Half-Cycle Pulse Ionization of Diamagnetic Zeeman States in Rydberg Atoms.

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HALF-CYCLE PULSE IONIZATION OF DIAMAGNETIC ZEEMAN
STATES IN RYDBERG ATOMS

A Dissertation
Submitted to the Graduate Faculty of the
Louisiana State University and
Agricultural and Mechanical College
in partial fulfillment of the
requirements for the degree of
Doctor of Philosophy
in
The Department of Physics and Astronomy

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To my teachers
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ABSTRACT

We study the dynamics of highly excited (Rydberg) atoms in laboratory-scale magnetic fields via their ionization by THz half-cycle pulses (HCPs). A half-cycle pulse is a unipolar electromagnetic pulse that can probe an atomic electron at all points along its orbit around the atomic core. The dynamical evolution of diamagnetic Zeeman states subjected to an ultrashort (picosecond) HCP is calculated for both hydrogen and sodium atoms.

Diamagnetic Zeeman states are localized parallel and perpendicular to the magnetic field. We use a grid-based pseudo-potential method to determine these eigenstates of the combined Coulomb-diamagnetic potential. We study states with principal quantum number $n$ between $15 - 30$ in the $\ell$-mixing regime at magnetic fields of $1 - 6T$. These initial states are subjected to the electric field of an HCP and their subsequent time evolution is calculated by solving the time-dependent Schrödinger equation. The HCP width is varied between the short pulse (impulsive) and the long pulse limit, keeping the total momentum transferred a constant. We calculate the total ionized fraction, and also the spectrum of the ionized photoelectrons.

We find that, in the impulsive limit, where the width of the pulse is much smaller than typical time scales in the system, the calculated amount of ionization and the photoelectron spectra are different depending on the localization of the initial state. These differences are shown to be due to the different initial momentum distributions of the parallel and perpendicular states. As the duration of the HCP is made longer, we find that ionization...
is suppressed as compared with the impulsive limit. The states localized perpendicular to the magnetic field are found to be much more sensitive to the HCP width than the parallel states, reflecting the fact that the two classes of states interact with different parts of the Coulomb-diamagnetic potential during the HCP. The ionization characteristics are shown to scale classically with the principal quantum number. The results are presented in such a manner that they can be verified by experiment.
1. INTRODUCTION

The study of ultrafast phenomena is becoming increasingly important in scientific research. The availability of extremely short laser pulses allows scientists to study processes that change over small time scales, much like using a strobe light to produce a series of frozen frames of a quick movement. Ultrashort pulses are used in experiments that transcend disciplinary boundaries — to observe protein folding, to study responses of muscles to stimuli, to analyze molecular dissociation, in atomic spectroscopy and to create novel atomic states. Indeed, the 1999 Nobel prize for Chemistry was awarded for the study of chemical transformations using femtosecond laser pulses. The theoretical understanding of ultrafast dynamics in physical systems is, therefore, very important.

We are interested in the dynamics of a class of physical systems which are abundant in nature and in modern technology — atoms in electric or magnetic fields. In medical imaging techniques such as magnetic resonance imaging (MRI), atoms are subject to a magnetic field strength of about one Tesla whereas atoms found in distant pulsars are subject to roughly one hundred million ($10^8$) Tesla. Atoms have been subject to external electric fields in the arc and vapor lamps of the past as well as the sophisticated lighting systems of today. With state of the art technology, not only can we apply strong, static (constant) electric and magnetic fields but also time-varying ones of intense lasers. The study of atoms in both static and dynamic external fields of a large range of field strengths is extremely challenging.
To understand atomic systems, physicists probe them by exciting the atoms and studying the light emitted as they de-excite (the excitation spectra) or by detaching one or more electrons from the atom and measuring the energy and angular distribution of the electrons (ionization spectra). Very intense and ultrashort laser pulses can be produced in the laboratory which allow extremely fast and deep probing of atomic systems. In order to understand ultrafast processes, it is important to study not only the stationary properties of the atoms themselves, but also their dynamical response to rapidly changing external stimuli. Optical pulses with special properties (for example, "half-cycle pulses") can also be made with which very delicate measurements of even individual atomic states can be performed. The study of atomic systems in static as well as dynamic (time-varying) external fields is, broadly, the subject of this dissertation.

1.1 Historical Background and Motivation

The first experiments with atoms in external fields were performed near the beginning of the twentieth century. The spectrum of atoms in a static magnetic field, the Zeeman effect [1], was measured in 1896 and of atoms in a static electric field, the Stark effect [2], was measured in 1913. At that time, quantum mechanics was still in its nascent stages of development and the Zeeman effect was explained by classical vector models of the atom [3]. One of the successes of the early Bohr quantum model was the first satisfactory treatment of the hydrogen atom in an electric field [4]. Interestingly, these early theoretical results were little altered by later calculations in 1926 with the stationary state perturbation theory of quantum mechanics [5]. The
success of perturbation theory in explaining these effects was due to the relatively low strength of the laboratory fields available then ($\approx 100$KV/cm, 5000 gauss) compared to the strong fields experienced by the electron moving around the nucleus (5.142 million KV/cm, 2350 million gauss). The external field then acts as a small disturbance (perturbation) to the electron which is bound strongly to the nucleus.

In 1939, Jenkins and Segre first observed a broadening of the spectral lines of barium in the presence of a strong magnetic field [6]. In a companion paper [7], Schiff and Snyder explained this effect using a perturbative theory - the quadratic or diamagnetic Zeeman effect (so named because the energy was proportional to the square of the magnetic field). However, the resolution of the spectroscopes available at that time was low and further progress required technical developments. In 1969, Garton and Tomkins [8] used instruments with high dispersion to resolve the diamagnetic Zeeman spectrum. They also made the first observations of the Quasi-Landau spectrum — equally spaced absorption peaks around and beyond the ionization limit. In between these two regions, namely, the low energy region with the resolved diamagnetic spectrum and the near-threshold region of the Quasi-Landau levels, they observed a region where the spectral structures were “confused”. This was termed the region of inter-$n$ mixing because the diamagnetic energy mixes states of adjacent $n$ — the principal quantum number. With the increase in technological sophistication, experiments were performed to map all the three regions with extremely high resolution [9] and the results were compared with numerical calculations. Classical mechanics of an atom in
a magnetic field predicted that there were regions in these maps that corresponded to regular behavior and others which corresponded to irregular behavior of the diamagnetic atom [10]. Some features of the Quasi-Landau region were explained using such classical models and the ‘confusion’ in the structure in the n-mixing regime was attributed to a transition from order to chaos in the classical diamagnetic system [11].

Since the early days, laboratory fields have become much stronger and the availability of lasers and high vacuum have permitted experiments with much higher resolution. These phenomena can no longer be completely explained by classical or semiclassical models and a theoretical description using quantum mechanics is needed. Perturbation theory, which was extremely successful at low field strengths, becomes inefficient at higher field strengths. A variety of non-perturbative methods have been developed to study atoms in external fields. Further, for an atom with loosely bound electrons, i.e., electrons in excited states, even relatively low external fields have a non-perturbative effect. Electrons in these states can be described correctly only by solving the Schrödinger equation.

The Schrödinger equation for the Stark problem in hydrogen can be solved exactly, i.e., with no approximations, by transforming the equations to parabolic coordinates [12]. In the Zeeman problem, no exact solution to the Schrödinger equation has been found in any coordinate system to date. This makes the investigation of the time evolution of the diamagnetic Zeeman system extremely difficult. Thus, a hydrogen atom in an external static electric field can be described completely at all field strengths. The theoretical
description of an atom in a magnetic field, however, is an unsolved problem and different approaches have been adding to the mosaic of information about this system \[13, 14\].

In recent years, it has been possible to probe Rydberg atoms [15], i.e., atoms that are highly excited (to a high principal quantum number \( n \)) experimentally. This is an exciting development because Rydberg atoms are testing grounds for the Correspondence Principle. Introduced by Bohr in 1923, this principle requires that the result of any quantum mechanical calculation reduce to the corresponding classical calculation in the appropriate limit [16], which in this case, is a high principal quantum number \( n \). At these high excitations, the spacing between the discrete energy levels of the atom is small, so that neighboring energy levels are easily mixed. Quantum mechanically, this state of the atom is represented by a wave packet which is a superposition of several states. A Rydberg wave packet can approach the classical ideal of a spatially localized particle travelling along a well defined trajectory. We are interested in studying diamagnetic Zeeman states as well as wave packets so that we can probe the region of transition between classical and quantum behavior.

The study of diamagnetic Rydberg wave packets is interesting also because the corresponding classical problem — the diamagnetic Kepler problem, exhibits chaotic trajectories [17]. Chaos is an area of recent excitement in classical physics where systems obeying classical deterministic equations show extreme sensitivity to initial conditions. That is, a system starting from an infinitesimally different initial condition than a previous one, can end in an
exponentially different final state, even though its evolution is governed by the same equations of motion. The classical equations of motion for an electron in the combined Coulomb-diamagnetic potential show this sensitivity [11]. A Rydberg atom in a high static magnetic field is therefore of immediate interest as a system where the quantum counterpart of the classically chaotic trajectories may be studied.

The first probes used to study atomic systems were vapor and discharge lamps. However, these were of low intensities and could only weakly excite atoms to low lying excited states. To study Rydberg states, the atoms had to be excited to much higher levels. The bandwidth of these lamps was large, i.e., they contained many frequencies and incoherently excited a large number of states making it impossible to study single processes directly. With the invention of the laser, an intense beam of light of a single frequency (monochromatic light) could be used to study many more atomic processes in detail, particularly coherent processes (where the relative phases of the states is important). To study time-varying processes, the probes must be shorter than the changes in the system under study. Therefore, optical (laser) pulses are used to perform time-resolved spectroscopy. Using current technology, ultrashort pulses can be used to study processes that take place in a mere tenth of a picosecond ($10^{-12}$s) or less.

The motivation for our study is the recent development of a new spectroscopic method for studying Rydberg atoms — their ionization using a picosecond Half-Cycle Pulse (HCP) [18, 19]. A HCP is essentially a unipolar electromagnetic pulse whose electric field lies almost completely in one half
of the optical cycle. Ordinarily, an electromagnetic wave cannot absorb or impart momentum since the momentum transferred over a full optical cycle averages to zero. During ionization, the recoil momentum of the ejected electron must be absorbed by some agent, and since the electromagnetic wave cannot, the atomic core must do so. Therefore, in traditional spectroscopic methods like photoionization, an electron is ionized only when it is near the atomic core. The ionization characteristics reflect the state of the atom for a very short time — when the electron is near the atomic core. That is, the ionization occurs only when the electron has a high momentum. Alternatively, in the dipole approximation, the electric field varies only in time and a pulse with zero integral of the electric field can transfer momentum to a bound electron only when the potential is anharmonic, again near the nucleus. A HCP, by contrast, can impart or take away linear momentum and thus ionize an electron anywhere on its trajectory (even when it has a low or zero momentum). HCPs have been used in the study of wave packet dynamics away from the core [20, 21, 22] and also to create wave packets [23, 24]. If the pulse width of the HCP is much smaller than the characteristic time scale of the wave packet (which, for example, is the Kepler orbital period for a radial wave packet), then time-delay spectroscopy can be performed [20, 21]. Recently, this technique has been used to study the ionization of Stark states, showing an asymmetry in the ionization probability depending on the localization of the states that are ionized [25, 26]. Since the diamagnetic system is also characterized by localization of states (parallel and perpendicular to
the magnetic field), ionization by HCP's may be expected to yield rich and interesting results in this system as well.

1.2 Overview

This dissertation documents the theoretical study of the ionization of diamagnetic Zeeman states and wave packets by a half-cycle pulse. Chapter 2 contains a short summary of the properties of atoms in external fields, with the main focus on those properties that are used in the development of the theoretical methods and in the interpretation of the results. These are collated from previously published experimental and theoretical work. The figures published therein are reproduced using our theoretical methods. Chapter /refch3 gives the details of our probe of the system - the ionization by a HCP. The technique of impulsive momentum retrieval[24, 22], where a short HCP is used to obtain information about the momentum distribution of the electron, is described. The coherent control of wave packet generation using HCPs is also discussed.

The theoretical methods used in our study are presented in Chap. 4. We use a pseudopotential method to discretize the Hamiltonian on a non-linear grid and solve the stationary Schrödinger equation. The evolution of non-stationary states in time is studied by a Peacemann-Rachford propagation technique[27]. Using a simple model of the HCP, the electron is ionized both in the short pulse (impulsive) regime and the long pulse regime. The photo-electron spectra are produced by an 'energy window' method [28] which is applied for the first time to a non-separable potential. We apply these techniques to the calculation of the ionization dynamics of hydrogen and sodium.
atoms in a diamagnetic potential. The results are presented in Chap. 5. We show that the ionization of diamagnetic Zeeman states depends strongly on their localization structure. Using a classical model, we explain the suppression of ionization as the width of the HCP gets longer. Using the impulsive momentum retrieval technique, we obtain the momentum distributions of the states of a diamagnetic atom. The effects of the non-hydrogenic core on the HCP ionization profiles are studied in sodium.

The HCP ionization of atoms in a diamagnetic potential enhances our understanding of the diamagnetic Zeeman effect. Our theoretical methods permit not only the accurate calculations of the known static properties, but also a study of the dynamics of these states under the influence of a time-dependent external field. The ionization by a very short HCP yields information about the momentum distribution in these states. Using this theory, we can study the dynamical evolution of an atomic system (the propagation of a wave packet or the time-evolution of an atomic state under the influence of an electromagnetic pulse) as well as predict their ionization behavior. The technique facilitates the study of different alkali atoms and the results are presented in a manner that they can be explored immediately by an experiment. In fact, our results have already prompted plans for experiments which will soon be under way.
2. ATOMS IN EXTERNAL FIELDS

Atoms in an external electric or magnetic field are very interesting because of the rich physical insights that can be obtained by studying them. These systems exhibit competing symmetries - spherical symmetry due to the Coulomb potential and axial or cylindrical symmetry due to the electric or magnetic field respectively. Theoretically, this presents a challenge because the Cartesian, cylindrical or spherical coordinate systems that are normally used depending on the symmetry of the system, cannot be used to solve these problems exactly. These systems also have states which exhibit a strong localization of the electron probability density. This localization, while in itself an interesting feature of perturbations in degenerate systems [29], can also be exploited in the further study of atomic systems in external fields.

We first study the case of an electron in a pure Coulomb potential — field-free hydrogen. This enhances the understanding of the Stark problem which can be completely solved in parabolic coordinates. Then one can compare and contrast with the Zeeman problem, where no such separable coordinate system exists and which is the main focus of this work. The effect of a non-Coulombic core is then seen in the analysis of alkali metal atoms, e.g., sodium, in the same external fields.
2.1 Field-free Hydrogen

The electron in a hydrogen atom lies in a spherically symmetric Coulomb potential due to the positively charged nucleus.

\[ V(r) = -\frac{1}{r}. \]  

(2.1)

It immediately follows that angular momentum and parity are conserved quantities in this system. The Schrödinger equation that describes the stationary states of an electron in this potential is

\[ -\frac{\nabla^2}{2} \Psi + V(r) \Psi = E \Psi. \]  

(2.2)

We use atomic units throughout with \( e = m_e = \hbar = 1 \) (see Appendix A). In spherical coordinates, this equation is separable into its radial and angular components and the radial equation corresponding to an angular momentum \( \ell \) is

\[ -\frac{1}{2r^2} \frac{d}{dr} \left( r^2 \frac{d\psi_{n\ell}}{dr} \right) + V_{\text{eff}}(r) \psi_{n\ell}(r) = E_n \psi_{n\ell}(r), \]  

(2.3)

where the effective potential is

\[ V_{\text{eff}}(r) = -\frac{1}{r} + \frac{\ell(\ell + 1)}{2r^2}. \]  

(2.4)

The eigenvalues \( E_n = -\frac{1}{2n^2} \) and eigenvectors \( \Psi(r) \) which are the products of the radial functions \( \psi_{n\ell}(r) \) and spherical harmonics \( Y_{\ell m} \) have long been known. An excellent pictorial representation of the radial wave functions can be found in the text *Theoretical Atomic Physics* [30]. The spacings between the energy levels of a hydrogen atom vary with the principal quantum number \( n \) as \( \frac{1}{n^3} \). The spacings are larger when the electron is deeply bound.
within the potential and very narrow when the electron has a near-threshold energy. Since the energy eigenvalues depend only on \( n \), they are degenerate with respect to both \( \ell \) and \( m \). For each value of \( n \), \( \ell \) can vary from 0 to \( n - 1 \), and for each value of \( \ell \), \( m \) can vary from \(-\ell\) to \(+\ell\). Degeneracies in energy are associated with conserved quantities and symmetries in a system. The degeneracy in \( m \) is characteristic of a central force field, for which the potential depends only on the radial distance. The \( \ell \) degeneracy is characteristic of the Coulomb field, as distinguished from other central force fields \([16]\). This degeneracy, sometimes referred to as accidental degeneracy in the literature, is associated not with a geometrical symmetry but a dynamical symmetry - represented by the \( O(4) \) group of which the angular momentum vector and the Runge-Lenz vectors are generators \([31]\). In equivalent classical terms, the angular momentum and the Lenz vector are constants of motion for an electron in a Coulomb potential.

