Experiments performed under this grant were designed to exploit the extreme electric field sensitivity of highly-excited Rydberg atoms to test methods for asserting coherent control over inter- and intra-atomic dynamics. The work focused on the application of Rydberg wavepacket techniques to investigate information storage and processing within model quantum systems. New methods for manipulating and probing quantum dynamics in Rydberg atoms were developed and used to read and write information to and from Rydberg data registers, and to protect quantum information from decoherence caused by unmeasured environmental interactions. Specifically: (i) intense, short electromagnetic pulses were used to read classical information stored in the momentum-space probability distributions of single Rydberg electrons; (ii) well-controlled electric fields were utilized to exploit the coupling of electron spin and orbital angular momentum in Rydberg fine-structure doublets to fabricate arbitrary quantum bits and implement universal single qubit logic gates; and (iii) the effectiveness of several different schemes for reducing the deleterious action of decoherence on qubits stored in Rydberg atoms was examined.
1. Introduction and Motivation
Most atomic physics research has moved beyond passive spectroscopy of isolated atoms. Instead, lasers and other coherent fields are now commonly used as tools for active manipulation and interrogation of inter- and intra-atomic dynamics. Optical control methods are enabling entirely new classes of atomic physics research, with fundamental science and practical applications from designer many-body systems that simulate model condensed matter Hamiltonians, to quantum information storage and processing, to new radiation sources and detectors.

Our experiments are designed to exploit the extreme electric field sensitivity of highly-excited Rydberg atoms for testing new methods for controlling quantum dynamics. In contrast to ground- or low-lying excited-state atoms, the application of non-resonant, time-dependent electric fields of a few to a few thousand V/cm can severely alter the electronic wavefunctions within Rydberg atoms. Well-characterized control fields used in conjunction with broad-band laser pulses, can facilitate the creation of coherent superposition-states (i.e. wavepackets) whose evolution can be precisely steered to affect inter- or intra-atomic processes.

In the following section we provide a brief overview of our methods for creating and probing Rydberg atoms. We then summarize the results obtained with AFOSR support from December 15, 2004 to December 14, 2007. We note that a no cost extension was requested and approved for this grant, FA9550-05-1-0064, moving the official end date to December 2008. However, the NCE was requested solely to maintain fiscal continuity within the University of Virginia system until funds for the renewal grant, FA9550-08-1-0066, were allocated. All funds from FA9550-05-1-0064 were exhausted prior to December 2007 and, therefore, results obtained after December 15, 2007 are attributed to the renewal grant.

2. Experimental Methods
All of the experiments performed under this AFOSR project involved ensembles of Rydberg atoms, identically prepared in vacuum. Ground-state alkali or alkaline-earth atoms in a low density ( <10^{12} cm^{-3} ) atomic beam were laser excited to Rydberg states with principal quantum numbers, 15<\textless n<40. Excitation of Rydberg eigenstates was performed as needed using nanosecond pulsed dye lasers. For other measurements, non-stationary superposition states were coherently populated using short, broad-band laser pulses. These pulses were obtained directly from the fundamental or low-order harmonics of a 120 fsec, 790 nm Ti:Sapphire amplifier. Standard pulse-shaping methods were used to modify the spectral-phase and amplitude of the short laser pulses, enabling fine control over the properties of the Rydberg wavepackets.

Subpicosecond THz pulses were also employed as tools for modifying and probing Rydberg electron dynamics. These pulses are also useful for a variety of other applications from time-domain THz spectroscopy, to THz imaging, to coherent control of the orientation of polar molecules. In our
laboratory, high power THz pulses are produced by gating biased, large aperture, GaAs photoconductive switches with 120 fsec, 790 nm laser pulses. The freely propagating radiation produced by each gate pulse has the form of a nearly unipolar “half-cycle” electric field pulse (HCP). The HCP field is linearly polarized parallel to the bias field across the GaAs wafer, and its amplitude is proportional to the bias voltage. HCPs are commonly used in our laboratory to create and probe Rydberg wavepackets via impulsive excitation and ionization, respectively.

