

**IONIZATION, PHOTOELECTRON DYNAMICS AND ELASTIC
SCATTERING IN RELATIVISTIC, ULTRA-STRONG FIELD**

by

Sui Luo

A dissertation submitted to the Faculty of the University of Delaware in partial fulfillment of the requirements for the degree of Doctor of Philosophy in Physics

Fall 2015

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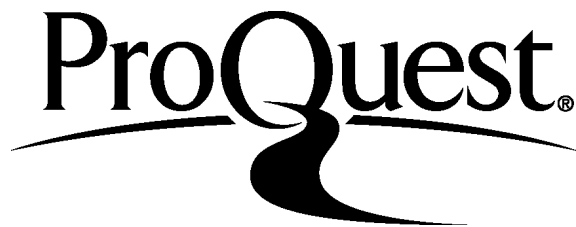
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SCATTERING IN RELATIVISTIC, ULTRA-STRONG FIELD**

by

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ABSTRACT

Ultrastrong laser-matter interaction has direct bearing to next generation technologies including plasma acceleration, laser fusion and attosecond X-ray generation. The commonly known physics in strong field becomes different as one progress to ultrastrong field. The works presented in this dissertation theoretically study the influence of relativistic effect and magnetic component of the laser field on the ionization, photoelectron dynamics and elastic scattering processes.

The influence of magnetic component ($\mathbf{B}_{\text{laser}}$) of circularly polarized (CP) ultrastrong fields (up to $3 \times 10^{22} \text{ W/cm}^2$) on atomic bound state dynamics is investigated. The Poincaré plots are used to find the changes in trajectory energies are on the order of a few percent for intensities up to $1 \times 10^{22} \text{ W/cm}^2$. It is found that at intensities where ionization approaches 50% for the bound state, the small changes from $\mathbf{B}_{\text{laser}}$ of the circular polarized light can actually result in a several-fold decrease in ionization probability. The force on the bound electron exerted by the Lorentz force from $\mathbf{B}_{\text{laser}}$ is perpendicular to the rotating plane of the circular polarized light, and this nature makes those trajectories which are aligned away from the minimum in the potential barrier stabilized against tunneling ionization. Our results provide a classical understanding for ionization in ultrastrong fields and indicate that relativistic effects in ultrastrong field ionization may most easily be seen with CP fields.

The photoelectron energy spectra from elastic rescattering in ultrastrong laser fields (up to $2 \times 10^{19} \text{ W/cm}^2$) is studied by using a relativistic adaption of a semi-classical three-step recollision model. The Hartree-Fock scattering potentials are used in calculating the elastic rescattering for both hydrogenlike and noble gas species. It is found that there is a reduction in elastic rescattering for intensities beyond $6 \times 10^{16} \text{ W/cm}^2$ when the laser Lorentz deflection of the photoelectron exceeds its wavefunction spread. A relativistic rescattering enhancement occurs at $2 \times 10^{18} \text{ W/cm}^2$, commensurate with relativistic motion of a classical electron in a single field cycle. The good comparison between the results with available experiments suggests the theory approach is well suited to modeling scattering in the ultrastrong intensity regime.

We investigate the elastic scattering process as it changes from strong to ultrastrong fields with the photoelectron angular distributions from Ne, Ar, and Xe. Noble gas species with Hartree-Fock scattering potentials show a reduction in elastic rescattering with the increasing energy of ultrastrong fields. It is found that as one increases the returning photoelectron energy, rescattering becomes the dominating mechanism behind the yield distribution as the emission angle for all the species extends from 0° to 90° . The relativistic effects and the magnetic field do not change the angular distribution until one is well into the $\Gamma_r \gg 1$ regime where the Lorentz defection significantly reduces the yield. As we proceed to the highest energy, the angular emission range narrows as the mechanism changes over to backscattering into narrow angles along the electric field.

Chapter 1

INTRODUCTION

1.1 Motivation

Light-matter interaction has been a continuously active research area in fundamental science in the past 30 years. Its research has direct bearing to topics fundamental to atomic, optical, molecular and plasma physics. A range of studies, both theoretical and experimental, have been conducted on the subjects of multiphoton ionization [1, 2], tunneling ionization [3-6], high-harmonic generation [7-10], and rescattering [11-13]. Strong field ionization and rescattering has been used to measure electron dynamics [14], collisionally excite multiple electrons [15], and perform molecular tomography [16]. The research results have also provided enormous inspirations for research in other disciplines including chemistry, biology and engineering.

Ever since its invention in 1960, the laser has been the driving force behind the light-matter interaction research. With the development of ultrastrong (with intensities above 10^{19} W/cm²) and ultrashort (with pulse duration only a few tens of femtoseconds, 10^{-15} s) lasers, a vast number of next generation practically applications become possible. Among them, three highlighted applications are plasma acceleration [17, 18], laser fusion [19, 20] and attosecond X-ray radiation generation [21, 22]. Plasma accelerators use ultrastrong laser fields to accelerate particles to high energies (in the order of GeV) within a distance much shorter than the conventional accelerators. It is the fundamental technology for future innovation of affordable and

compact accelerators which can be used in various applications, ranging from high energy physics to medical diagnostics. On the other hand, laser fusion tries to replicate the fusion process happening in the sun by using ultrastrong lasers to squeeze the hydrogen atoms and turn them into helium, a process which promises the ultimate solution to the earth's energy crisis. Finally, the attosecond X-ray radiation enables scientists to visualize reaction pathways and dynamics on the molecular level which used be non-observable with traditional radiation sources. All the technologies are deemed to yield far-reaching impact on our life in the future.