2.2 Hydrogen in a Static Electric Field: The Stark Effect

The potential experienced by an atomic electron in a static electric field \( \varepsilon_z \) in the \( z \) direction is

\[
V(r) = -\frac{1}{r} + \varepsilon_z z. \tag{2.5}
\]

The system exhibits competing symmetries — spherical symmetry due to the Coulomb potential and axial symmetry due to the Stark potential. Parity and angular momentum, \( \ell \), are no longer conserved; only the magnetic quantum number, \( m \), is a good quantum number in this system.
Figure 2.1: Stark map of hydrogen. Energy curves of hydrogen in an electric field with increasing field strength corresponding to the \( n=20 \) \( (m=0) \) manifold of field-free hydrogen. The highest energy state (dot-dashed line) is localized uphill the Coulomb-Stark potential and the lowest energy state (dashed line) is localized downhill the Coulomb-Stark potential.
The Stark map in Fig. 2.1 shows the energy eigenvalues of a hydrogen atom in an electric field as a function of increasing field strength. Such a map was first plotted with measurements in 1979 [32]. The energy curves begin at the field free values which correspond to the $n-m$ degenerate $n = 20$ ($m = 0$) states of field-free hydrogen.

The electric field lifts the $\ell$-degeneracy of the states in field-free hydrogen. From the point of view of perturbation theory, the Stark operator $z$ connects the field-free degenerate states of opposite parity and the perturbation is of the first order in the electric field strength. The non-degenerate ground state ($n = 1$) of hydrogen which has even parity, however, has no first order Stark effect. The Stark states are superpositions of the field-free states and therefore have a mixture of angular momenta. At low field strengths, only states from the same $n$-manifold are superposed and this is called the $\ell$-mixing regime. At higher field strengths, states from adjacent $n$-manifolds are also mixed and this is called the $n$-mixing regime. This is indicated by the crossing of the energy curves in the Stark map. In hydrogen, these are true crossings, i.e., the states are truly degenerate at the crossing and they cross each other without interaction. As the states move through a crossing, they are unchanged. This degeneracy corresponds to a conserved quantity [33]; indeed, the $z$-component of the Runge-Lenz vector (besides $L_z$) is a constant of the motion in this system.

The Schrödinger equation representing this system can be solved exactly in parabolic coordinates [12]. The energies of the Stark states calculated to
second order in the field are given by [34, 35]

\[ E_{nn_1n_2m} = -\frac{1}{2n^2} + \frac{3\varepsilon n}{2}(n_1 - n_2) \]
\[ -\frac{\varepsilon^2 n^4}{16}[17n^2 - 3(n_1 - n_2)^2 - 9m^2 + 19], \] (2.6)

where \( n_1 \) and \( n_2 \) are parabolic quantum numbers related to the usual spherical quantum numbers through the relation

\[ n_1 + n_2 + |m| + 1 = n. \] (2.7)

The second order shift can be seen to break the \( m \) degeneracy. For \( m = 0 \), the allowed values of \( n_1 - n_2 \) are \( n - 1, n - 3, \ldots, -n + 1 \). Depending on the sign of \( (n_1 - n_2) \), the energies are downshifted or upshifted from the field-free energy. The red or downshifted “downhill” states are localized along the \(-z\) axis and the higher energy blue or upshifted “uphill” states are localized along the \(+z\) axis. The states for which \( (n_1 - n_2) \simeq 0 \) are localized near the \( z = 0 \) plane. This is inferred from the expression for the electron probability distribution of the Stark states in spherical coordinates [34],

\[ |\Psi_{nn_1n_2m}|^2 = r^{2n-2}(1 + \cos \theta)^{2n_1+|m|}(1 - \cos \theta)^{2n-2+|m|}e^{-2r/n}. \] (2.8)

Pictorial representations of these probability distributions for states with small quantum numbers \( n = 8 \) are well known [36].

### 2.3 Hydrogen in a Static Magnetic Field: The Zeeman Effect

An atom in a strong magnetic field is another example of a system which exhibits competing symmetries (spherical symmetry due to the Coulomb potential and cylindrical symmetry due to the magnetic field). This can be
Figure 2.2: The Coulomb-diamagnetic potential. The competing symmetries due to the Coulomb potential and the magnetic field in the z-direction are illustrated by the 3D plot and the contour plot of the total potential.
seen in the contour and 3D plots of the combined Coulomb-diamagnetic potential in Fig. 2.2.

The Hamiltonian for the H atom in a magnetic field in the $z$ direction \[34\] (one atomic unit of magnetic field = \(2.35 \times 10^5\) Tesla) is

\[
H = \frac{P^2}{2} - \frac{1}{r} A(r) L \cdot S + \frac{L \cdot B}{2} + S \cdot B + \frac{B^2}{8} (x^2 + y^2).
\] (2.9)

The $L \cdot S$ term is negligible except at very low values of the magnetic field; i.e., for the magnetic field strengths that we are interested in, the spin and angular momentum are decoupled and the contribution of spin may be ignored. The paramagnetic terms, which are linear in $B$, add a constant energy to the Hamiltonian, yielding an overall phase factor in the time-dependent wave function, and may also be ignored. This Hamiltonian also conserves parity; thus, for the Coulomb-diamagnetic problem, the magnetic quantum number $m$, as well as parity, are conserved quantities. For each value of $m$, the unperturbed Hamiltonian is $(n - |m|)$-fold degenerate. The degeneracy of the $\ell$-states, $|m| \leq \ell < n$, is then lifted by the diamagnetic potential which is quadratic in the magnetic field. The Schrödinger equation representing the diamagnetic atom is not separable in any co-ordinate system, in contrast to its counterpart, an atom in an electric field, which separates in parabolic co-ordinates.

The diamagnetic map in Fig. 2.3 shows the energy eigenvalues of a hydrogen atom in a magnetic field as a function of increasing field strength. First plotted with measurements in 1980 \[9\], the energy curves begin at the field free values which correspond to the degenerate states of field-free hydrogen. The diamagnetic spectrum is characterized by two regions: low energies and
Figure 2.3: Diamagnetic Zeeman map of hydrogen. Energy curves of hydrogen in a magnetic field with increasing field strength corresponding to the \( n = 20 \ (m = 0) \) even parity manifold of field-free hydrogen. The avoided crossings of these curves with those of the \( n = 21 \) and \( n = 19 \) states are seen. Dashed line: Lowest energy state, Dot-dashed line: Highest energy state, Thick solid line: Separatrix state.
magnetic fields — the $\ell$-mixing regime, where the energy curves of the adjacent $n$-manifolds do not overlap; and higher energies or magnetic fields — the $n$-mixing regime, with significant overlap of adjacent $n$-manifolds. In the $\ell$-mixing regime, the eigenstates are divided into two classes, both showing strong spatial localization [37, 38]. This type of localization is known to be a general feature of perturbed degenerate systems with competing symmetries [29]. The lowest energy state of a single $n$-manifold is localized along the direction of the magnetic field in the potential valley while the highest energy states are localized perpendicular to the magnetic field on a potential ridge. The highest energy "ridge" states are the diamagnetic counterpart of the "uphill" states in the Stark system. States with intermediate energies, the "separatrix" states, are not localized.

The diamagnetic spectrum corresponding to a single $n$-manifold has states of opposite parity which are doubly degenerate at the lowest energy and equally spaced at high energy. The diamagnetic energy term in the Hamiltonian is proportional to the square of the magnetic field and to the fourth power of $n$, the principal quantum number (due to $\langle r^2 \rangle$). In a single $n$-manifold of hydrogen, the diamagnetic energies [29] in the $\ell$-mixing regime lie on a scale of 0 to $\frac{5}{4}$, with the lowest state having energy 0 and the highest energy state having an energy of $\frac{5}{4}$. Between the two, the non-degenerate states which have the least energy spacing are called the separatrix states and they have an energy of $\frac{1}{4}$. Thus the separatrix states are closer to the lowest energy states than to the highest energy states of the same manifold. Both the highest and lowest energy states have predominantly low angular
Figure 2.4: Localization of diamagnetic Zeeman states. Electron probability densities of diamagnetic Zeeman states at a magnetic field of 6T (in the $z$-direction) corresponding to the $n=20$ ($m=0$) even parity manifold of field-free hydrogen. The system is in the $\ell$-mixing regime. The lowest energy states (top) are localized parallel to the magnetic field and the highest energy states (middle) are localized perpendicular to the magnetic field. The separatrix states (bottom) are not localized. The probability distributions are multiplied by $r^2$ to aid visibility.
momentum contributions, $\ell < \sqrt{n}$, and the few separatix states have all the high angular momenta [29] as seen in Fig. 2.5. Previous studies [37] have successfully explained the principal features of the $\ell$-mixing regime, as well as the near-threshold Quasi-Landau region [39].

As the strength of the magnetic field increases, the states from one $n$-manifold begin to interact with the states from adjacent $n$-manifolds. The energy curves of the states in the adjacent $n$-manifolds approach each other and form avoided crossings. This is the $n$-mixing regime. The fact that avoided crossings occur and the two states are not truly degenerate at the avoided crossing indicates the absence of a dynamical symmetry. However, when the width of the avoided crossings is measured, the number of avoided crossings falls off exponentially with increase in the width [9]. This implies the presence of an approximate symmetry which was then shown to be related to the Runge-Lenz vector [40]. Another interpretation of the very narrow avoided crossings is that the localization of the highest energy state of the $n$th manifold and the lowest energy state of the $(n+1)$th manifold are orthogonal to each other. These two states which form the avoided crossing do not have a significant overlap and retain their localization properties at crossing field strengths [38].

At higher energies and magnetic fields, well in the $n$-mixing region, complex structure is seen in the spectrum, some of which can be explained by trajectories in the chaotic region of the classical diamagnetic Kepler problem [41]. These features are due to dynamical effects, and both quantum and
Figure 2.5: Angular momentum distribution in diamagnetic Zeeman states. Distribution of angular momentum, $\ell$, in diamagnetic Zeeman states of hydrogen at a magnetic field of 6T corresponding to the $n=20$ ($m=0$) even parity manifold. The ridge (dot-dashed line) and the valley (solid line) states have mostly low angular momentum contributions ($\ell \leq \sqrt{n}$) and the separatrix states (dashed line) have the high angular momentum contributions.
classical calculations have been done to explain them. Classically, chaotic orbits correspond to orbits in that energy range which do not close (soft chaos) [11]. One method of representing the evolution of classical systems is the Poincaré surface of section. Starting from an initial condition, the trajectory of a particle evolving according to classical equations of motion fills three dimensional space. Choosing a plane in this space, the evolution of the system in time is marked by the appearance of a point each time the trajectory intersects this plane. If the orbit is closed (with period 1), then the point is the same for each traversal. For stable orbits, the trajectories of the particle starting from points close to that orbit approach the stable orbit as the system evolves to an infinite time. This makes a regular pattern on the section. If an orbit is chaotic, the trajectories of the particles starting from points adjacent to the orbit diverge exponentially and rapidly fill up space. These appear as dots in Poincaré section maps.

Another method of looking at chaos in the classical system is to find eigen trajectories of the system, i.e., trajectories corresponding to the quantized integral of the action, and see if they are periodic [17]. In experimental spectra, one looks for structure that has a periodicity by looking at the Fourier transform of the spectrum. The absence of periodicity is an indicator of the onset of chaos [41]. In quantum mechanical studies, statistical distributions of energy level spacings in hydrogen show a change in nearest neighbor spacings from Poisson to Wigner, corresponding roughly to the change in the classical dynamics from quasi-integrable (at low energies and magnetic fields) to chaotic (at high energies or magnetic fields) [42]. This can be understood
by invoking the arguments of approximate symmetry. In the quasi-integrable region, the avoided crossings are narrow and the statistical distribution of the energy level spacings peaks at zero as in the Poisson distribution. As the system moves away from quasi-integrability, the approximate symmetry breaks down and the avoided crossings are wider. Thus the distribution of the energy level spacings peaks at a non-zero value. This method of studying the diamagnetic system is popular because it is more economical to compute the eigenvalues of a system than the eigenvectors. Another method used is to artificially put a gaussian wave function in a potential and compute its propagation in the potential [43]. However, these methods do not give any information about the stationary state wave functions or the electron probability distributions and cannot show their dynamical evolution. The power of our method lies in the ability to calculate the initial state wave functions, propagate them in the presence of external fields, and use them to calculate experimentally measurable quantities such as the ionization probabilities.

2.4 Alkali-metal Atoms

Alkali-metal atoms are similar to hydrogen since they have one valence electron. The behavior of the outer electron may be understood as a single electron moving in the combined potential of the nucleus and the inner shell electrons, i.e., the core. This combined potential is central but only approximately of the Coulomb form. This prevents the states with the same quantum number \( n \) from having the same energy (as in the Coulomb degeneracy in field-free hydrogen). The energy levels of field-free alkali metals can
be calculated from a relation that is very similar to that of hydrogen,

\[ E_{nl} = \frac{-1}{2(n - \mu \ell)^2}, \]  

(2.10)

where \( \mu \ell \) is called the quantum defect. This arises due to the core penetration of the wave function where the potential is not Coulombic. The radial wave functions with smaller \( \ell \) values penetrate the core while those with \( \ell > 2 \) hardly penetrate the core at all because of the large angular momentum barrier. Thus, the quantum defect depends on the \( \ell \) quantum number, being large for the \( \ell = 0 \) or \( s \) states and small for states with \( \ell > 2 \). For example, in sodium, the states with \( s \) and \( p \) character are separated by a large difference in energy from the corresponding field-free value in hydrogen. The state with \( d \) character is much less separated from the other states and the rest of the states with quantum numbers \( n, 2 < \ell < n - 1 \) have hydrogen-like energies.

On the application of an external electric or magnetic field, the widely separated states of \( s \) and \( p \) character do not mix with the rest of the manifold at low field strengths. The localization characteristics of the rest of the hydrogenlike states of the same manifold stay the same as in hydrogen. The presence of the quantum defect causes anticrossings in the adiabatic Stark map, i.e., the dynamical symmetry due to the Coulomb potential no longer exists. The angular momentum distribution is also similar to that in hydrogen with the exception of the \( s \)-wave contributions. The diamagnetic interaction is proportional to the square of the radial distance and is appreciable at large radial distances. The quantum defect is large for low \( \ell \) contributions which are significant near the atomic core. Therefore, it can be expected that a phenomenon that depends on the localization properties
of the wave functions will show a behavior similar to that in hydrogen. However, dynamical phenomena which do involve core interaction, such as the time-evolution of a radial wave packet which interacts with the nucleus on the time scale of a Kepler orbital period, are expected to show differences in alkali metals as compared with hydrogen.
3. HALF-CYCLE PULSES

Ionization by half-cycle pulses (HCPs) is a recently developed spectroscopic method of studying Rydberg atoms [18, 19]. An HCP is essentially a unipolar electromagnetic pulse whose electric field lies almost completely in one half of the optical cycle. Fig. 3.1(a) shows a numerical fit to a measured half-cycle pulse field [26]. The pulse consists of a large lobe of short duration, followed by a long but shallow tail of the opposite polarity. At large times, this tail is too weak to ionize an electron. Moreover, in the laboratory experiment, the HCP is a focussed pulse and the electron will leave the focal region before it experiences the negative tail. Therefore an HCP is effectively a unipolar pulse and is represented as such in our model, shown in Fig. 3.1(b). We emphasize that an HCP is neither a single photon nor an electromagnetic wave; but a pulse with a unipolar shape.

3.1 Properties of a Half-cycle Pulse

The characteristic that sets HCP ionization apart from traditional spectroscopy is that an HCP can ionize an atomic electron all along its trajectory. It is well known that a single photon or pure electromagnetic wave cannot impart momentum to a free particle [44]. Therefore, in traditional spectroscopic methods such as photoionization, an electron can be ionized only when it is near the atomic core so that the core may absorb the recoil momentum during ionization. This means that the ionization reflects the state of the atom in a very short time — while the electron is near the nucleus and has a
Figure 3.1: A half-cycle pulse. (a) Pulse shape fitted to an experimental HCP field as in Ref.[26]. (b) Our model of an HCP with the same peak HCP field and full width at half maximum (FWHM) as in (a).
high momentum. An HCP, by contrast, can absorb and impart momentum and can ionize an electron anywhere on its trajectory.

An HCP can also be used to create wave packets. When an HCP interacts with an atomic system, its electric field mixes the atomic states to produce a wave packet. The coherent bandwidth of a subpicosecond HCP is large enough that many states of different angular momentum and principal quantum numbers are coupled in a single pulse. This method has been used to create novel dynamic states in atoms [23, 24] which is a key to the coherent control of atoms and molecules. An HCP can also be used to probe the temporal evolution of wave packets. If the width of the HCP is much smaller than the characteristic time scale in a system (for example, the Kepler orbital period in a radial wave packet), then the dynamical evolution of the system may be studied by time-resolved spectroscopy [20, 21]. Such an HCP, whose duration is much shorter than typical time scales in an atomic system, can be used along with a classical impulse approximation or sudden impact approximation to retrieve the momentum probability distribution (in the direction of the HCP’s electric field) of the system [22, 24].

These properties of an HCP have prompted several investigations other than those cited above. The dynamical stabilization of atoms by periodically kicking the atom with a series of HCPs has been demonstrated recently [45]. A theoretical study of the HCP ionization of elliptic states [46] in atoms in crossed, weak electric and magnetic fields is also interesting, as in the limit of setting one of the fields to zero, this reduces to the extreme Stark states on one hand (magnetic field \( B = 0 \)) and into circular states (static electric
field $\varepsilon_s = 0$) on the other. The same properties have been exploited in the ionization of atoms using quarter-cycle pulses (QCPs) [47].