Experiments are performed using atomic beam sources enclosed in vacuum chambers. In each chamber, the laser/atom interaction region is positioned between a pair of parallel electric-field plates. During and/or after their initial excitation, Rydberg atoms are exposed to additional laser pulses and THz or rf fields. These coherently alter the Rydberg wavefunction to induce specific dynamics or probe wavepacket evolution as required for a given experiment. A 1GS/sec arbitrary waveform generator (AWG) is available to produce complex rf waveforms for this purpose. After each laser shot, a high-voltage ramp applied to the field plates pushes any ions in the interaction region toward a microchannel plate detector. The ions may be created via photoionization due to a laser or HCP probe, or may be produced via field-ionization of Rydberg states during the high-voltage ramp. The time-dependent ion signal is used to determine the Rydberg-state energy distribution and the relative abundance of Rydberg atoms and ions in the interaction region at the instant the field ramp is applied. Experiments are designed so that probe pulses encode relevant information regarding the Rydberg excitation strength or wavepacket dynamics into the photoionization probability or the final energy distribution of the Rydberg electrons.

Some experiments utilize an imaging detector consisting of a microchannel plate stack backed by a phosphor coated anode to facilitate the measurement of the spatial distribution of ions and Rydberg states within the interaction region. The fluorescence at each point on the phosphor screen is proportional to the ionization probability at a particular location within the interaction region. By mapping one or more experimental parameters (e.g. pump-probe time delay, electric field amplitude, laser wavelength, etc.) onto different spatial dimensions within the interaction region, many parallel measurements can be made in a single laser shot.

3. Accomplishments

Given the great current interest in exploiting the quantum nature of matter to enable new paradigms in computing and information processing, during the grant period, our AFOSR research focused on the application of Rydberg wavepacket control techniques to investigate information storage and processing within these model quantum systems. Accordingly, we developed new methods for manipulating and probing quantum dynamics in atoms. We utilized these techniques to read and write information to and from Rydberg data registers, and to protect quantum information from decoherence caused by unmeasured environmental interactions. Specifically, we: (i) used intense, short electromagnetic pulses to read classical information stored in the momentum-space probability distributions of single Rydberg electrons; (ii) utilized well-controlled electric fields to exploit the coupling of electron spin and orbital angular momentum in Rydberg fine-structure doublets to fabricate arbitrary quantum bits and implement single qubit logic gates; and (iii) examined the effectiveness of several different schemes for reducing the deleterious action of decoherence on qubits stored in Rydberg atoms. We also constructed a Rb magneto optical trap (MOT) which is being used to explore coherent interactions between cold Rydberg atoms, a focus of the renewal grant.
A. Probing Classical Information Stored in Electronic Probability Distributions

The availability of sophisticated methods for engineering and visualizing electronic wavefunctions with specific spatial structure or eigenstate composition has opened the door to the possibility of reading and writing "atomic bar-codes," on individual atoms. The nodal structure of the momentum-space probability distribution of a single Rydberg electron is a potential candidate for testing this notion. Accordingly, we recently demonstrated significant improvements in the resolution of our impulsive momentum retrieval (IMR) technique which might serve as a bar code reader. In principle, the electronic momentum distribution for a bound electron at any instant in time can be recovered, exactly, from repeated destructive measurements of the probability for ionizing the electron as a function of the strength of an instantaneous impulse. In that case, the momentum distribution is a scaled derivative of the ionization probability vs. impulse curve. In the laboratory, we can extract approximate momentum distributions using physically realizable HCPs with durations much less than the characteristic time-scale for electron motion on the atom.

The sensitivity achieved in our original demonstrations of this IMR technique was insufficient to resolve quantum nodal structure. However, by using HCP amplitude modulation for direct differentiation of the ionization probability and by imaging the Rydberg ionization probability across the spatial profile of the ionizing HCP beam, we have recently demonstrated that momentum distributions can be recovered without scanning the HCP field strength. By eliminating errors associated with experimental drifts during a HCP ionization scan, this new method has dramatically improved our ability to view fine structure in electronic probability distributions within Rydberg atoms. As shown in Fig. 1, we recently imaged small features associated with nodal structure in the probability distributions for certain Rydberg Stark states. Furthermore, we developed a semiclassical model which accurately predicts the momentum image distortions that occur for some distributions. These distortions are caused by the aliasing of high momentum features to low momentum due to the motion of the electron during the non-zero duration of the HCPs. Extremely accurate images can only be obtained for high angular momentum electron distributions which have negligible probability density at small radius (i.e. large momentum). Thus, our improved IMR method might also be profitably applied for imaging the motion of high angular momentum wavepackets whose dynamics cannot be accurately measured using optical frequency probes.

