherence between atoms via DD interactions (Chapter 6). These three sub-projects help us understand better how the DD interactions influence electron dynamics in an ensemble, and explore the use of controlled atom-atom coupling to induce coherent wavepacket motion within those atoms. The latter provides an effective demonstration of coherent control beyond unimolecular photo-reactions.

1.2 Rydberg Atoms and 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 (DD) 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 DD interactions, Rydberg atoms should not be considered as individuals but rather a system. Details of DD interactions, both from a classical view and a 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 electron charge and ε_0 is the permittivity of free space. The conversion factors between a.u. and SI units are shown in Table 1.1.

Quantity	Value in atomic units	Value in SI units
Length	1	$5.2917721092(17)\times10^{-11}$ m
Energy	1	$4.35974417(75)\times10^{-18}$ J
Time	1	$2.418884326505(16)\times10^{-17}$ s
Velocity	1	$2.1876912633(73)\times10^{6}$ m·s^{-1}
Force	1	$8.2387225(14)\times10^{-8}$ N
Temperature	1	$3.1577464(55)\times10^{5}$ K
Pressure	1	$2.9421912(19)\times10^{13}$ Pa
Electric field	1	$5.14220652(11)\times10^{11}$ V·m^{-1}
Electric potential	1	$2.721138505(60)\times10^{1}$ V
Electric dipole moment	1	$8.47835326(19)\times10^{-30}$ 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 distinct projects. Each project contains both an experimental description and details on relevant simulations. Additional introductory material and common aspects of all experiments are found 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 daily operation procedures. More details about some instruments can be found in their respective manuals and the dissertations from previous students who worked in this lab.

Chapter 3 introduces physics concepts commonly involved in the experiments, and their mathematical expression in simulations. It is not practical to include in this dissertation every single line of simulation code that was used during the research, 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 collective decay (i.e. "superradiance") 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 for highly excited Rydberg atoms in the typical magneto-optical-trap geometry.

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 do not evolve 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 possibilities for future experiments following these results.

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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.