3.2 Production of a Half-cycle Pulse

An HCP is produced using a thin GaAs semiconductor wafer of 0.5mm thickness and $\simeq 3\text{cm}^2$ surface area[48]. An electric field of strength less than 10KV/cm is applied parallel to the surface of the wafer. (This bias field is a typical value. HCP's have been produced by the application of bias fields up to 100KV/cm, but their characterization has not been reliable.) This biased semiconductor acts as a photoconducting switch. The bias field is shorted across the semiconducting surface by illuminating one side of the wafer with a 100fs, 770nm laser pulse from a Ti:sapphire oscillator, which drives the GaAs into conduction. The electrons rapidly accelerating through the photoconductor radiate energy. A significant portion of this energy (up to 80%) is transmitted through the wafer in the form of a spatially coherent, $\simeq 0.5\text{ps}$, single-polarity, HCP. The HCP is polarized in the direction of the bias field in the wafer and its peak electric field is proportional to the bias field. The HCP has a large bandwidth of the order of 1 THz. The bandwidth of the HCP is determined by using an electric field autocorrelator and the temporal pulse shapes are inferred by using cross correlation techniques, and a good description of this technique is found in Refs. [48, 49].

The HCP that is produced has a strong, short unipolar component and a very long weak tail of the opposite polarity. In recent experiments, this weak tail has been cut off by using a photoswitch as described in Ref. [50]. The characterization of a spatially propagating HCP [51] shows that a focussed
HCP changes its shape as it propagates. The unipolar character of an HCP is restricted to a small spatial location which depends on the focal length of the lens used. The interaction of the atoms with the main (unipolar) lobe of the HCP is ensured by a narrow extraction slit.

### 3.3 Theoretical Model of a Half-cycle Pulse

The HCP used to ionize an electron is modeled as a time-dependent electric field. Our model uses the form in the equation below, however, several other shapes like a square or gaussian pulse have been used in earlier theories[52]. The shape of the HCP does not substantially affect the physics described in this section.

\[
E(t) = \begin{cases} 
\hat{z}E_{HCP} \sin\left(\frac{2\pi}{t_{pulse}} t\right) & 0 \leq t \leq t_{pulse}, \\
0 & t > t_{pulse}, 
\end{cases}
\]

where \(E_{HCP}\) is the maximum field amplitude in atomic units and \(t_{pulse}\) is 1.5 times the full width at half maximum (FWHM) of the electric field. The momentum transferred by such a pulse to a free electron in the absence of any other interactions is

\[
Q = -\int_0^{t_{pulse}} dt \, E(t).
\]

In the limit that the pulse width is very small, no other interactions take place during the pulse, and the effect of the HCP is to boost the electron by a momentum \(Q\)

\[
\Psi_f(r, t) = e^{iQ \cdot r} \Psi_i(r, t).
\]

This is the impulse approximation of an HCP.
3.4 Impulsive Momentum Retrieval

An accurate measurement of wave functions in any system is extremely difficult because the information needed for the complete representation of a state is both its probability amplitude and phase. Even with the knowledge of the spatial probability distribution $|\Psi(r)|^2$, one cannot find the momentum probability distribution $|\Psi(p)|^2$ directly. By using an HCP in the impulsive limit, it is possible to retrieve new information about the electron probability distribution in momentum space [22, 24].

For a classical electron of momentum $p_i$, the final momentum after its interaction with an HCP is $p_f = p_i + Q$, where $Q$ is the momentum transferred by the HCP. If the electron does not move during the HCP, then the change in its energy is equal to

$$\Delta E = \frac{p_f^2}{2} - \frac{p_i^2}{2} = p_i \cdot Q + \frac{Q^2}{2}. \quad (3.4)$$

To ionize an electron, this change in energy must exceed the binding energy $|E_b|$. Since the electric field of the HCP is in the $z$-direction, the initial $z$-momentum of an electron that will have threshold energy after the pulse is given by

$$p_{0z} = (|E_b| - \frac{Q^2}{2})/Q. \quad (3.5)$$

The probability that an electron is ionized in this classical impulse approximation is just the probability that its initial momentum in the $z$-direction is greater than $p_{0z}$ [24]. That is, the fraction of electrons ionized by an HCP
which transfers a momentum \( Q \) is

\[
I(Q) = - \int_{-\infty}^{+\infty} dp_x \int_{-\infty}^{+\infty} dp_y \int_{p_0}^{\infty} dp_z |\Psi(p)|^2
\]

\[
= \frac{|E_b| + Q^2/2}{Q^2} |\tilde{\Psi}(p_z)|^2, \tag{3.6}
\]

where \( |\tilde{\Psi}(p_z)|^2 \) is equal to \( |\Psi(p)|^2 \) integrated over \( p_x \) and \( p_y \). \( I(Q) \) can now be inverted to give

\[
|\tilde{\Psi}(p_z)|^2 = \frac{Q^2}{|E_b| + Q^2/2} \frac{dI}{dQ}. \tag{3.7}
\]

Making a graph of the ionization as a function of the impulsive momentum transferred, the slope of the curve directly yields information about the momentum distribution of the initial state.

### 3.5 HCP Ionization of Stark States

The HCP ionization of sodium Rydberg states in the presence of an additional static electric field shows an asymmetry in the ionization dynamics, which depends on the localization of the electron probability distributions in the state - uphill and downhill in the Stark potential (described in Chap. 2) [25]. This asymmetry is explained by invoking a classical argument. The energy transferred by the HCP to the ionized electron is

\[
\Delta E = - \int_{t_{\text{pulse}}}^{t} dt \mathbf{E}_{\text{HCP}}(t) \cdot \mathbf{v}(t); \tag{3.9}
\]

where \( \mathbf{v}(t) \) is the instantaneous velocity of the electron. Whenever the velocity of the electron is not aligned with the direction of the HCP field, it results in a smaller gain in energy. In the vicinity of the nucleus, the velocity of the electron changes direction. Therefore, the electron gains more net
energy if the HCP were to impart momentum to the electron in such a way that it does not interact with the nucleus or atomic core. The experimental results show good qualitative agreement with theoretical calculations [26]. Since the diamagnetic Zeeman states are also localized, their ionization by a HCP promises interesting results as well.

3.6 Atomic Wave Packets

An atomic wave packet is a coherent superposition of stationary states of an atom. The large coherent bandwidth of an HCP makes it possible to superpose a large number of states, thereby creating a wave packet. In ordinary spectroscopy, individual stationary or quasi-stationary states are excited and studied. However, stationary states are inherently quantum objects, even for extremely large quantum numbers, so that a single state cannot describe a classical electron in a Kepler orbit. An HCP can produce a three-dimensionally localized wave packet [53] which approaches the classical picture of an electron orbiting the atomic core. By changing the HCP field strength, we can kick the electron from an initial atomic state making either a bound wave packet (for a soft kick) or a continuum wave packet (for a hard kick). For a general review of atomic wave packets, see Ref.[54]. We describe some properties of wave packets and use these concepts in the analysis of the HCP ionization of diamagnetic Zeeman states.

3.6.1 Properties of a Wave Packet

3.6.1.1 Localization

The most obvious property of a wave packet is its localization. A localized wave packet obeying quasi-classical laws of dynamics is composed of...
a large number of stationary states, with the main contribution from their short wavelength (or high momentum) components. Since a wave packet is produced by the optical (laser) excitation of an atom in a low lying state to a superposition of a large number of Rydberg states, it follows that the bandwidth of the exciting laser pulse must be very large; i.e., the temporal width of the pulse must be very small. For example, to produce a "classical limit" atomic state, a picosecond pulse is used (which has a bandwidth of the order of a THz) [55].

Consider an atomic wave packet formed with states labeled $n$ and let the central frequency of the exciting laser pulse correspond to a state $\bar{n}$. Assume that the wave packet is strongly localized in space, i.e., its spatial extent $\Delta x$ is much smaller than $L$, the characteristic length associated with the classical orbit [56]. From the uncertainty principle, the energy of the wave packet is,

$$\Delta E \propto v \Delta p \propto \hbar \omega_{cl} \frac{L}{\Delta x},$$

where $v$ and $\Delta p$ are the characteristic values of the velocity and the uncertainty in the momentum respectively, and $\omega_{cl}$ is the frequency spacing between the adjacent $n$ states. This means the "width" of the number of states excited

$$\Delta n \propto \frac{\Delta E}{\hbar \omega_{cl}} \propto \frac{L}{\Delta x} \gg 1.$$  

Thus, a strongly localized, particle-like wave packet must have a large number of stationary states. The larger the number of states superposed, the greater the localization [56].
The superposition of many states of low angular momentum but of several different manifolds (with different principal quantum numbers) excites a wave packet that is localized radially \cite{57}. A radial wave packet exhibits oscillatory motion in the radial direction at the classical oscillation period $2\pi \bar{n}^3$ \cite{58}. A wave packet that is localized in the polar and azimuthal angles can be produced by the superposition of states in the same manifold (which includes higher angular momenta) \cite{57, 59}. A wave packet that is localized in all three dimensions has been observed very recently \cite{53}.

### 3.6.1.2 Dephasing and Revival

In an atom, the energy levels are only quasi-equidistant at high energies. The classical frequency $\omega_{cl}$ is therefore a weak function of the energy itself. The classical period $T_{cl}$ is controlled by the smallest energy spacing among the various components of the wave packet.

\[
T_{cl}(E) = \frac{2\pi}{\omega_{cl}} \quad (3.12)
\]

\[
= 2\pi \bar{n}^3. \quad (3.13)
\]

Taking into consideration the slight differences in the energy level spacing at high $\bar{n}$, the energy in the region can be written as \cite{56}

\[
E \approx E_n + 2\pi \hbar \frac{(n - \bar{n})}{T_{cl}} \pm 2\pi \hbar \frac{(n - \bar{n})^2}{T_{rev}} + \ldots, \quad (3.14)
\]

where $\pm$ refers to the sign of $\partial \omega_{cl}/\partial E$ and

\[
T_{rev} = \frac{2T_{cl}}{\hbar |(\partial \omega_{cl}/\partial E)|}. \quad (3.15)
\]

For an atom in a weak static electric field, the Stark energies are equidistant to first order in the field. But for an atom in a strong electric field or a
strong magnetic field, the energy spacings are not equidistant and the term with $T_{rev}$ is important.

The wave function of the packet may be written as

$$\psi(x,t) \approx \sum_k C_k(t) \psi_k(x) \exp \left( -i \left( \frac{2\pi k}{T_{cl}} + \frac{2\pi k^2}{T_{rev}} \right) \right); \quad (3.16)$$

where $k = n - \bar{n}$. The evolution of the wave packet may be understood as follows. For times $t < T_{cl}$, the wave packet merely undergoes dispersion, i.e., the initially localized wave packet moves over a classical trajectory and spreads out. After time $t \approx T_{cl}$, the wave packet regains its shape because of the quasi-equidistant energies of the component states. Therefore, after a time equal to the classical oscillation period, the wave packet, which has moved along a classical trajectory, returns to its original position and its original shape. In this sense, the packet evolves according to the Correspondence Principle. For times greater than the classical oscillation period $T_{cl}$, the individual components of the wave packet begin to dephase. This leads to the decay of the wave packet after many periods of classical motion. However, at time $t = T_{rev}$, the additional dephasing phases are exact multiples of $2\pi$, so that the classical evolution of the wave packet is restored. This is called the 'revival' of the wave packet [60]. A classical state of an atom has also been produced where the features of classical evolution, dephasing and revival are clearly seen [55].

3.6.2 Coherent Control

A wave packet is produced when an electron in a tightly bound state is rapidly excited to a superposition of Rydberg states producing a localized distribution of probability density [54]. The spatial extent of the wave packet
is of the order of the classical Bohr radius of the initial state and is therefore much smaller than the characteristic lengths associated with the Rydberg states. From the discussion in the previous sections, it is seen that the properties of a wave packet depend upon the initial distribution of the wave packet, the central energy of the packet, the energy distribution and, most importantly, the phase of the constituent states. Another way of stating the same is that the wave packet depends on the shape, central frequency, bandwidth and coherence of the exciting pulse. The coherence of the pulse is transferred to the wave packet during excitation. Therefore, the properties of the wave packet may be controlled by appropriately tailoring the coherence properties of the exciting pulse. For example, the frequency of the pulse may be chirped (changed linearly over time), or two pulses may be used to generate wave packets at two different times.

Such techniques have been used to produce novel atomic states like the Schrödinger cat states [61], a classical state of the atom [55], etc., and the production of Trojan wave packets (non-dispersive wave packets that orbit at a large distance from the nucleus) [62] has been proposed. Quantum control may also be used to control the multi-dimensional dynamics of electrons in atoms; indeed, the production of a three-dimensional wave packet [53] is an example of this. With appropriate pulses, the optical ionization of atoms can be stabilized [63], or populations in Rydberg states can be redistributed, and a state can be driven through multiple excitation routes to the same final state [64]. Using these techniques, the equivalent of Young's double slit experiment has been demonstrated in an atom [65]. Recently, a
method to control the THz emission from Stark wave packets by controlling the parameters of the excitation pulse has been proposed [66, 71]. Coherent control is also used in chemistry to dissociate selectively chemical bonds in large molecules [67]. Using shaped pulses, it is possible to create specifically desired target states ("sculpting a wave packet", to borrow a phrase) [68].

Wave packets are created, on one hand, to realize localized probability distributions that approach the classical ideal of a particle-like electron moving in an orbit around the nucleus. These are used to probe the region of transition between quantum and classical behavior. On the other hand, wave packets with specifically shaped probability distributions are created to produce novel atomic states. These can be produced by specifically tailored HCPs, by using a HCP along with a laser pulse, or by using a series of HCPs. By changing the coherence of the exciting pulse, in our case - an HCP, we can control the nature of the wave packets generated.
4. THEORETICAL METHODS

In this chapter, we describe the theoretical methods used in our study of the HCP ionization of atoms in external fields. The Schrödinger equation for an electron in a field-free atomic potential is solved using a grid based pseudopotential method. The matrix elements of the external electric or magnetic potential are constructed in a restricted basis of the field-free states and the Stark or diamagnetic Zeeman states are obtained. These states may be studied individually or a wave packet may be formed and studied. A wave packet is formed by the laser excitation of a deeply bound electron to a superposition of excited states. An individual state or a wave packet is then probed via ionization by a half-cycle pulse. We show the methods of time-propagation of the states evolving in the field of the HCP both in the short pulse (impulse approximation) regime and otherwise. The energy window technique of determining the photoelectron spectra and the ionized population of states is also demonstrated. The results of the study using these theoretical methods are presented in the following chapter.

4.1 Construction of Eigenstates of the Field-free Atom

We begin by finding the eigenstates of the unperturbed atomic Hamiltonian. The Schrödinger equation for an electron in an unperturbed atom is

\[ H_0 \Psi(r) = \left( -\frac{\nabla^2}{2} + V(r) \right) \Psi(r) = E \Psi(r). \]  

(4.1)

In hydrogen, the potential \( V(r) \) is Coulombic, i.e., \( V(r) = -1/r \) and the Hamiltonian is separable in spherical co-ordinates. For alkali metal atoms,
we use a non-local pseudo-potential \([69]\):

\[
V(r) = \sum_{\ell} V_\ell(r)|\ell\rangle\langle\ell| + V_{pol}(r) - \frac{1}{r},
\]

where \(V_\ell(r)\) is an \(\ell\)-dependent short range contribution and \(V_{pol}(r)\) is a polarization term. Both the short range and the polarization terms are strongly dependent on atomic structure. The short range term represents the shielding of the nuclear charge by the core electrons, as well as the orthogonality constraints imposed by the exclusion principle. The polarization of the ion charge cloud is accounted for through dipole and quadrupole potentials.

The solution is found by expanding the wave function in a mixed basis of discretized radial functions times spherical harmonics, while retaining a finite number of spherical harmonics:

\[
\Psi(r_j, \theta, \phi) = \sum_{\ell=0}^{\ell_{max}} \Phi_\ell(r_j) Y_\ell^m(\theta, \phi),
\]

where \(j\) is an index corresponding to a radial grid point. The resulting equation for \(\Phi_\ell(r_j)\) is then discretized (See Appendix B). The use of second order approximations in the discretization yields an eigenvalue equation of a symmetric tridiagonal matrix, which is then solved.

Following the method in Ref.[71], the numerical solution is found on a non-uniform radial grid (See Appendix C). The grid is closely spaced near the nucleus, with the spacing increasing to a constant value towards the outer edge. This makes it possible to represent Rydberg wave functions accurately. Both positive and negative energy states are represented on the same grid, thus allowing us to study the dynamics of bound states as well as
ionization problems. The unperturbed eigenstates, which we label as $\Phi_{n\ell}(r_j)$, are eigenstates of a real, symmetric, tridiagonal matrix.

To find the field-free basis states, we diagonalize a symmetric, tridiagonal matrix for each angular momentum $\ell$ value. A diagonalization can yield $N$ eigenstates and eigenvalues (where $N$ is the number of grid points) of which we require only the lowest few. The complexity of this procedure is of $O(N)$. The grid is chosen to yield eigenvalues with a maximum error of 0.01% by comparing them to known eigenvalues of the hydrogen atom. The radial functions are also in excellent agreement with the analytic solutions to the radial part of the Schrödinger equation for values of the principal quantum number up to $n = 35$.