Figure 1: Momentum distributions for two n=28 Rydberg Stark eigenstates in calcium. The quantum numbers k = -6 and k = -26 correspond to states with very small and very large negative dipole moments, respectively. The 3 different curves in each figure correspond to: the approximate distributions measured using the IMR method and a 500 fsec HCP (solid line); a semiclassical model that takes into account the motion of the electron during the HCP to simulate the experimental distributions (dashed line); and the "exact" momentum distributions (dotted line).

A manuscript describing the improved IMR method, new measurements enabled by it, and our semiclassical analysis of the pseudo-impulsive ionization has been published [J. Murray-Krezan and R.R. [insert citation here].
B. Creating Arbitrary Qubits and Implementing Single-Qubit Logic Gates in Rydberg Atoms

We have developed techniques to create and manipulate single quantum bits (qubits) in coherent superpositions of np fine-structure states in Li. We employ nanosecond dye lasers to excite atoms in a thermal beam, producing ensembles of identical, single-electron Rydberg wavepackets. In each excited atom, the electron is placed in a coherent superposition of np1/2 and np3/2 fine-structure states with |m_j|=1/2 [28]. The admixture of the orbital angular momentum states, |m_L| = 0,1 in the two fine-structure eigenstates as well as the energy splitting, ε, between the two eigenstates can be controlled by applying a small static electric field to the atoms. In a weak electric field (~0.4 V/cm for n = 28), the two eigenstates are equal admixtures of the m_L = 0 and |m_L| = 1 states. Moreover, due to the spin-orbit coupling, the energy splitting between the two fine-structure states is roughly independent of small variations in electric and magnetic fields. Apparently the spin-orbit interaction creates an approximate “decoherence free subspace” (DFS) in which the wavepacket is virtually immune to dephasing caused by stray fields.

Qubits with arbitrary amplitude and phase can be produced using a fast rising electric field pulse (~ 2 V/cm at n = 28) at a time, t = t_0. The pulse decouples the electron spin and orbital angular momentum and projects the wavepacket onto the |m_L| = 0,1 eigenstates. Thus, we can specify the state vector, |ψ_m(T)> = [cosθ , i sinθ e^(iφ(T))] , which represents our qubit at times T = t-t_0. The qubit amplitude, cosθ, depends on the phase acquired in the excitation basis, $θ = \int_0^T E dt$ , and can be controlled by varying the projection time, t_0. The qubit phase, φ(T) = $\int_0^T E dt$ , is acquired at a rate equal to the energy difference E between the two qubit levels in the new, m_L basis.

We confirm our ability to create arbitrary qubits by measuring the qubit amplitude and phase. The qubit amplitude is obtained directly from field ionization measurements which can distinguish between electron population in the m_L = 0 and |m_L| = 1 states. To measure the qubit phase, we construct an intra-atomic interferometer from a sequence of electric field pulses which serve as as quantum-state “beam” splitters and combiners. The combined operation is equivalent to a √NOT quantum logic gate. The interferometer converts the phase difference, φ(T), between two input states, [m_L] = 0 and 1, into an amplitude difference between two output states in the same basis. Provided all atoms in the ensemble have the same phase, then the difference in the measured probabilities P_0 and P_1 for finding the atom in the |m_L| = 0 and 1 states is P_0 - P_1 = sin[2θ] sin[φ(T)]. However, due to environmental decoherence and inhomogeneous dephasing, atoms in the ensemble acquire phase at different rates, such that the amplitude, C(T), of the T-dependent oscillations in P_0 and P_1 decrease with increasing T. As described in more detail below, the amplitudes of these oscillations provide a direct measure of the coherence of the system.

For short times though, decoherence is negligible, and electric field pulse sequences can be used to implement quantum logic operations on the ensemble of fine-structure qubits. Indeed, we have demonstrated high-fidelity universal quantum NOT, √NOT gates and Hadamard transforms for single qubits. As noted above, we have used these gates as tools to manipulate and measure the qubit amplitude and phase.
In addition, we have created Rydberg populations in which two distinct qubits have been simultaneously encoded (in np and n'p doublets). We have shown that we can selectively apply (with slightly reduced fidelity) universal quantum operations to one qubit without affecting the other. We have also developed a controlled-NOT (CNOT) gate that will or will not flip the qubit amplitudes \([\cos\theta, \sin\theta] \rightarrow [\sin\theta, \cos\theta]\) depending on the value of the qubit phase, \(\varphi\). Unfortunately, high fidelity operation of this gate appears to be possible for only a limited range of qubit amplitudes. A manuscript describing the arbitrary Rydberg qubit preparation and universal quantum gate operation is in preparation and will be described in detail in Mary Kutteruf’s Ph.D. dissertation.

### C. Suppressing Decoherence in Rydberg Qubits

Decoherence is present at some level in any quantum system, and it represents a primary obstacle to realizing large-scale quantum information processing. Using Rydberg qubits, we have explored methods for controlling or eliminating decoherence. These techniques might find general application in more complex systems.

When measuring the phase of our arbitrary qubits, we find that \(C(T)\) decays (see Fig. 2) with increasing delay, \(T\). By applying a quantum NOT operation to the qubit at time \(T\), we observe a partial revival of \(C(T)\), a “quantum state echo” at a time \(2T\). This echo is analogous to spin echoes observed in NMR, and its amplitude \(C(2T)\) is a direct measure of the qubit coherence. In spite of the fact that we do not know the precise cause of the observed decoherence, by applying a sequence of NOT operations in rapid succession we have been able to maintain essentially perfect qubit coherence for the lifetime of the Rydberg atoms in our interaction region (see Fig. 2). This control technique is a form of “bang-bang” dynamical decoupling and is analogous to spin-flip narrowing in NMR. By rapidly flipping the state-vector, the pulse sequence eliminates the slower acquisition of phase noise within the atoms in the ensemble. A manuscript detailing these results has been published [R.S. Minns, M.R. Kutteruf, H. Zaidi, L. Ko and R.R. Jones, Physical Review Letters 97, 040504 (2006)].

![Figure 2: Oscillations in \(P_0\) as a function of the phase-measurement time, \(T\). The main figure shows signals observed with no NOT gates applied (black), a single NOT gate applied near 500 nsec (red), 17 NOT gates applied (green), and 63 NOT gates applied (magenta). The insets](image)
show temporally magnified views of the measured and predicted echoes following the 17 and 63 pulse sequences.

As noted above, the electric field-independence of the fine-structure states in weak fields makes this excitation basis an approximate decoherence-free subspace (DFS). In the time-domain, the formation of this DFS can be attributed to continuous flipping of the qubit caused by Rabi flopping between the $m_L=0$ and $|m_L|=1$ levels due to the spin-orbit interaction. Provided the acquisition of phase noise due to environmental field fluctuations is slow compared to the driven qubit flip-rate, the continuous flipping of the quantum-state results in a near perfect cancellation of decoherence, in direct analogy with the pulsed, bang-bang dynamical decoupling scheme. Indeed, the decoherence rate in the excitation basis (as determined from the decrease in the measured oscillation amplitudes in $P_0$ and $P_1$ as a function of the projection time, $t_0$) is considerably lower than in the (non-actively controlled) high-field basis. However, as shown in Fig. 3, decoherence does occur in the excitation basis. Interestingly, both the intrinsic DFS and the active bang-bang decoupling scheme exploit the spin-orbit interaction to flip the quantum state. In the bang-bang sequence however, this mechanism is only active for roughly half of the time. Therefore, it may seem surprising that the bang-bang pulse sequence is more effective in suppressing decoherence in our system. By choosing the delay between NOT operations the quantum state is flipped in both the high- and low-field bases, eliminating phase noise acquired in both. A similar approach might bolster the effectiveness of dynamic decoupling schemes in other systems as well.