4.2 Matrix Elements of the Perturbing Potential

An external electric or magnetic field perturbs the atomic states and mixes the field-free wave functions. We now find the stationary state wave functions and energy eigenvalues of an atom in an external field. The matrix elements of the perturbing potential corresponding to the applied external field are found in the unperturbed basis. We choose a restricted basis with principal quantum numbers ranging from $n_{\text{min}}$ to $n_{\text{max}}$. Throughout, we work with states with magnetic quantum number $m = 0$; therefore the angular momentum quantum number, $\ell$, ranges between 0 and $n_{\text{max}} - 1$.

4.2.1 Matrix Elements of the Stark Potential

The potential due to an applied electric field $\varepsilon_s$ in the $z$-direction is

$$V(r) = \varepsilon_s z = \varepsilon_s r \cos \theta.$$  \hfill (4.4)
The matrix elements of the Stark potential are written as

\[ \langle V \rangle = \langle \Psi_{n'\ell'}(r) | \varepsilon_s r \cos \theta | \Psi_{n\ell} \rangle . \]  

(4.5)

\[ = \varepsilon_s \sum_{n\ell n'\ell'} \langle \Phi_{n'\ell'} | r | \Phi_{n\ell} \rangle \langle \ell' | \cos \theta | \ell \rangle . \]  

(4.6)

The matrix elements of \( \cos \theta \) are non-zero only for values of \( \ell' = \ell \pm 1 \) and are [72]

\[ \langle \ell + 1 | \cos \theta | \ell \rangle = \frac{\ell + 1}{\sqrt{(2\ell + 1)(2\ell + 3)}} . \]  

(4.7)

The electric field connects the angular momentum components of the opposite parity, thereby breaking the reflection symmetry of the unperturbed atomic Hamiltonian.

4.2.2 Matrix Elements of the Diamagnetic Potential

The electron of an atom in an external magnetic field \( \mathbf{B} \) in the \( z \)-direction is under the influence of a combined Coulomb-diamagnetic potential. We construct the matrix elements of the diamagnetic potential

\[ V_B = \frac{B^2}{8}(x^2 + y^2) , \]  

(4.8)

in a restricted unperturbed basis \( \Phi_{n\ell} \) with \( n \) ranging from \( n_{\text{min}} \) to \( n_{\text{max}} \). In spherical coordinates,

\[ V_B(r) = \frac{B^2}{8}r^2 \sin^2 \theta . \]  

(4.9)

The only non-zero matrix elements are those which connect states of angular momentum \( \ell \) with those of \( \ell \) or \( \ell \pm 2 \). The matrix elements are [72, 73]

\[ \langle n'\ell' | \sin^2 \theta | n\ell \rangle = 2 \frac{\ell^2 + \ell - 1}{(2\ell - 1)(2\ell + 3)} \delta_{n'\ell'} \]  

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\[-\frac{(\ell + 2)(\ell + 1)}{(2\ell + 3)\sqrt{(2\ell + 5)(2\ell + 1)}} \delta_{\ell,\ell+2}
\]
\[-\frac{\ell(\ell - 1)}{(2\ell - 1)\sqrt{(2\ell + 1)(2\ell - 3)}} \delta_{\ell,\ell-2}. \tag{4.10}\]

Since the total potential conserves parity, we can find odd and even parity states (which contain odd or even \(\ell\) contributions) independently.

The radial part of the matrix elements are computed numerically. By comparing them with the analytical expressions which are known for hydrogen [74], we ensure their accuracy. The basis states are grouped by the angular momentum quantum number as \(|n_{\text{min}},\ell_{\text{min}}\rangle, |n_{\text{min}}+1,\ell_{\text{min}}\rangle, \cdots, |n_{\text{max}},\ell_{\text{min}}\rangle, |n_{\text{min}},\ell_{\text{min}}+1\rangle, \cdots, |n_{\text{max}},\ell_{\text{max}}\rangle, \cdots, |n_{\text{max}},\ell_{\text{max}}\rangle\). For example, the structure of the diamagnetic potential matrix \(\langle n_1 \ell_1 | V_B | n_2 \ell_2 \rangle\) in a restricted basis of states with \(n\) between 1 and 5 is seen in the matrix below, where each entry \(V_{ij}\) contains the elements with all the relevant \(n\) values.

\[
\langle V_B \rangle = \begin{pmatrix}
V_{00} & 0 & V_{02} & 0 & 0 \\
0 & V_{11} & 0 & V_{13} & 0 \\
V_{20} & 0 & V_{22} & 0 & V_{24} \\
0 & V_{31} & 0 & V_{33} & 0 \\
0 & 0 & V_{42} & 0 & V_{44}
\end{pmatrix} \tag{4.11}
\]

The matrix elements form a symmetric, banded matrix which can then be diagonalized using standard algorithms.

**4.3 Eigenstates of the Full Hamiltonian**

The diagonalization of the Stark or diamagnetic Hamiltonian reduces to finding eigenvalues \(E^k\) and eigenvectors \(a_i^k\) of a real, symmetric, banded, matrix. We vary the number of basis states to check the convergence of the energy eigenvalues. Typically, we use a basis of 220 states around \(n \simeq 20\).
The eigenstates

\[ |k\rangle = \sum_{l} a^k_l(r)|l\rangle \]  \hspace{1cm} (4.12)

are again products of radial and angular functions; therefore, the dynamics and the symmetry of the problem may be separated easily. The eigenvalues are in excellent agreement with those produced by large numerical calculations (using a basis of over 40,000 states) [70] over the range of values for the magnetic field that we are interested in. Our calculations of the localization of the probability distributions (Fig. 2.4) and \( \ell \) contributions (Fig. 2.5) of the eigenstates are in excellent agreement with previously published quantal calculations [29].

The eigenstates are labeled by \( k \) which ranges from \(-(n-1), -(n-1) + 2, \) to \( +(n-1) \) in the Stark case and from 1 to \( \frac{n}{2} \) in the even parity diamagnetic Zeeman case. Taking the \( n = 20 \) Stark manifold as an example, the downhill or lowest energy state is labeled \( k = -19 \) and the uphill or highest energy state is labeled \( k = +19 \). Similarly, in the \( n = 20 \) diamagnetic Zeeman manifold of even parity, the highest energy or ridge state (described in Chap. 2) is labeled \( k = 10 \), the lowest energy or valley state is labeled \( k = 1 \) and the separatrix state is labeled \( k = 4 \). In the case of the Stark states, the label \( k \) is equal to the difference \( n_1 - n_2 \) between the parabolic quantum numbers, but in the case of the diamagnetic states, \( k \) is just a labelling index.

The time evolution of individual eigenstates can be performed by multiplication with the appropriate phase.

\[ |k(t)\rangle = e^{-iE_k t}|k(0)\rangle. \]  \hspace{1cm} (4.13)
The excitation and ionization properties of the eigenstates \( |k\rangle \) of the full Hamiltonian may be studied individually by examining the time-dependent dynamics when starting in a single eigenstate. It is also possible to make a coherent wave packet by promoting an electron initially in a deeply bound state (that is not affected by the external magnetic field) with a laser pulse to a superposition of higher perturbed (Stark or diamagnetic) states.

### 4.4 Ionization by a Half-cycle Pulse

The HCP used to ionize the electron is modeled as a time-dependent electric field of the form:

\[
E(t) = \begin{cases} 
\hat{a} E_{HCP} \sin\left(\frac{2\pi t}{2\tau_{pulse}}\right) & 0 \leq t \leq \tau_{pulse}, \\
0 & t > \tau_{pulse}, 
\end{cases}
\]

where \( E_{HCP} \) is the maximum field amplitude in atomic units and \( \tau_{pulse} \) is 1.5 times the full width at half maximum (FWHM) of the electric field.

The time evolution of a state in the combined Coulomb-diamagnetic potential under the influence of a HCP is treated by the Peacemann-Rachford method [27]. In the total Hamiltonian \( H = H_0 + H_I \), \( H_0 \) connects adjacent radial points of functions with the same \( \ell \) value, whereas the interaction \( H_I = H_B + H_{HCP} \) couples functions of different \( \ell \) values (\( \ell \to \ell, \ell \pm 1, \ell \pm 2 \)) at the same radial point.

\[
\Psi(r,t+\delta t) = \sum_{n=0}^{\infty} \frac{(-i)^n}{n!} \int_0^t dt_1 \int_0^t dt_2 \ldots \int_0^t dt_n 
T[H(t_1) H(t_2) \ldots H(t_n)]\Psi(r,t),
\]

where, \( T \) represents the time-ordering operator. To second order, the short time propagator is

\[
\Psi(r,t+\delta t) = e^{-iH(t+\frac{\delta t}{2})\delta t}\Psi(r,t).
\]
We expand the propagator as
\[
\Psi(r, t + \delta t) = [1 + iH_0 \frac{\delta t}{2}]^{-1}[1 + iH_f \frac{\delta t}{2}]^{-1}
[1 - iH_f \frac{\delta t}{2}][1 - iH_0 \frac{\delta t}{2}]\Psi(r, t).
\] (4.17)

The Peacemann-Rachford propagator agrees with the full propagator up to the third order in $\delta t$. The first two operations on $\Psi(r, t)$ are straightforward, the next two require finding the solution to five-term and three-term recurrence relations in $\ell$ and $r$, respectively. The computational complexity of this operation is of first order in $N_r \times N_\ell$. The HCP field exerts a torque on the electron and consequently many higher $\ell$ values are mixed in during the time evolution. This effect is also observed by monitoring the average angular momentum $\ell$ as a function of time. This average increases during the HCP field. Although our initial state may have only low angular momentum components, we use up to $\ell \simeq 200$ during the time evolution of a state under the influence of a HCP in the combined Coulomb-diamagnetic potential. Since the commutator $[H_0, H_f] \neq 0$, the Peacemann-Rachford propagator is not unitary. Therefore, the time step $\delta t$ must be small enough to insure that the higher-order terms in the expansion of the exponential in Eq. 4.16 are negligible. We monitor the norm of the wave function at every time step as a check on this property. We can perform 120,000 such space-time steps in 1 CPU second on a DEC-alpha workstation.

4.4.1 The Impulse Approximation

If the width of the HCP is much smaller than typical time scales in the system, a semi-classical approximation can be successfully used to obtain the
ionization characteristics. Eq. 4.16 may be written as

$$\Psi(r, t + \delta t) = e^{-i(H_I + H_0)\delta t}\Psi(r, t).$$  \hspace{1cm} (4.18)

$$= e^{-iE(t)z\delta t}\sum_k C_k(t_p)e^{-iE_k\delta t}|k\rangle.$$  \hspace{1cm} (4.19)

$$\Psi(r, r_p) \approx e^{-i\int_0^{r_p} E(t)z\delta t}\sum_k C_k(t_p)e^{-iE_kr_p}|k\rangle.$$  \hspace{1cm} (4.20)

As $\frac{r_p}{\tau_{Keppler}} \ll 1$,

$$\Psi_{\text{impulse}}(r) = e^{iQz}\Psi(r).$$  \hspace{1cm} (4.21)

In the impulse approximation, the HCP essentially boosts the wave packet with the total momentum $Q$ in the direction of the electric field of the HCP. In the $r - \ell$ basis, the matrix elements of $\cos \theta$ are found as in the case of the Stark matrix elements. Since the HCP field couples angular momenta, we use a basis with angular momentum $\ell$ up to 200. The eigenvalues of this matrix range between -1 and 1 and the matrix $S$ which is used to make a similarity transformation between the $r - \ell$ basis and the diagonal basis of $\cos \theta$ is found. The matrix

$$e^{iQz} = e^{iQr \cos \theta}$$

$$= S^{-1}e^{iQr S \cos \theta S^{-1}} S.$$  \hspace{1cm} (4.22)

The application of the above operator to the initial wave function $\Psi(r)$ boosts its momentum by $Q$ in the $z$-direction.

4.5 Photoelectron Spectra and Ionization Fractions

After an HCP has been applied to an electron initially in an eigenstate $|k\rangle$, the energy spectrum of the ionized photoelectrons is obtained by projecting
the final state wave function (at any time after $\tau_{\text{pulse}}$) onto the eigenstates of the final state Hamiltonian $H_f = H_0 + H_B$ as in Ref. [28]. This is a fast and efficient method of extracting spectral features from a numerical wave function.

The probability of finding an electron of energy $E_i$ in the final state with a resolution of $2\gamma$ is given by the window function

$$P(E_i, H_f) = \langle \Psi_f | \frac{\gamma^{2n}}{(H_f - E_i)^{2n} + \gamma^{2n}} | \Psi_f \rangle. \quad (4.23)$$

For $n = 1$, this window function has a Lorentzian profile. As $n$ is increased, the shape of the window approaches a rectangular shape. Increasing $n$ improves the accuracy of the energy analysis, but the resolution of this method is $2\gamma$ for all $n$.

For $n = 2$, Eq. 4.23 can be evaluated by solving two successive linear equations for the vector $|\chi_i\rangle$,

$$\begin{align*}
(H_0 - E_i + \sqrt{i}\gamma)(H_0 - E_i - \sqrt{i}\gamma)|\chi_i\rangle &= |\Psi_f\rangle.
\end{align*} \quad (4.24)$$

$P(E_i, H_f)$ is given by $\langle \chi_i | \chi_i \rangle$. The total ionized fraction is also given by the area under this curve: $\sum_i P(E_i)$.

The resolution of this method is equal to $2\gamma$. As $\gamma$ is decreased to increase the resolution of the energy spectrum, spectral features due to the finite size of the radial grid (the box states) can be seen. This can be avoided by copying the wave function to be resolved into a larger grid and padding the outer points with zeros. However, this is a slow and expensive process and is used only if there are spectral features in the wave functions which are
narrower than the separation of the box states. All the results presented in this dissertation are at a resolution of 4 meV.

Since the applied magnetic field \((6T)\) is very small compared to the magnetic field that an electron experiences due to its motion in the field of the nucleus \((\approx 10^5T)\), we can simplify this calculation by setting \(H_f = H_0\) which makes Eq. 4.23 separable in \(r\) and \(\ell\).

### 4.5.1 Alternating Direction Iteration

In the energy analysis of the wave functions in the Coulomb-diamagnetic potential, the wave function to be analysed is projected on to the field-free basis states. The validity of this approximation is checked by finding \(P(E_i, H_f)\), using the method of Alternating Direction Iteration (ADI) [27, 75] with the full Hamiltonian \(H_f = H_0 + H_B\). This method uses the unperturbed photoelectron ionization probability \(P(E_i, H_0)\) as a first guess and iteratively determines the final photoelectron ionization probability \(P(E_i, H_f)\) to successively higher orders in the perturbation (which is of second order in the magnetic field).

Following Ref. [28], Eq. 4.23 can be written as

\[
P(E_i, H_f) = \langle \chi | \chi \rangle; \quad (4.25)
\]

where \(\chi\) is the solution to the matrix equation

\[
(H_0 + V_B - E_i + i\gamma) |\chi\rangle = \gamma |\Psi_f\rangle. \quad (4.26)
\]

Writing \(H = H_0 - E_i + i\gamma\) and \(V = V_B\), for a complex number \(r\), the above equation can be written as

\[
(H + V + r) |\chi\rangle = \gamma |\Psi_f\rangle + r |\chi\rangle. \quad (4.27)
\]
\begin{equation}
(H + r)|\chi\rangle = \gamma|\Psi_f\rangle - (V - r)|\chi\rangle.  \tag{4.28}
\end{equation}

\begin{equation}
(V + r)|\chi\rangle = \gamma|\Psi_f\rangle - (H - r)|\chi\rangle.  \tag{4.29}
\end{equation}

Starting with a first guess, \(\chi_0\) which is the solution to \(H|\chi_0 = \gamma|\Psi_f\rangle\), the iterative solutions are obtained by the prescription

\begin{equation}
|\chi_{n+1}\rangle = T_r|\chi_n\rangle + |\phi\rangle;  \tag{4.30}
\end{equation}

where

\begin{equation}
T_r = [V + r]^{-1}[H - r][H + r]^{-1}[V - r],  \tag{4.31}
\end{equation}

and

\begin{equation}
|\phi\rangle = [V + r]^{-1}\{1 - [H - r][H + r]^{-1}\}\gamma|\Psi_f\rangle.  \tag{4.32}
\end{equation}

The photoelectron ionization probability \(P(E_i, H_f)\) differs from the first guess \(P(E_i, H_0)\) by a small amount of the order of \(10^{-5}\) and converges rapidly. There is also no discernible change in the photoelectron spectra as compared to those calculated by projecting onto the unperturbed states. \(P(E_i, H_f)\) converges even at the addition of a random error of \(10\%\) to the initial guess \(\chi_0\) added at every energy point \(E_i\). To our knowledge, this is the first time that the energy window method has been used with a non-separable potential.

**4.6 Higher-order Impulse Approximation**

In a recent paper [76], a higher-order impulse approximation was used to explain the stabilization of atoms subjected to a series of HCPs [77]. The higher-order impulse approximation described in the above paper is based on a transformation from the length to the acceleration gauge. Using the
operator method of solving the time-dependent Schrödinger equation [78], we show that this approximation is gauge independent. This method has been successfully used to describe atomic processes in external static and dynamic fields. For example, the operator method was used to describe the photodetachment of $H^-$ in the presence of a static electric field [79] (see Appendix D). We use the same method to arrive at the higher-order impulse approximation of Ref. [76].