We have also demonstrated that the coherence time of qubits in the low field basis can be extended through the use of continuous, rather than pulsed fields for dynamic decoupling. Specifically, the residual decoherence remaining in the approximate DFS is substantially suppressed by driving the qubit with an AC-field whose frequency is resonant with the energy splitting between the two constituent levels in the wavepacket. The AC-drive effectively phase-locks the qubit, preventing the acquisition of phase errors caused by environmental interactions. The mechanism is similar to spin-locking in NMR. In a dressed-state picture, this phase-locking can be viewed as an insensitivity of the qubit quasi-energy levels to small amplitude fluctuations. This active dynamic decoupling method should be applicable to extend qubit storage times in a variety of other systems. A manuscript describing these results has been published [R.S. Minns, M.R. Kutteruf, M.A. Comissionso, and R.R. Jones, Journal of Physics B 41, 074012 (2008).]

![Figure 3: Measured probability in the $m_L = 0$ state, following the sudden projection out of the excitation basis, as a function of the projection delay, $t_0$. The black curve shows the result without any rf field. The red and blue curves show the measured probability if the projection is performed immediately following the application of a resonant, 3.2 MHz rf pulse with an amplitude of 0.035 V/cm and a duration of 6.25 μs (red), or a 3.2 MHz rf pulse with an amplitude of 0.035 V/cm and a duration of 6.25 μs (blue).]
pulse with an amplitude of 0.045 V/cm pulse and a duration of 18.8 μs (blue). The oscillation amplitudes reflect the qubit coherence. The resonant rf radiation dresses the qubit so that it remains coherent for substantially longer times.

D. Development of a Rb Magneto Optical Trap

Postdoctoral researcher, Dr. Thibault Vogt, constructed from scratch, a Rb magneto-optical trap (MOT) which is being used in our continuing experiments on controlled, coherent interactions in nearly frozen Rydberg gases. The MOT chamber houses a microchannel plate detector with a phosphor screen anode for spatially resolved, state-selective detection of Rydberg atoms in the MOT. A set of parallel rods enables the application of uniform electric fields while maintaining optical access for the trapping lasers. The MOT was a critical addition to our laboratory and its development would have taken considerably more time had it not been for Dr. Vogt’s expertise.

4. Personnel

i) Total Support (December 15, 2004 – December 14, 2007)
- Prof. Robert Jones – Principal Investigator – 2.0 months summer salary
- Dr. Russell Minns – Postdoctoral Associate – 19 months salary + research supplies
- Dr. Thibault Vogt – Postdoctoral Associate – 7 months salary + research supplies
- Jeremy Murray-Krezan – Graduate Student - 17 month stipend + research supplies
- Mary Kutteruf – Graduate Student - 8 month stipend + research supplies
- Aaron Wallo – research supplies

ii. Personnel Transitions

During the report period Jeremy Murray-Krezan successfully defended his Ph.D. dissertation and Aaron Wallo completed the requirements for his M.S. degree. Dr. Murray-Krezan is currently employed at the Naval Research Laboratory while Mr. Wallo is working for the Naval Surface Warfare Center. Postdoctoral researchers Dr. Russell Minns and Dr. Thibault Vogt have taken second postdoctoral research positions at University College London and Peking University, respectively. Principal Investigator Robert Jones was promoted to Francis H. Smith Professor of Physics at the University of Virginia.

iii. Honors and Awards

Jeremy Murray-Krezan received a 12 month NSF IGERT fellowship (2005).
Mary Kutteruf received a 12 month NSF IGERT fellowship (2005).

5. Publications


A.W. Wallo, A Half Cycle Pulse Assisted p to p Transition in Li, M.S. Thesis, University of


Note: The three publications listed below resulted from work performed prior to the original ending date (Dec. 14, 2007) for grant FA9550-05-1-0064. However, these were published in 2008 and, therefore, are also included in the publication list for the renewal grant which began on Feb. 15, 2008.


6. Interactions and Transitions
Results from this project were described by the PI in 9 invited talks at conferences and university colloquia. In addition postdocs and graduate students presented our AFOSR supported results in 6 contributed talks at conferences.

7. Inventions or Patent Disclosures
None during the report period.