The Schrödinger equation for a bound electron in a time varying electric field $E(t)$ is

$$i\dot{\Psi}(\mathbf{r}, t) = \left[\frac{\mathbf{p}^2}{2} + V(\mathbf{r}) + E(t)z\right]\Psi(\mathbf{r}, t) \quad (4.33)$$

Following the operator method [78], a unitary time evolution operator is defined such that the time dependent wave function is a product of exponentials.

$$\Psi(t) = \exp[-i\phi(t)]\exp[-i\mu(t)A]\exp[-\lambda(t)C]$$

$$\times \exp[-i\delta(t)E]\exp[-i\nu(t)B]\Psi_0; \quad (4.34)$$

where, $\Psi_0$ is the unperturbed wave function before the application of the electric field. That is, $\Psi_0$ is the solution to the eigenvalue equation

$$H_0\Psi_0 = \left[\frac{\mathbf{p}^2}{2} + V(\mathbf{r})\right]\Psi_0 = E_0\Psi_0. \quad (4.35)$$

We choose the following,

$$A = z, \quad (4.36)$$

$$B = \frac{\mathbf{p}^2}{2} + V(\mathbf{r}), \quad (4.37)$$
\[ \dot{\mu} = E(t), \quad (4.38) \]
\[ \dot{\nu} = 1; \quad (4.39) \]

so that the Schrödinger equation satisfies

\[ i\dot{\Psi} = [\dot{\mu}A + \dot{\nu}B]\Psi. \quad (4.40) \]

A and B are time independent operators and \( \mu \) and \( \nu \) are time dependent c-numbers. A and B do not commute:

\[ \begin{align*}
C &= [A, B] = ip_z \\
\dot{a} &= [A, C] = -1 \\
E &= [C, B] = \frac{dV}{dz} \\
\dot{a}' &= [A, [A, C]] = 0 \\
F &= [C, [C, B]] = \frac{d^2V}{dz^2} \\
\dot{a}'' &= [A, E] = 0 \\
G &= [E, B] = \sum_j \frac{i}{2} \left( p_j \frac{d^2V}{dx_j dz} - \frac{d^2V}{dx_j dz} p_j \right)
\end{align*} \quad (4.41)-(4.47) \]

For the purpose of this approximation, we ignore the derivatives of \( V(r) \) of order higher than 1, but in principle, this expansion can be extended.

Inserting Eq. 4.34 in Eq. 4.33 we get,

\[ \dot{\Psi} = -i\dot{\phi}\Psi \\
- i\dot{\mu}A\Psi \\
- \lambda \exp[-i\phi] \exp[-i\mu A] C \exp[-\lambda C] \exp[-i\delta E] \exp[-i\nu B]\Psi_0 \]
\[-i\delta \exp[-i\phi] \exp[-i\mu A] \exp[-\lambda C] \exp[-i\delta E] \exp[-i\nu B] \Psi_0 \]
\[-i\nu \exp[-i\phi] \exp[-i\mu A] \exp[-\lambda C] \]
\[\times \exp[-i\delta E] B \exp[-i\nu B] \Psi_0. \quad (4.48)\]

Using the Baker-Campbell-Hausdorf expansion
\[e^{aA}B = \left( B + a[A, B] + \frac{1}{2!} a^2[A, [A, B]] + \cdots \right) e^{aA}. \quad (4.49)\]

Equation 4.48 becomes
\[\Psi = -i \left( \dot{\phi} + \mu A - i\lambda C + \lambda \mu + \delta E + \nu B - \lambda \nu E \right. \]
\[-i\mu \nu C + \frac{\mu^2}{2}\nu) \Psi. \quad (4.50)\]

Inserting the above in Eq. 4.40 and gathering the coefficients of each operator, we get
\[\dot{\phi} + \lambda \mu + \nu \frac{\mu^2}{2} = 0. \quad (4.51)\]
\[-i\lambda - i\nu \mu = 0. \quad (4.52)\]
\[\dot{\delta} - \dot{\nu} \lambda = 0. \quad (4.53)\]

By definition, \(\nu = t\) and \(\mu = \int dt E(t)\). Therefore, we can solve for these coefficients as follows.
\[\mu = \int_0^\tau dt E(t). \quad (4.54)\]
\[\nu = t. \quad (4.55)\]
\[\lambda = -\int_0^\tau dt \mu(t). \quad (4.56)\]
\[= -\int_0^\tau dt \int_0^t dt' E(t'). \quad (4.57)\]
\[\delta = \int_0^\tau dt \lambda(t). \quad (4.58)\]
\[
= - \int_0^{\tau_p} dt \int_0^t dt' \int_0^{t''} E(t'').
\]
\[
\phi = \int_0^{\tau_p} dt \frac{\mu^2}{2}.
\]
\[
= \int_0^{\tau_p} dt \frac{1}{2} \left[ \int_0^{t'} dt'E(t') \right]^2.
\]

Putting these in Eq. 4.34, we get
\[
\Psi(r, t) = \exp[-i\phi] \exp[iQz] \exp[-ipz \int_0^{\tau_p} dt \int_0^t -dt'E(t')] \cdot \exp[-i\frac{dV}{dz} \int_0^{\tau_p} -dt \int_0^t -dt' \int_0^{t''} -dt''E(t'')] \cdot \exp[-iH_0t] \Psi_0.
\]

The first term is an overall phase change with no physical effect, the second is a boost in the z-momentum, the third term is a translation in the z-direction and the fourth term is a correction in the potential. The operator \(\exp[-iH_0t]\) acts on the unperturbed wave function to give \(\Psi_0(r, t)\). The above expression is identical to equation (2) of ref. [76]. This approximation is valid when the pulse width is small so that the higher derivatives of the potential do not contribute to the wave function. In the sense of solving the time-dependent Schrödinger equation, this expression is a first-order approximation — a contrast to the term “higher-order impulse approximation”.

### 4.6.1 Effect of Pulse Shape

Let the electric field of the HCP take the form
\[
E_{HCP} = E; \quad (0 < t < \tau_p)
\]
\[
= 0; \quad \text{otherwise.}
\]

The total momentum transferred to an electron or the impulse \(Q\) is
\[
Q = -E\tau_p.
\]
From Eq. 4.59, we get

\[ \Psi(r, t) = \exp[-i \frac{Q^2}{6} \tau_p] \exp[iQz] \exp[-i \frac{Q}{2} \tau_p p_z] \]
\[ \cdot \exp[i \frac{dV}{dz} \frac{\tau_p^2}{6} Q] \Psi_0(r, t). \]  

(4.62)

Taking our model of the HCP,

\[ E(t) = \sin(\frac{\pi}{\tau_p} t); \quad (0 < t \leq \tau_p) \]  
\[ = 0; \quad \text{otherwise}, \]  

(4.63)

the total momentum transferred is given by the expression

\[ Q = -2 \frac{\tau_p}{\pi}. \]  

(4.64)

From Eq. 4.59, we get

\[ \Psi(r, t) = \exp[-i \frac{Q^2}{6} \tau_p] \exp[iQz] \exp[-i \frac{Q}{2} \tau_p p_z] \]
\[ \cdot \exp[i \frac{dV}{dz} \frac{\tau_p^2}{6} Q] \Psi_0(r, t). \]  

(4.66)

We can see that changing the pulse shape has no effect in this approximation.

As \( \tau_p \to 0 \),

\[ \Psi = \exp[-iQz] \Psi_0; \]  

(4.67)

which is the sudden impact approximation or the zeroth order impulse approximation. Keeping terms to first order in \( \tau_p \), we have

\[ \Psi = \exp[-i \frac{Q^2}{2} \tau_p] \exp[-iQz] \exp[iQ \tau_p p_z] \Psi_0, \]  

(4.68)

which is the first order impulse approximation but still contains only the zeroth order term in the potential. Keeping terms to second order in \( \tau_p \), we
get the complete expression which is the second order impulse approximation but has the first order approximation in the potential. The exponent in the second order correction term is proportional to the gradient of the potential in the $z$-direction. For a typical $Q$ of $\sim \frac{1}{n}$ and $\tau_p$ of 100fs, the second-order correction term is small only for states with large $n$ where the potential $V(r)$ is flat at the classical radius of $\sim n^2$.

For our system - the diamagnetic atom, the potential is

$$V(r) = -\frac{1}{r} + \frac{B^2}{8}(x^2 + y^2). \quad (4.69)$$

$$\frac{dV}{dz} = \frac{z}{r^3}. \quad (4.70)$$

Along the $z$-axis, as $z$ increases, this gradient rapidly goes to zero, particularly where the lowest energy states are localized. At a large distance from the $z$-axis, at a small value of $z$, the gradient is small but finite.

For alkali atoms where we use a pseudopotential, the expression $i\frac{dV}{dz}$ cannot be used, so we must retain the commutator $i[p_z, V(r)]$ and apply it during propagation.

4.6.2 The Second-order Impulse Approximation in the R-L Basis

Taking our model of the HCP, the wave function at a time $t$ is

$$\Psi(r, t) = \exp[-i\frac{Q^2}{6}\tau_p] \exp[-iQz] \exp[i\frac{Q}{2}\tau_pp_z]$$

$$\cdot \exp[-i\frac{dV}{dz}\frac{\tau_p^2}{6}Q] \Psi_0(r, t). \quad (4.71)$$

The initial wave function $\Psi_0$ is known in the $r$-$\ell$ basis. The boost operation alone can be done in the usual way. The second operation (corresponding to a translation) is harder since it mixes both the radial wave functions at
the adjacent grid points as well as the angular momenta. This becomes a giant 3D matrix which we cannot diagonalize. The problem is to perform this z-translation in a spherical basis, which we now describe.

The most time-consuming step in making this wave function is the z-shift. One idea is to use the operator for $p_z$ in the $r - \ell$ basis (see Appendix E), rewrite it for a non-linear grid and apply it in the expansion of the exponential until the expansion converges (ie, the norm of the next term being added is smaller than a very small number $10^{-20}$ or so). i.e.,

$$\exp\left[i\frac{Q}{2}\tau p_z\right]\psi = \exp[i\delta z p_z]\psi$$

$$= \left[1 + i\delta z p_z + \cdots + \frac{(i\delta z p_z)^n}{n!} + \cdots\right]\psi.$$ (4.72)

The problem with this method is that convergence is extremely expensive and too much numerical noise builds up in the norm. The highest momentum which controls the size of the $\delta z$ we can take is not the momentum of the state itself but the inverse of the smallest grid spacing - which in the non-linear grid is very small.

Another extension of the same method is to write

$$\exp[i\delta z p_z]\psi = \left(\exp\left[i\frac{\delta z}{m p_z}\right]\right)^m\psi$$ (4.73)

so that the exponent gets smaller and see if we can achieve convergence faster for each exponential expansion. This is also a very expensive method. To use either of the two methods to calculate a shift due to a 100fs HCP would take a year for a single $n=30$ state!

A stable method is to write the exponential itself as a propagator of sorts using a second-order symmetrized product formula algorithm [80]. The
operator $p_z$ in the $r - \ell$ basis couples both the adjacent radial points and the adjacent $\ell$ points. Therefore writing this in a $r - \ell$ basis makes it a three-dimensional matrix. $p_z$ is written as a sum of two operators $R$ and $\Theta$, which do not commute.

$$p_z = \Theta + R.$$  \hspace{1cm} (4.74)

Correct through second order in $\delta z$,

$$\exp[-i\delta z p_z] = \exp[-i\frac{\delta z}{2} R] \ast \exp[-i\frac{\delta z}{2} \Theta] \ast \exp[i\frac{\delta z}{2} \Theta] \ast \exp[i\frac{\delta z}{2} R].$$  \hspace{1cm} (4.75)

Each of the $R$ and $\Theta$ parts is split into the sum of two block diagonal matrices, each part containing either the odd or the even $\ell$ values (See Appendix F). This $2 \times 2$ splitting method has been previously used for cartesian grids [80] and recently for a non-uniform radial grid in calculations of multi-photon processes [81]. For small enough $\delta z$ (which depends only on the largest momentum in the state), the norm of the wave function is conserved. For $n = 20$ and a threshold impulse $Q$ of 0.052a.u., the norm is conserved for $\delta z \approx 0.1a.u$. That is, for a $z$-shift of about 100a.u., it would take a HCP of 100fs, taking 1000 of these ‘propagation’ steps. This is the method that we are currently using to carry out the higher-order impulse approximation in the $r - \ell$ basis.

The theoretical methods described in this chapter are used in calculating with a high degree of accuracy the (known) static properties of the atom in an external magnetic field such as probability distributions and localization characteristics. The true power of this theory lies in the ability to study
the dynamics in the system, be it the time evolution of wave packets or the effect of external fields on these atomic systems. In addition, ionization can also be studied, yielding not only the total ionized population (as given by previous theories) but also the spectra of the ionized photoelectrons. All the results (which are presented in the following chapter) are in a form that can easily be used by an experimentalist attempting to perform experiments on this system.
5. RESULTS AND DISCUSSION

We study the HCP ionization of alkali atoms in a magnetic field [82]. The ionization is studied in two limits — when the HCP width is much smaller than typical time scales in the system (the impulsive limit) and in the opposite limit when the HCP width is long enough to be comparable to typical time scales in the system. We show the suppression of ionization as the pulse width of the HCP increases. As $n$, the principal quantum number of the states increases, we show that the ionization properties are scaled and approach classical behavior. The effect of a non-hydrogenic core is studied by calculating the HCP ionization of sodium.

5.1 HCP Ionization of Diamagnetic Hydrogen

We begin with hydrogen in a magnetic field of $B = 6\, \text{T} = 2.55 \times 10^{-5}\, \text{a.u.}$ along the $z$-direction. The states corresponding to the $n = 20$ ($m = 0$) manifold are within the $\ell$-mixing regime. Choosing the lowest energy (valley), highest energy (ridge), and separatrix states of even parity from this manifold, we study their ionization behavior. The ionizing HCP is linearly polarized with its electric field vector in the $z$-direction.

5.1.1 Ionization in the Impulsive Limit

Figure 5.1 shows the ionized fractions of the valley, ridge and separatrix states as a function of the momentum transferred by the HCP in the impulse approximation. For a free, classical electron of momentum $p_i$, the final momentum after its interaction with a HCP is $p_f = p_i + Q$, where $Q$ is the
Figure 5.1: Impulsive ionization of diamagnetic states of H. Calculated ionized fractions of photoelectrons as a function of total momentum transferred (impulse approximation). Solid line: lowest energy state. Dashed line: highest energy state. Dot-dashed line: a separatrix state.
momentum transferred by the HCP. As discussed in Chap. 3, the change in its energy is equal to

$$\Delta E = \frac{p_f^2}{2} - \frac{p_i^2}{2} = p_i \cdot Q + \frac{Q^2}{2}. \quad (5.1)$$

To ionize an electron, this change in energy must exceed the binding energy $|E_b|$. Since the electric field of the HCP is in the z-direction, the initial z-momentum of an electron that has threshold energy after the pulse is given by

$$p_{0z} = (|E_b| - \frac{Q^2}{2})/Q. \quad (5.2)$$

The probability that an electron is ionized in this classical impulse approximation is just the probability that its initial momentum in the z-direction is greater than $p_{0z}$ [24]. In this approximation, the slope of the curve in Fig. 5.1 is proportional to the dispersion in the z-component of the momentum of the state.

Using the technique of Impulsive Momentum Retrieval, the momentum distributions of the three states is found and shown below. If a state has a large z-component to its momentum, it does not require much Q to be ionized and vice-versa. The highest energy state has a much steeper threshold than the lowest energy or separatrix states. One can see that the highest energy state, which is localized perpendicular to the magnetic field, has very little momentum in the z-direction so that there is very little likelihood of ionization for $Q$ less than the threshold value of 0.052 a.u. The dispersion in the z-momentum is also very small. The lowest energy and separatrix states
Figure 5.2: Momentum distributions of diamagnetic Zeeman states of H. The $z$-momentum distribution of $n = 20$ even parity states of hydrogen in a magnetic field of 6T. (a) Lowest energy state (parallel to the magnetic field), (b) highest energy state (perpendicular to the magnetic field) and (c) separatrix state.
have significant probability densities and momenta along the $z$-direction and therefore show similar ionization curves with appreciable ionization even at lower $Q$.

Figure 5.3 shows the ionization spectra obtained in the impulse approximation for the three states (ridge, valley, and separatrix) ionized by an HCP. We compare a soft kick, which transfers energy just above threshold and a hard kick, which completely ionizes the Rydberg electron. At threshold, only those parts of the electron probability distribution which have positive $z$-momentum get ionized. The distribution for the soft kick is therefore peaked at zero energy. For a hard kick, the spectra are symmetric and peaked at the energy corresponding to $Q^2/2 - |E_b|$, where $E_b$ is the energy of the bound state. From the discussion in the previous paragraph, this implies that the average $z$-momentum is equal to zero. Such a feature is also seen in the total ionization cross section for charged particle collisions which are peaked about the energy-momentum conserving value [83]. The ionization of an electron by a HCP, an electromagnetic pulse, shows characteristics of collisional ionization by charged particles when the pulse width of the HCP is very small (the impulsive limit). The spectrum of the lowest energy state in Fig. 5.3(a) shows an oscillatory structure corresponding to the two possible $z$-momentum directions — along and opposite the HCP field. The highest energy state (Fig. 5.3(c)), which has very little $z$-momentum, does not show this structure. The separatrix state seen in Fig. 5.3(b) shows a complex structure in its spectrum which reflects its complex momentum distribution.
Figure 5.3: Photoelectron spectra of diamagnetic states ionized by an HCP. Calculated differential ionization probabilities of diamagnetic Zeeman states of hydrogen ionized by a HCP (impulse approximation) as a function of photoelectron energy for (a) lowest (b) highest and (c) separatrix states. Solid line: hard kick, $Q = 0.2$ a.u. Dashed line: soft kick, $Q = 0.052$ a.u. The area under the curve yields the total ionized fraction.
5.1.2 Effect of Increasing HCP Width: The Non-impulsive Limit

In Fig. 5.4, we study the ionized fractions of the lowest, highest and separatrix $n = 20$ states as a function of the width of the HCP while keeping the total impulse $Q$ constant. $Q$ is chosen to be 0.052a.u. such that the energy $\frac{Q^2}{2} - |E_b|$ is just above threshold. In all three cases, the ionization is suppressed as the HCP width becomes comparable to the Kepler period $(2\pi n^3 \approx 1.2\text{ps})$. Clearly, the nature of this decrease in ionization depends on the localization of the state. The lowest energy states, localized in the direction of the magnetic field, show a sharp suppression of ionization when the HCP width becomes approximately equal to one half of the Kepler period. The highest energy states, localized perpendicular to the magnetic field, show a smooth decrease in ionization with increasing pulse width. The separatrix states show the least loss of ionization. The photoelectron spectra at $\tau_{\text{pulse}} = 0$, $\frac{\tau_{\text{Kepler}}}{2}$ and $\tau_{\text{Kepler}}$ seen in Fig. 5.5 show that the energy gained by the photoelectron decreases as the pulse width is increased, thus decreasing the ionized fraction. We now discuss the dynamics of this "energy loss" as a function of pulse width.

In the impulse approximation, the electron does not have time to move during the pulse and interacts only with the electromagnetic field of the HCP. In the opposite limit, when the pulse width is long, the electron may interact with the ion core and the diamagnetic potential, as well as the electromagnetic field during ionization. The change in energy of an electron due to the HCP (treating the electron as a classical particle) is given by:

$$\Delta E = - \int_0^{\tau_{\text{pulse}}} dt \, \mathbf{v} \cdot \mathbf{E}_{\text{HCP}},$$  \hspace{1cm} (5.3)
Figure 5.4: Non-impulsive ionization of diamagnetic states of H. Effect of width of HCP on ionization of $n = 20$ diamagnetic states (circles: lowest, squares: highest, triangles: separatrix). The total momentum transferred $Q$ is kept constant at 0.052 a.u.
Figure 5.5: Photoelectron spectra of diamagnetic states of H with increasing HCP width. Calculated photoelectron spectra of \( n = 20 \) diamagnetic states when the HCP width is (i) 0 (impulse approximation) (ii) \( \frac{\tau_{\text{Kepler}}}{2} \) (iii) \( \tau_{\text{Kepler}} \) for (a) lowest (b) highest and (c) separatrix states. The decrease in the total ionized fraction is accompanied by a smaller photoelectron energy.

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where \( v \) is the velocity of the electron. When interacting with the ion core, the electron changes its direction of motion thus gaining less net energy than it would have otherwise. The ionization dynamics may be expected, therefore, to show a dependence on the pulse width of the HCP at time scales corresponding to the interaction of the electron with the ion core, that is, the Kepler orbital period. The lowest energy state is localized along the direction of the magnetic field and can interact with the ion core on a time scale of one half the Kepler period (since the initial electron probability distribution is peaked near the classical turning point as seen in Fig. 2.4(a)). This is reflected in the sudden drop in the ionized fraction as \( \tau_{\text{pulse}} \sim \frac{t_{\text{Kepler}}}{2} \). The highest energy states are localized perpendicular to the magnetic field and interact differently with the core. The separatrix states are more delocalized and do not lose as much energy as the lowest or highest energy states.

5.1.3 Effect of Increasing \( n \), the Principal Quantum Number

These results may be understood better by studying the localized states of different manifolds.

Figure 5.6 shows calculations performed for four manifolds, \( n = 30 \) at \( B = 1T \), \( n = 20, 18, 16 \), at \( B = 6T \). These states are still within the \( \ell \)-mixing regime. The ionization probability (in the impulsive limit) as a function of \( Qn \), the product of the momentum imparted by the HCP and the principal quantum number (the classically scaled momentum \( \langle p \rangle_n = \frac{1}{n} \)), is seen to be universal for the states of the same localization. Figure 5.7 shows the ratio of the ionized fraction at a finite HCP width to that in the impulse.
Figure 5.6: Scaling of ionized fraction in the impulsive ionization of diamagnetic states of H. Ionization curves in the impulse approximation as a function of the total momentum transferred times \( n \) for diamagnetic states of hydrogen: \( n = 30 \) (diamonds), \( n = 20 \) (circles), \( n = 18 \) (squares) and \( n = 16 \) (triangles). Open points: lowest energy states. Filled points: highest energy states.
Figure 5.7: Scaling of ionization properties in the non-impulsive ionization of diamagnetic states of H. Plot of ratio of ionized fraction at finite HCP width to that in the impulse approximation for the same momentum transferred vs the ratio of HCP width to the Kepler orbital period $2\pi n^3$. $n = 20$ (circles), $n = 18$ (squares) and $n = 16$ (triangles). (a) lowest (b) highest energy states.

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approximation for threshold $Q$ plotted against the ratio of the HCP width to the classical Kepler period $\tau_{Kepler} \simeq 2\pi n^3$ for both the lowest and highest states. These are calculated for $n = 16, 18$ and 20 at $B = 6T$, with Kepler periods of $\sim 0.6$ ps, 0.9 ps and 1.2 ps, respectively, ionized by pulses with $Q$ values corresponding approximately to the threshold energies of the states. The curves lie on top of each other, indicating that the Kepler orbital period is the significant time scale for each of these localized states. The drop in the ionization probability of the lowest energy states at approximately $\tau_{Kepler}/2$ is steeper as the value of $n$ increases. This suggests that as $n$ increases, the system approaches classical behavior wherein an electron starting from a point in the orbit furthest from the ion core (i.e., near the classical turning point) can interact with the core at times $\tau_{Kepler}/2$ and again at $3\tau_{Kepler}/2$.

5.1.4 Ionization of $n = 30$ States by a 0.5ps HCP

For a magnetic field of 1T, the $n = 30$ ($m = 0$) states of hydrogen are also in the $\ell$-mixing regime. It has been assumed that the impulse approximation is good when the width of the HCP is much smaller than the Kepler orbital period corresponding to the state to be ionized. We test this assumption by comparing the ionization of the states by a 0.5ps HCP with the ionization by an impulsive kick. The Kepler orbital period corresponding to states with $n = 30$ is roughly 4ps. Figure 5.8 shows the ionized fractions of the three classes of states as a function of $Q$, the momentum transferred, keeping the pulse width of the HCP 0.5ps. The corresponding values in the impulsive limit are also indicated. Clearly, the impulse approximation is a good approximation for the lowest energy and separatrix $n = 20$ states ionized by
Figure 5.8: Ionization of $n = 30$ states by a 0.5ps HCP. Calculated ionized fractions of photoelectrons as a function of total momentum transferred in $n = 30, m = 0$, even parity diamagnetic states of hydrogen. Solid line: 0.5ps HCP. Dashed line: impulse approximation. (a) Lowest energy state (b) Highest energy state (c) separatrix state.
a 0.5ps HCP. For the highest energy state, the same sensitivity to pulse width described in the previous subsections manifests itself here. It is interesting that even at very large $Q$, there is a significant deviation from impulsive behavior.

5.1.5 The Higher-order Impulse Approximation

We attempted to explain this sensitivity of the highest energy state to the HCP width by using the higher-order impulse approximation described in the previous chapter. The higher-order impulse approximation consists of three contributions — first, the momentum boost in the $z$-direction which is the usual impulse approximation, second, a translation in the $z$-direction and third, a contribution from the derivative of the potential which is a smaller effect. This approximation was computed using a stable symmetrized product formula algorithm. We find that the higher-order impulse approximation is not a good approximation for the states that we ionize.

5.2 HCP Ionization of Diamagnetic States of Sodium

We have repeated many of the hydrogen calculations using a pseudopotential appropriate for describing valence orbitals in sodium. In sodium, the quantum defect in the $s$ and $p$ channels causes two states, one of $s$-character and one of $p$-character, to split off in energy from the rest of the unperturbed hydrogenic states of a single $n$-manifold. When a magnetic field of $B = 6T$ is applied, these degenerate states do not contain significant contributions from the $\ell = 0$ or $\ell = 1$ angular momenta but retain all the other features just as in hydrogen. We choose the lowest energy, separatrix and highest energy states of the remaining $n = 20$, $m = 0$ even parity manifold and the $s$ split
Figure 5.9: Impulsive ionization of diamagnetic states of sodium. Calculated ionized fractions of photoelectrons as a function of total momentum transferred (impulse approximation) in $n \approx 20$, $m = 0$, even parity diamagnetic states of sodium. Solid line: lowest energy state. Dashed line: highest energy state. Dot-dashed line: separatrix state. Filled circles: $s$ split-off state.
Figure 5.10: Photoelectron spectra of diamagnetic states of sodium ionized by an HCP. Calculated ionization spectra of diamagnetic states in sodium ionized by HCP (impulse approximation) for (a) lowest (b) separatrix (c) highest and (d) s split-off states. Solid line: hard kick, $Q = 0.19$ a.u. Long-dashed line: soft kick, $Q = 0.05$ a.u.
off state nearest in energy. When ionized by the HCP in the impulsive limit, the highest, lowest and separatrix states of the $n = 20$ manifold show similar behavior as in hydrogen, as shown in Fig. 5.9. The nearest $s$ split-off state, which has mainly $\ell = 0$ character, shows significantly different behavior. Its photoelectron spectrum is also very different from that of the localized diamagnetic states as shown in Fig. 5.10. But, for the localized states, the quantum defect does not change the half-cycle pulse ionization characteristics qualitatively.
6. CONCLUSION

6.1 Summary

In summary, we have presented an efficient theoretical technique for studying the dynamical evolution of atomic systems in external fields. We have shown that the HCP ionization dynamics of Zeeman states in the $\ell$-mixing regime depends strongly on the localization of the initial diamagnetic state. In the limit of very short HCPs ($t_{\text{pulse}} \ll t_{\text{Kepler}}$), the determination of the ionized fraction as a function of the momentum transferred by the pulse is a direct measure of the $z$-momentum of these states.

As the width of the HCP increases (keeping the total momentum transferred a constant), the ionization of the diamagnetic states is suppressed. The decrease in ionization is also accompanied by a decrease in the energy gained by the Rydberg electron during ionization, as seen in the calculated photoelectron spectra. The decrease in the ionization fraction is also sensitive to the localization of the initial states. We find that the states that are localized perpendicular to the magnetic field are more sensitive to the increase in HCP width than the states localized parallel to the magnetic field. This effect is understood qualitatively at present.

Similarities in the HCP ionization of localized diamagnetic states in hydrogen and sodium point to the features of ionization being a general characteristic of localized diamagnetic states of atoms in the $\ell$-mixing regime. These measurements may be made at reasonable laboratory magnetic fields.
Indeed, our preliminary results have prompted plans for such experiments which will soon be under way[100].

6.2 Future Work

The techniques used in this work can be used to study several interesting problems. This dissertation presents the HCP ionization of diamagnetic Zeeman states. Two HCPs may be used in a pump-probe study, where the first HCP is used to excite or ionize a wave packet and the second HCP is used to detect it. Thus, time resolved spectroscopy may be carried out as in the case of Stark wave packets [21]. The creation of wave packets in the diamagnetic atom by using specifically tailored HCPs is an example of a system that can be coherently controlled to produce interesting target states. These novel states can then be ionized by an HCP.

Another example of a system with localized states is an atom in parallel electric and magnetic fields [29]. Our theoretical techniques can easily be extended to study this system. The presence of two external fields in this system translates to an extra element of control in the study of quantum control processes.

It has been proposed that the study of Rydberg wave packet dynamics will yield information about the stable orbits in the diamagnetic system [57]. Using an HCP, a wave packet may be made with which the dynamics of the diamagnetic system in the near-threshold regions can be investigated [43]. The ionization of diamagnetic states in the n-mixing regime may be used to study the region of transition between order and chaos in the diamagnetic system. Since we can calculate the time-evolution of a wave function, we can
also study wave packet evolution along classical structures [101] and core scattering in alkali-metal atoms[102].
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APPENDIX A: ATOMIC UNITS

Atomic units (a.u.) were first introduced by Hartree [84] in 1928. In these units, the charge of an electron, \( e \), the mass of an electron, \( m_e \), and the "rationalized Planck's constant", \( \hbar \), are set equal to unity [35].

\[
e = m = \hbar = 1.
\] (A.1)

One immediate consequence of this definition is that the speed of light, \( c \), as defined by the fine structure constant \( \alpha \) as

\[
\alpha = \frac{e^2}{\hbar c}.
\] (A.2)

is

\[
c \approx 137.
\] (A.3)

\( \alpha \) is also the velocity of an electron in the first Bohr orbit.

A.1 Atomic Units of Some Physical Quantities

1 a.u. of charge = \( 1.6 \times 10^{-19} \text{C} \).
1 a.u. of mass = \( 9.1 \times 10^{-31} \text{kg} \).
1 a.u. of length = \( 5.29 \times 10^{-11} \text{m} \).
1 a.u. of energy = \( 27.21 \text{eV} \).
1 a.u. of time = \( 2.41889 \times 10^{-17} \text{s} \).
1 a.u. of electric field strength = \( 5.142 \times 10^9 \text{V/cm} \).
1 a.u. of magnetic field strength = \( 2.35 \times 10^5 \text{T} \).
A.2 A Few Useful Conversions

1 a.u. of energy is equal to twice the ionization potential of hydrogen (the famous 13.6eV) and is equal to 219474.6 cm\(^{-1}\).

1 eV is equal to 8068.92 cm\(^{-1}\) (wavenumbers).

1 ps is equal to 41341.27 a.u. of time.

6 T is equal to 15.32 \times 10^{-5} a.u. of magnetic field strength.

300 V/cm is equal to 5.83 \times 10^{-8} a.u. of electric field strength.

The bandwidth of a transform limited pulse is given by \(\frac{4\pi a^2}{r_p}\). A 1 ps pulse has a bandwidth of \(\sim 14.72\) cm\(^{-1}\).

A.3 Classical Scaling of Atomic Properties

A few properties of Rydberg atoms that classically scale with the principal quantum number, \(n\), are: The binding energy \(\propto -\frac{1}{n^2}\).

Energy level spacing \(\propto \frac{1}{n^3}\).

The radiative lifetime \(\propto n^3\).

The orbital radius \(\propto n^2\).

The Kepler orbital period \(\propto n^3\).

The classical momentum \(\propto \frac{1}{n}\).

The first-order Stark shift \(\propto n^2\).

Stark crossing field \(\frac{1}{3n^2}\).

Threshold of field ionization \(\frac{1}{9n^4}\).

The diamagnetic energy \(\propto n^4\).

Zeeman crossing field \(\propto \frac{1}{n^2}\).
APPENDIX B: DISCRETIZATION OF THE HAMILTONIAN ON A NON-UNIFORM GRID

We would like to solve the Schrödinger equation for an electron in a central potential (Coulomb or an alkali pseudopotential) by discretizing the Hamiltonian on a non-uniform radial grid [71]. The radial grid has N+1 points with indices from 0 to N and the grid ranging from \( r_{\text{min}} \approx 0 \) to \( r_{\text{max}} \). We assume that the grid is large enough to contain the wave function completely, i.e.,

\[
\psi(r_{\text{max}}) = 0. \quad (B.1)
\]

We also assume that the wave function is well-behaved so that

\[
\lim_{r \to 0} r \psi = 0, \quad (B.2)
\]

\[
\lim_{r \to 0} r \frac{\partial \psi}{\partial r} = 0. \quad (B.3)
\]

In order to avoid numerical overflow, the radial distance at the 0\(^{th}\) grid point is not zero, but

\[
r_0 = -r_1, \text{ i.e.,} \quad \frac{r_1 + r_0}{2} = 0. \quad (B.4)
\]

The action \( S \) is defined as [75, 85]

\[
S = \int_{t_1}^{t_2} \mathcal{L}(\Theta, \Theta^*), \quad (B.5)
\]

where \( \mathcal{L} \) is

\[
\mathcal{L} = \langle \Psi | i \frac{\partial}{\partial t} - H_0 | \Psi \rangle, \quad (B.6)
\]

\[
= \langle \Psi | i \frac{\partial}{\partial t} - T - V | \Psi \rangle; \quad (B.7)
\]

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where $T$ and $V$ are the kinetic and potential energy operators respectively. When the variation in the action with respect to $\Psi^*$ for fixed end points $t_1$ and $t_2$ is set to 0, it yields an equation of motion for $\Psi$. This is equivalent to $\Psi^*$ satisfying the Euler-Lagrange condition

$$\frac{d}{dt} \left( \frac{\partial \mathcal{L}}{\partial \dot{\Psi}^*} \right) - \frac{\partial \mathcal{L}}{\partial \Psi^*} = 0. \quad (B.8)$$

In spherical coordinates, the wave function $\Psi$ may be written as a sum over radial functions $\psi_{\ell}(r)$ times spherical harmonics $Y_{\ell m}$. That is,

$$\Psi = \sum_{\ell} \psi_{\ell}(r) Y_{\ell m}(\theta, \phi). \quad (B.9)$$

The expression for $\mathcal{L}$ can be written separately for each value of $\ell$. Thus for a particular value of the angular momentum $\ell$, the expectation value of the kinetic and potential energies can be written as

$$\langle T \rangle = \frac{1}{2} \int_0^{r_{\text{max}}} dr \cdot r^2 \cdot |\partial \psi_{\ell}| \quad (B.10)$$

$$\langle V \rangle = \int_0^{r_{\text{max}}} dr \cdot r^2 \psi_{\ell}^* \left( V_{\ell}(r) + \frac{\ell(\ell + 1)}{2r^2} \right) \psi. \quad (B.11)$$

Using these integral expressions permits us to deal efficiently with the boundary conditions as $r \rightarrow 0$.

To discretize the function $\mathcal{L}$ on a non-linear grid, we define an integration rule. Functions are evaluated at each grid point $r_j$. Derivatives are evaluated at the midpoint between grid points. Distance increments are calculated as the distance between the half-points. That is,

$$f(r) \rightarrow f_j(r_j). \quad (B.12)$$

$$\left| \frac{\partial f}{\partial r} \right|^2 r^2 dr \rightarrow \left[ \frac{f_{j+1} - f_j}{r_{j+1} - r_j} \right]^2 \left( r_{j+1} - r_j \right) \left[ \frac{r_{j+1} + r_j}{2} \right]^2. \quad (B.13)$$

$$\Delta r_j \rightarrow \frac{r_{j+1} + r_j}{2} - \frac{r_j + r_{j-1}}{2} = \frac{r_{j+1} - r_{j-1}}{2}. \quad (B.14)$$
This three-point integration rule ensures that the discretized equation is correct to the second-order in the grid spacing. For a single $\ell$, $\mathcal{L}$ is written as

$$
\mathcal{L} = \sum_{j=1}^{N} \psi_j^* \psi_j r_j^2 \left[ \frac{r_{j+1} - r_{j-1}}{2} \right] - \sum_{j=1}^{N} \psi_j^* \tilde{V}_j \psi_j r_j^2 \left[ \frac{r_{j+1} - r_{j-1}}{2} \right] - \frac{1}{2} \sum_{j=1}^{N} \frac{(\psi_{j+1}^* - \psi_j^*)(\psi_{j+1} - \psi_j)}{(r_{j+1} - r_j)} \left[ \frac{r_{j+1} + r_j}{2} \right]^2 ; \quad (B.15)
$$

where $\tilde{V}_j$ is the effective potential including the centrifugal barrier.

$$
\tilde{V}_j = V_j + \frac{\ell(\ell + 1)}{2r_j^2} . \quad (B.16)
$$

Collecting the terms with $\psi_j^*$ in the expression for $\mathcal{S}$, and defining a radial function $g_j$ such that

$$
g_j = r_j \sqrt{\Delta r_j \psi_j} , \quad (B.17)
$$

we get

$$
\mathcal{L} = \sum_{j=1}^{N} g_j^* i g_j - \sum_{j=1}^{N} g_j^* \tilde{V}_j g_j
= \frac{1}{2} \sum_{j=1}^{N} g_j^* \left\{ \frac{g_{j+1}}{(r_{j+1} - r_j)} \sqrt{\frac{4}{(r_{j+1} - r_j)(r_{j+2} - r_{j+1})(r_{j+1} - r_{j-1})}} \frac{1}{r_{j+1} r_j} \left( \frac{r_{j+1} + r_j}{2} \right)^2 
+ \frac{g_{j-1}}{(r_j - r_{j-1})} \sqrt{\frac{4}{(r_j - r_{j-1})(r_j - r_{j-2})(r_j - r_{j-1})}} \frac{1}{r_j r_{j-1}} \left( \frac{r_j + r_{j-1}}{2} \right)^2 
- \frac{2g_j}{r_{j+1} - r_{j-1}} \left( \frac{1}{r_j - r_{j-1}} \left( \frac{r_j + r_{j-1}}{2r_j} \right)^2 
+ \frac{1}{r_{j+1} - r_j} \left( \frac{r_{j+1} + r_j}{2r_j} \right)^2 \right) \right\} . \quad (B.18)
$$
Imposing the Euler-Lagrange condition with respect to $g_j^*$ gives the time-evolution equation for $g_j$. This is a symmetric tridiagonal matrix.

$$i\dot{g}_j = \tilde{V}_j g_j - \frac{1}{2} [C_j g_{j+1} - 2D_j g_j + C_{j-1} g_{j-1}], \quad (B.19)$$

where,

$$C_j = \frac{2}{(r_{j+1} - r_j)} \frac{1}{(r_{j+2} - r_j)(r_{j+1} - r_{j-1})} \times \frac{1}{r_{j+1} r_j} \left[ \frac{r_{j+1} + r_j}{2} \right]^2, \quad (B.20)$$

$$D_j = \frac{1}{r_{j+1} - r_j} - 1 \left( \frac{1}{r_j - r_{j-1}} \left[ \frac{r_j + r_{j-1}}{2r_j} \right]^2 \right. \left. \frac{1}{r_{j+1} - r_j} \left[ \frac{r_{j+1} + r_j}{2r_j} \right]^2 \right), \quad (B.21)$$

$$\tilde{V}_j = V_j + \frac{\ell(\ell + 1)}{2r_j^2}. \quad (B.22)$$

From the definition of $g_j$, we can see that $g_j$ is normalized such that if

$$\sum_j \Delta_j r_j^2 |\psi_j|^2 = 1, \quad (B.23)$$

then

$$\sum_j |g_j|^2 = 1. \quad (B.24)$$

For a defined non-linear grid, the coefficients $C_j$ and $D_j$ can be computed and the eigenvalues $E_{nl}$ and the eigenfunctions $g_j^\ell$ of the real symmetric tridiagonal matrix in Eq. B.19 can be found by using standard algorithms.
APPENDIX C: THE NON-UNIFORM RADIAL GRID

We use a non-uniform radial grid [71] to discretize the field-free Hamiltonian in an alkali metal. From the analytical expressions for the radial part of the eigenfunctions of the hydrogen atom [30], we know that radial wave functions are oscillatory towards the origin but relatively smooth far away. The 'wavelength' of these oscillations near the origin is independent of energy (principal quantum number). This is advantageous because states with different principal quantum numbers can be represented on the same radial grid.

We use a radial grid of \( N_r \) points. We specify the smallest grid spacing required \( \Delta_{\text{min}} \) and the maximum grid spacing allowed \( \Delta_{\text{max}} \). To represent hydrogenic wave functions accurately, the grid spacing is smallest near the origin and constant near the outer edge of the box. An acceleration parameter \( \alpha \) determines the rate at which the grid spacing changes from \( \Delta_{\text{min}} \) near the origin to \( \Delta_{\text{max}} \) near the outer edge of the box as shown in Fig. C.1. The grid is defined as

\[
    r_i = r_{i-1} + \Delta_{\text{min}} + (1 - e^{-\alpha r_{i-1}})(\Delta_{\text{max}} - \Delta_{\text{min}}), \quad (C.1)
\]

with the convention that the first grid point is at

\[
    r_1 = +\Delta_{\text{min}}. \quad (C.2)
\]

Grid points \( r_0 \) and \( r_{N+1} \) are defined but used only to take care of the edge effects. The maximum extent of the grid is \( r_N \). This is determined by the

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Figure C.1: The non-uniform radial grid. The grid spacing in a non-uniform radial grid is small near the origin and a constant near the outer edge of the radial box.
grid parameters. We choose the extent of the grid to be about 2.5 times the classical radius of the state that we wish to study.

Typical grid parameters used in this study are

\[ N = 3500 \]
\[ \Delta \text{min} = 0.01 \]
\[ \Delta \text{max} = 5 \]
\[ \alpha = 0.0005. \] (C.3)

These accurately represent hydrogenic wave functions for principal quantum numbers up to \( n = 35 \).

The quality of the grid depends sensitively on \( \alpha \) and \( \Delta \text{max} \). \( \alpha \) must be small enough that the grid is closely spaced for a sufficient radial distance so that all the oscillations in the wave functions are correctly represented, but large enough that the grid spacing increases towards the outer edge where the wave functions are smooth. The grid is very sensitive to \( \Delta \text{max} \) which must be small enough to allow for a relatively small \( \alpha \), but large enough so that the number of grid points required does not increase too much. Merely increasing \( N \), the number of gridpoints, just increases the size of the grid with points spaced at \( \Delta \text{max} \). The grid is not very sensitive to \( \Delta \text{min} \) which must be smaller than the smallest oscillation in the wave function.

We choose the grid parameters by comparing the numerical hydrogenic wave functions produced with the analytical wave functions. The energy eigenvalues have a maximum error of 0.01% and the wave functions are in
excellent agreement with the analytic solutions to the radial part of the Schrödinger equation for \( n \) up to 35.
APPENDIX D: PHOTODETACHMENT IN COMBINED STATIC AND DYNAMIC ELECTRIC FIELDS

Through an exact solution of the time-dependent Schrödinger equation for an electron in a static electric field plus the time-dependent electric field of the detaching radiation, the photodetachment cross-section of $H^-$ is calculated. Careful attention is paid to ensuring proper limiting behavior as the frequency of the time-dependent field goes to zero. We do not find observable effects of a cross-term between the two fields on the detachment cross-section. Our results point to possible gauge dependence and other difficulties of S-matrix formulations of multi-photon detachment and ionization [79].

Following a detailed experimental study [86] of $H^-$ just above threshold in the presence of a strong electric field ($\approx 100$KV/cm), theoretical analyses [87, 88, 89] accounted for the principle observed effects: a finite cross-section at the zero-field detachment threshold of 0.75 eV, an exponential fall-off of the cross-section for lower photon energies due to detachment aided by tunneling through the static field's potential, and oscillations in the cross-section about the zero field value for energies above 0.75 eV. This last may either be viewed as the effect of the sloping static field potential on the outgoing $p$-wave ("Airy function oscillations") or as the interference between two pathways for the escaping electron, one directly into the escape direction and the other after reflection of an oppositely moving wave from the static field barrier. Such simple, and analytical, treatments of the effect of a static field on a free outgoing $p$-electron, with neglect of final state interactions

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between it and the parent H atom left behind, sufficed to give a complete and detailed accounting of the observed data \([87, 88]\). Treatment of such final state interactions has also been carried out subsequently \([90]\).

With the advent of intense lasers, there has also been interest in multi-photon detachment and in non-perturbative phenomena due to the dynamic electric field of the detaching laser \([91, 92]\). In particular, Gao and Starace \([91]\) re-investigated the problem through an exact solution for the outgoing electron in combined static and dynamic electric fields. When applied to \(H^-\), they claimed that a cross-term between the two fields leads to somewhat different results from previous studies even in the weak laser-field limit, the cross-section near the zero static-field detachment threshold being lowered. This is the question we address here through an alternative derivation of this exact solution that pays careful attention to its proper limiting behaviors. Our results do not support the claims of lowered cross-sections arising from a cross-term between the static and dynamic fields. On the other hand, our analysis suggests that caution is necessary in S-matrix formulations because results seem to depend on the choice of gauge for the electromagnetic potentials.

As in Ref. \([91]\), and adopting the same notation, we consider a uniform static electric field \(E_x = E_x z\) and the time-dependent electric field of the laser (effects of its magnetic field are as usual neglected as smaller) to give a total field

\[
E(t) = E_x z + E_0 \sin(\omega t).
\]

\[
= E_x z + (E_{0x} \hat{x} + E_{0y} \hat{y} + E_{0z} \hat{z}) \sin(\omega t). \quad (D.1)
\]
Describing the outgoing electron as moving in such a time-dependent field, and neglecting any residual interactions with the H atom left behind, the Schrödinger equation,

\[ i\dot{\psi}(\mathbf{r},t) = \left( \frac{\mathbf{p}^2}{2} + \mathbf{E}(t) \cdot \mathbf{r} \right) \psi(\mathbf{r},t). \]  

(D.2)

can be solved through separation in Cartesian coordinates. Throughout, an over-dot will denote differentiation with respect to \( t \) and we set \( \hbar = m_e = e = 1 \). Whereas Ref. [91] did so through a passage to momentum space, we develop our solutions in coordinate space through a technique of solving such time-dependent equations by operator algebra [78]. By working in coordinate space and directly with the electric field themselves, we avoid questions that arise in the momentum space formalism, particularly the gauge choice for the vector potential \( \mathbf{A} \).

For a general time-dependent equation,

\[ i\dot{\phi}(t) = [\dot{\mu}(t) A + \dot{\nu}(t) B] \phi(t). \]  

(D.3)

where \( A \) and \( B \) are possibly non-commuting operators not themselves explicitly dependent on time, the general solution can be developed in terms of an evolution operator, \( U(t,0) \), which is of the form of a product of exponentials, each involving \( A, B, \) and successive commutators of them, along with time-dependent functions \( \mu(t), \nu(t), \lambda(t), \delta(t),... \) which obey first order classical differential equations [78]. Each Cartesian component in (D.2) involves only the operators \( \mathbf{p}^2 \) and the linear coordinate so that apart from their commutator proportional to \( \mathbf{p} \), no further operators appear. The resulting solutions involve four exponential factors.

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For the $x$- and $y$- components, these solutions have been previously recorded [78] and are

$$
\psi(x, t) = \exp \left[ -i \frac{E_{0x}^2}{\omega^3} \left( \frac{1}{8} \sin(2\omega t) - \sin(\omega t) + \frac{3\omega t}{4} \right) \right] 
\times \exp \left[ -i \frac{E_{0x}}{\omega} x(1 - \cos(\omega t)) \right] 
\exp \left[ -i \frac{E_{0x}}{\omega^2} k_x (\sin(\omega t) - \omega t) \right] 
\times \exp \left[ ik_x x - i k_x^2 t \right] , \quad (D.4)
$$

with an exactly similar expression in $y$.

The solution in the $z$-coordinate can be similarly derived through the procedure in [78] to give

$$
\psi(z, t) = \exp \left[ -i \frac{E_{0z}^2}{\omega^3} \left( \frac{1}{8} \sin(2\omega t) - \sin(\omega t) + \frac{3\omega t}{4} \right) \right] 
+ \frac{E_{0z} E_z}{\omega^3} (\cos(\omega t) - 1 + \frac{\omega^2 t^2}{2}) 
\times \exp \left[ -i \frac{E_{0z}}{\omega} z(1 - \cos(\omega t)) \right] 
\exp \left[ -i \frac{E_{0z}}{\omega^2} p_z (\sin(\omega t) - \omega t) \right] 
\times \exp (-i\varepsilon_z t) \phi_A(z) ; \quad (D.5)
$$

where $\varepsilon_z$ is the eigenvalue and $\phi_A(z)$ the Airy eigenfunction [93] satisfying

$$
\left( \frac{p_z^2}{2} + E_z z \right) \phi_A(z) = \varepsilon_z \phi_A(z). \quad (D.6)
$$

These results in (D.4) and (D.5) parallel exactly the similar expressions in Ref. [91] with the one important difference that we have paid careful attention to the boundary condition at $t = 0$, that (D.4) reduce to plane waves in $x$ and $y$ with no extraneous phase factors and (D.5) to the Airy
stationary state with again each of the three exponentials in the top two lines of this equation reducing exactly to unity. Construction through the evolution operator ensures this [78]. In contrast, Ref. [91]'s solutions retain at \( t = 0 \), when the field vanishes, redundant phase factors because of their choice of the vector potential \( \mathbf{A} = \frac{e \mathbf{E}}{\omega} \cos(\omega t) \) to describe this electric field while working in momentum space. Indeed, were we to modify their results by replacing \( \cos(\omega t) \) above by \( (\cos(\omega t) - 1) \) in \( \mathbf{A} \), which still describes the same electric field, the two results would become identical. As we will see, this seemingly slight difference in the choice of a gauge has important consequences. In our formalism, however, we work directly with the electric field in (D.1).

The appearance in (D.4) and (D.5) of the characteristic combinations, \( \sin(\omega t) - \omega t \) and \( \cos(\omega t) - 1 + \omega^2 t^2 / 2 \), in place of the trigonometric functions alone as in Ref. [91] has another profound consequence. Our solutions remain well-behaved in the limit \( \omega \to 0 \) (which is closely related to \( t \to 0 \) because of the combination \( \omega t \)) whereas some of the similar terms in Ref. [91] and, in particular, the cross-term involving \( E_0 z E_s \) in (D.5), blow up in this limit. This is a persistent problem in the literature on intense dynamic fields [94], that several results seem not to admit passage to the static field limit as one should expect of them. It is worth emphasising that within the momentum space formulation as in Ref.[91], different choices of \( \mathbf{A} \), differing only in a constant which does not change the electric field, lead to different wave functions. In particular, these functions can differ drastically in the \( \omega \to 0 \) limit so that particular care may be necessary for considerations of
the static field limit. We also note that these gauge questions have to do with alternative wave functions for alternative vector potentials, not whether the dipole matrix element for photoabsorption uses length, velocity or acceleration forms.

To calculate photodetachment of $H^-$, we follow Ref. [91] in using (D.4) and (D.5) to describe the final state wave function along with a simple, much used "one-electron" representation of the ground state of $H^-$ [87, 88, 91, 95],

$$\psi_i(r, t) = \frac{B}{r} \exp(-kr) \exp(-i\xi t), \quad (D.7)$$

where $k = (-2\xi)^{1/2}$, $\xi = -0.75\text{eV}$ being the energy of this ground state, and $B$ is a normalization parameter, equal to 0.31552 in atomic units [88, 91]. This wave function has long [95] proved very successful in describing photodetachment for the energy range of interest and has been employed in all the past work that we compare with. The S-matrix element is given as in Eq. (27) of Ref. [91] by

$$S_{fi} = (2\pi)^{-1/2} B \int_{-\infty}^{\infty} dt \int dr \psi^*(x, t) \psi^*(y, t) \psi^*(z, t) \exp(-i\xi t), \quad (D.8)$$

with the wave functions drawn from (D.4) and (D.5). Examining next the weak laser-field limit, we expand the factors $\psi^*$ to first order in $E_0$, and retain terms proportional to it to get

$$S_{fi} = \frac{i^{2/3} B E_0 E_s^{1/6}}{\omega^2} \left[ A\prime(-\xi) \int_{-\infty}^{\infty} dt (\sin \omega t - \omega t) \exp[i(\xi - \xi_i)t] \right]$$

$$= -i \frac{(E_s/2)^{1/3}}{\omega} A(-\xi) \int_{-\infty}^{\infty} dt (\cos \omega t - 1 + \frac{1}{2}\omega^2 t^2)$$

$$\times \exp[i(\xi - \xi_i)t], \quad (D.9)$$
where $\varepsilon_f = \frac{1}{2}(k_x^2 + k_y^2) + \varepsilon_z$ and we have defined a dimensionless energy $\xi = \varepsilon_z(2/E_z^2)^{1/3}$. The derivative in the first term, denoted by a prime, is with respect to $\xi$, and arises from the $E_{oz}p_z$ operator in (D.5).

The above result in (D.9) parallels exactly the one in Ref. [91], again with the replacement of $\sin \omega t$ and $\cos \omega t$ by the forms that vanish up through terms of order $\omega^2 t^2$. Before turning to the time integrations, the structure of (D.9) and the origin of its second term in the curly brackets already points to problems with the claim in Ref. [91] that this is a new contribution not present in earlier treatments. Whereas the first term in (D.9) arises from the expansion of the $E_{oz}p_z$ in (D.5), thus carrying the dipole operator which leads to the transition element $\langle \psi | E_{oz}p_z | \psi_i \rangle$, the second term has its origins in the cross-term $E_{oz}E_z$ of (D.5). But this is purely a phase with no involvement of atomic operators, and therefore incapable of causing transitions.

Further confirmation of this conclusion that there is no cross-term proportional to $E_{oz}E_z$ is provided by carrying out the time integrations. In Ref. [91], these integrations were immediate, upon combining the $\exp(-i\omega t)$ piece of the sine and cosine with the other exponential in the integrand to give, upon integration, $2\pi \delta(\varepsilon_f - \varepsilon_i - \omega)$. With the transition probability $W_{fi}$ defined as

$$\lim_{t \to -\infty} \frac{|S_{fi}|^2}{t} = 2\pi W_{fi} \delta(\varepsilon_f - \varepsilon_i - \omega), \quad (D.10)$$

the cross-section

$$\sigma = \frac{8\pi \omega}{cE_0^2} \int W_{fi} dk_x dk_y d\varepsilon_z, \quad (D.11)$$

was then evaluated.
We now handle these operations by evaluating the integrals in (D.9) between limits $-T$ and $T$ analytically and computing $\sigma$, finally taking the $T \to \infty$ limit numerically. In the first part of this process, the analytical integration from $-T$ to $T$, we retain only the contributions which lead to $\delta(\epsilon_f - \epsilon_i - \omega)$ in the limit $T \to \infty$ as the only ones that correspond to absorption of a single photon. As $T$ increases beyond a few atomic units, our calculated photo-detachment cross-section converges rapidly. The second term in (D.9) does not contribute in accordance with our discussion above and, as illustrated in Fig. D.1, our results reproduce exactly those of earlier work [87, 88] even for static field strengths exceeding 1MV/cm. All these results coincide as shown by the solid curve. Only if both terms in the curly bracket in (D.9) are retained, while simultaneously dropping the terms in $-\omega t$ and $-1 + \frac{1}{2}\omega^2 t^2$, do we recover the results of [91] as shown by the dashed curve. We conclude, therefore, that a proper treatment leads to no reduction in the cross-section around the zero static-field threshold as claimed in Ref. [91]. The handling of time integrations with the $-\omega t$ and the $-1 + \frac{1}{2}\omega^2 t^2$ terms would also confront a calculation such as the one in Ref. [91] had this alternative gauge been used for $A$. The S-matrix integrations in (D.9) would then not simply reduce to delta functions but also involve the derivatives that we have encountered.

In view of the discrepancy with the previous results in [91], we offer the following discussion. In the spirit of the S-matrix formulation, the electric field in (D.1) is assumed to be switched on starting at $t = -\infty$ and switched off at $t = \infty$ adiabatically, although this is not explicitly implemented in
Figure D.1: Laser photodetachment of $H^-$ in a static electric field. Photodetachment of $H^-$ in combined static (1 MV/cm) and detaching laser's electric fields. Solid line: Our results, dashed line: Ref. [91] Earlier calculations of Refs. [87] and [88] are essentially indistinguishable from the solid line.
carrying out the time integrations, just as in Ref. [91]. Therefore, our cal-
culations follow exactly the same procedure as did Ref. [91] except that our
final state wave functions in (D.4) and (D.5) differ from those in Ref. [91] as
pointed out above. We were led to these additional terms involving $-\omega t$ and
$-1+\omega^2 t^2$ by our emphasis on the proper limiting behavior as $t \to 0$ or $\omega \to 0$.
Therefore, it may be argued that Ref. [91] and this paper deal with different
problems, differing in when the electric fields are turned on. But, as we have
pointed out, these additional terms may also be viewed as arising from the
two different gauge choices for $A$ in Ref. [91], namely $(cE_0/\omega)(\cos \omega t - 1)$,
with or without that $-1$. As shown in Fig. D.1, the difference between the
dashed and solid lines can be attributed entirely to this difference, which in
itself poses the question of gauge invariance of the cross-sections presented
in Ref. [91].

Part of these questions of the switching on and off of the electric fields
can be settled by developing explicit solutions of (D.1) and (D.2) with some
specific form of $E(t)$ which vanishes smoothly as $|t| \to \infty$, but we also dis-
agree with Ref. [91] in other regards. Most importantly, it seems to us
that notwithstanding any multiple order of interactions with the static field
$E_s$, one photon absorption in the limit of weak laser fields must involve an
amplitude proportional to $E_0$, along with the matrix element of a dipole op-
erator, whether $\mathbf{r}$ or $\mathbf{p}$, and a corresponding energy-conserving delta function
$\delta(\varepsilon_f - \varepsilon_i - \omega)$. Therefore, such one photon transitions can only be attributed
to terms which have such a structure upon expanding exponentials in (D.4)
and (D.5) to first order in $E_0$. The $E_0E_s$ term of [91] does not satisfy these
requirements. Likewise, in a related argument which may help clarify the points of disagreement, were we to seek two-photon transitions by expanding the exponentials to order $E_0^2$, we would not expect any contribution from the first terms involving $E_0^2 \sin 2\omega t$ in (D.4) and (D.5), because they contain no atomic operators. In this, we would differ from Ref. [91] and other such treatments whose Floquet expansions get a contribution from these terms, the so-called "ponderomotive potential" then appearing in their resulting energy-conserving delta function for such two (or multiple) - photon transitions. Since this ponderomotive potential depends on the continuous variable $E_0^2/\omega^2$, it need not be an integer multiple of $\omega$, placing it in conflict with the photon picture of multi-photon absorption. We, on the other hand, would face no such conflict. See also Ref. [96].

Finally, after completion of our work, we have seen a recent paper Ref. [97] on the one- and two-photon photodetachment of $H^-$ in combined static and dynamic fields, taking into account final state interactions of the electron with the residual H atom. As already stated, our study was not concerned with this interaction. But we note that Ref. [97]'s treatment of such an interaction leads also to contributions similar in structure to the "cross-term" in $E_0^2 E_2$, namely, to a term proportional to $Ai(-\xi)$ in (9); see (72) of Ref. [97]. In disentangling the two effects, of the cross-term and the electron-atom final state interaction, and in coming to the conclusion that the latter is small, Ref. [97] has compared with the previously calculated effects of the cross-term in Ref. [91]. In view of our questioning of any depression of the cross-section around the zero-field detachment threshold from the cross-term, the
conclusion of Ref. [97] on the unimportance of the electron-atom final state interaction will have to be revisited.

We have also seen a recent paper Ref. [98] on $H^{-}$ photodetachment in a static electric field and a pulsed laser field. They consider quantum and semi-classical approaches different from both ours and Ref. [91]. These authors also note in a footnote to their equation (6) that only a term in the derivative of but not in $Ai(-\xi)$ contributes, in agreement with our conclusions.

We acknowledge useful discussions with Drs. A.F. Starace and K. Unnikrishnan.
APPENDIX E: THE DISCRETIZED $Z$-MOMENTUM OPERATOR

In the $r - \ell$ basis, the wave function has a form

$$|\Psi\rangle = \sum_{\ell=0}^{\ell_{\text{max}}} \phi_{\ell}(r)|\ell\rangle. \quad (E.1)$$

The $p_z$ operator acts on this wave function as

$$p_z|\Psi\rangle = -i \frac{\partial}{\partial z} \sum_{\ell=0}^{\ell_{\text{max}}} \phi_{\ell}(r)|\ell\rangle. \quad (E.2)$$

The expression for $\frac{\partial}{\partial z}[f(r)Y_{\ell m}(\theta, \phi)]$ is well known [99] and is used to write

$$\frac{\partial}{\partial z}[\phi(r)|\ell\rangle] = a_{\ell} \left( \frac{\partial}{\partial r} \phi - \frac{\ell}{r} \phi \right) |\ell + 1\rangle + a_{\ell-1} \left( \frac{\partial}{\partial r} \phi + \frac{\ell + 1}{r} \phi \right) |\ell - 1\rangle; \quad (E.3)$$

where

$$a_{\ell} = \frac{\ell + 1}{\sqrt{(2\ell + 1)(2\ell + 3)}}. \quad (E.4)$$

To discretize this operator on the non-uniform radial grid, we adopt the same method as described in Appendix C. That is, we discretize the expression

$$L = \sum_{\ell} \int r^2 dr a_{\ell-1} \phi_{\ell}^* \left( \frac{\partial}{\partial r} \phi_{\ell-1} - \frac{\ell - 1}{r} \phi_{\ell-1} \right)$$

$$- a_{\ell} \left( \frac{\partial}{\partial r} \phi_{\ell} + \frac{\ell}{r} \phi_{\ell} \right) \phi_{\ell+1}. \quad (E.5)$$

By using the same discretization scheme as before, we have

$$L_{\ell} = a_{\ell-1} \left[ \frac{(\phi_{\ell+1}^j + \phi_{\ell}^i)^*}{2} (\phi_{\ell-1}^j - \phi_{\ell-1}^i) \left( \frac{r_{j+1} + r_{j}}{2} \right)^2 \right]$$

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Collecting the terms with $\phi^j_\ell^*$ and defining the function $g^j_\ell$ as before,

$$\mathcal{L} = \sum_\ell (g^j_\ell)^* \left\{ a_{\ell-1} \left[ g^{j+1}_{\ell-1} \frac{1}{r_j + r_{j+1}} \sqrt{(r_{j+2} - r_j)(r_{j+1} - r_j)} \left( \frac{r_{j+1} + r_j}{2} \right)^2 - g^{j-1}_{\ell-1} \frac{1}{r_j - r_{j-1}} \sqrt{(r_{j+1} - r_j)(r_{j+1} - r_{j-1})} \left( \frac{r_{j+1} + r_j}{2} \right)^2 \right] \right. \\
+ a_{\ell} \left[ g^{j+1}_{\ell+1} \frac{1}{r_j + r_{j+1}} \sqrt{(r_{j+2} - r_j)(r_{j+1} - r_j)} \left( \frac{r_{j+1} + r_j}{2} \right)^2 + g^{j-1}_{\ell+1} \frac{1}{r_j - r_{j-1}} \sqrt{(r_{j+1} - r_j)(r_{j+1} - r_{j-1})} \left( \frac{r_{j+1} + r_j}{2} \right)^2 \right] \right\}.$$  

(E.6)

This expression is further simplified by using

$$\left( \frac{r_{j+1} + r_j}{2} \right)^2 - \left( \frac{r_j + r_{j-1}}{2} \right)^2 = \frac{(r_{j+1} - r_{j-1})}{4}$$

times $[r_{j+1} + r_j - 1 + 2r_j]$;  

(E.8)

$$\frac{r_{j+1} + 2r_j + r_{j-1}}{4r_j} = \frac{(r_{j+1} - r_j) - (r_j - r_{j-1})}{4r_j} + 1.$$  

(E.9)

Applying the Euler-Lagrange condition we get

$$\left( \frac{\partial}{\partial z} g^j_\ell \right)_\ell = a_{\ell-1} \left[ \alpha_j g^{j+1}_{\ell-1} - \frac{\delta_j + \beta_{\ell-1}}{r_j} g^j_{\ell-1} - \alpha_j g^{j-1}_{\ell-1} \right] + a_{\ell} \left[ \alpha_j g^{j+1}_{\ell+1} - \frac{\delta_j + \beta_{\ell}}{r_j} g^j_{\ell+1} - \alpha_j g^{j-1}_{\ell+1} \right];$$  

(E.10)

where,

$$\alpha = \frac{1}{\sqrt{(r_{j+2} - r_j)(r_{j+1} - r_{j-1})}} \frac{1}{r_j r_{j+1}} \left( \frac{r_{j+1} + r_j}{2} \right)^2,$$  

(E.11)
\[ \delta = \frac{(r_{j+1} - r_j) - (r_j - r_{j-1})}{4r_j} , \]  

\[ \beta_\ell = \ell + 1. \]  

(E.12)  

(E.13)

In the limit of a uniform grid, \( \delta \to 0 \) and the equation reduces to that obtained by the discretization on a uniform grid.

Thus, the operator \( p_z \) acting on a wave function in the \( r - \ell \) basis on a non-uniform radial grid yields a three dimensional matrix which is symmetric and tridiagonal in both the \( r \) and the \( \ell \) indices.
APPENDIX F: THE Z-TRANSLATION OPERATOR

The operator $e^{i\Delta z p_z}$ produces a translation of $\Delta z$ in the $z$-direction. To see the effect of this operation on the wave function in the spherical basis

$$e^{i\Delta z p_z} \sum_\ell \phi(r)|\ell\rangle,$$

we study the form of the $p_z$ operator as described in the last equation of Appendix E. Since the matrix form of $p_z$ itself cannot be diagonalized, we write it as a sum of two parts,

$$p_z = R + \Theta.$$  \hspace{1cm} (F.2)

$$e^{i\Delta z p_z} = e^{i\Delta z R/2} e^{i\Delta z \Theta/2} e^{i\Delta z R/2}.$$ \hspace{1cm} (F.3)

up to terms which are correct through second-order in $\Delta z$ because $[R, \Theta] \neq 0$.

The $R$ and $\Theta$ parts are both tridiagonal matrices which may be written as a sum of two block diagonal matrices consisting of $(2 \times 2)$ blocks [80].

$$R = R_e + R_o.$$ \hspace{1cm} (F.4)

$$\Theta = \Theta_e + \Theta_o.$$ \hspace{1cm} (F.5)

$$e^{i\Delta z R/2} \approx e^{i\Delta z R_e/2} e^{i\Delta z R_o/2}.$$ \hspace{1cm} (F.6)

$$e^{i\Delta z \Theta/2} \approx e^{i\Delta z \Theta_e/2} e^{i\Delta z \Theta_o/2}.$$ \hspace{1cm} (F.7)

The $R$ part consists of those terms with coefficients $\alpha$ as defined in the Appendix E, and the other terms form the $\Theta$ part. The matrix $R$ has a form
The matrix $\Theta$ also has a very similar structure. The $2 \times 2$ matrices can be diagonalized easily. So to make a translation of $\Delta z$ using the $R$ matrix, $e^{i\Delta z R}$, the wave function is transformed to the basis in which the $\Theta$ components are diagonal and then for each $\ell$ value, it is operated upon by the operators $e^{i\Delta z R_\epsilon}$ and $e^{i\Delta z R_\delta}$, then transformed back to the original $r - \ell$ basis. This operation is repeated with the $\Theta$ part in the appropriate basis.

$\Delta z$ is chosen to be small so that the norm is conserved. We ensure that the operator $e^{i\Delta z p_z}$ changes $\langle z \rangle$ by $\Delta z$. This algorithm is stable. The complexity of the algorithm is of the order of $(N_r \times N_\ell)$ for each $z$-step.
February 11, 2000

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VITA

Chitra Rangan is a native of Chennai (formerly Madras), India. The first child of Mr. and Mrs. T.P. Rangan, she grew up in an environment where education is valued. As a tenth-grader, she won the prestigious National Talent Search Scholarship in 1986. She graduated with a bachelor's degree in physics from the University of Madras, India, in 1991, with university medals in English and French.

She graduated with a master's degree in physics from the Indian Institute of Technology, Madras, in 1993. Her master's thesis was in the area of quantum optics where she researched the noiseless, undistorted propagation of an electromagnetic pulse (squeezed soliton) through an optical fiber. She won three fellowships for graduate study, of which she accepted a Junior Research Fellowship at the Indian Institute of Astrophysics, Bangalore.

Since 1994, she has been pursuing doctoral study in atomic, molecular and optical physics at the Department of Physics and Astronomy at Louisiana State University. Her research has been presented at the DAMOP (APS) meeting (1998), APS Centennial meeting (1999) and the Gordon Research Conference in Atomic Physics (1999). She and her engineer husband Narayan live in Michigan. She plans to pursue an academic career in physics after graduating in May 2000.

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