The successes of these hopeful technologies require in-depth and comprehensive understanding of laser-matter interaction in the ultrastrong field context. As one progress from strong field to ultrastrong field, the commonly known physics becomes different and new phenomena emerge. The once widely used plane-wave approximation and dipole approximation need to be examined for their validities at the new regime. The speed of the electron in the ultraintense laser field becomes comparable to the speed of the light and electron relativistic dynamics need to be studied. As the electric field becomes even larger than the atomic Coulomb field, the once neglected magnetic component becomes significant and its influences on the atomic ionization, electron dynamics and rescattering process need to be investigated. It is also the purpose of the works presented in this dissertation to understand all these interesting, fundamental, yet complicated questions. Specifically, we accomplish this goal by focusing on the theory effort to model the ionization, relativistic dynamics, and rescattering in ultraintense field, and use computer cluster to conduct trajectory ensemble simulation.

1.2 Ionization in Ultrastrong Field

Our understanding of ionization has experienced three phases historically as the intensity of light sources increases.

The first phase is the observation of photoelectric effect, where metals emit photoelectrons when light shines upon them [23]. The theory by Albert Einstein in 1905 well explained the observation by quantizing the energy carried in the light. Specifically, the energy carried by a single photon is $\hbar\omega$, where ω is the frequency of the light. When the photon energy is larger than the minimum energy required to remove a delocalized electron from the surface of the metal, ϕ , photoelectric effect happens. And the kinetic energy of the freed electron in the continuum is calculated with $K = \hbar\omega - \phi$. Photoelectric effect essentially resembles the single photon ionization process (Figure 1.1(a)) in the context of light-gas interaction, where an electron absorbs a photon with energy equal or larger than the ionization potential of the atom. The energy scale is typically in the order of tens of eV and with light intensities typically in the order of 10^{12} W/cm² or lower.

The second phase happened in the 1960's and 1970's. The development of Q-switching [24] and mode locking technologies pushed the maximum intensity of the laser up to 10^{14} W/cm², which led to the discovery of multiphoton ionization (Figure 1.1(b)) [1, 2]. In the multiphoton ionization process, a bound state electron is ionized by simultaneously absorbing N photons with the total energy ($N\hbar\omega$) larger than its ionization potential (IP). At these intensities ($10^{12} \sim 10^{14}$ W/cm²) the electric field potential can be safely treated as a perturbation to the original Hamiltonian and the lowest-order perturbation theory (LOPT) was employed to accurately model this process [25, 26]. The n -photon ionization rate is calculated as $\Gamma_n = \sigma_n I^n$, where n is the minimum number of photons needed for ionization, σ_n is the generalized cross

section and I is the laser intensity. As the intensity gets stronger, a phenomenon known as above-threshold ionization (ATI) is observed within the context of multiphoton ionization, where the number of absorbed photons is larger than the minimum number of photons required for ionization [27, 28]. The fact that the oscillation energy of electron driven by the electric field is now larger than the photon energy suggests the nature of ATI is non-perturbative. ATI thus marks the division between the perturbative and non-perturbative regime.

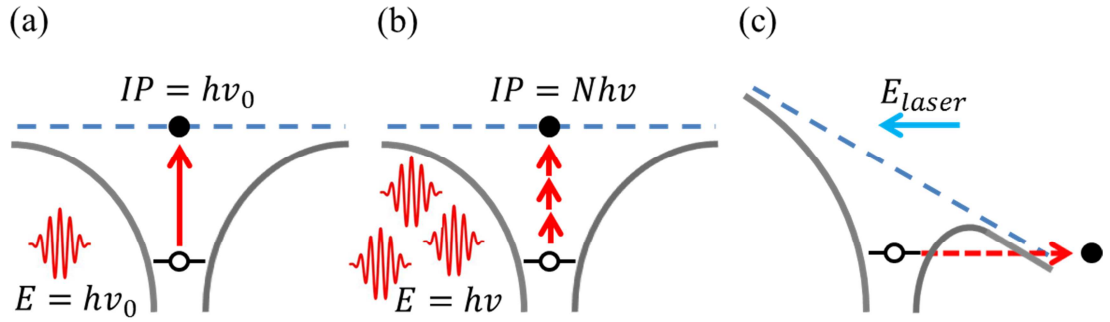


Figure 1.1 Schematic diagram for ionization mechanisms, (a) single photon ionization (photoelectric effect), (b) multiphoton ionization and (c) tunneling ionization.

In the third phase, the invention of chirped pulse amplification (CPA) [29, 30] in the 1980's enables the generation of laser with intensity up to the order of 10^{19} W/cm² and with pulse duration down to the order of femtosecond (10^{-15} s). A new mechanism known as tunneling ionization (Figure 1.1(c)) becomes possible [3, 4]. In both single photon ionization and multiphoton ionization mechanisms, the incoming light is treated as a flux of photons. But in tunneling ionization, the light is considered as electromagnetic field. At these intensities, the original Coulomb potential is

significantly modified by the electric field of the laser (the electric field experienced by atoms in ultrastrong field has a typical value of 10^{11} V/cm, comparing to the Coulomb binding field of atomic hydrogen 5×10^9 V/cm). The effective potential can now be written as $V_{\text{eff}} = -Z/r + \vec{E} \cdot \vec{r}$, where Z is the screened charge of the nucleus, r is the radial distance from the nucleus and $\vec{E}$ is the electric field of laser. This drastic distortion in the potential enables the bound state electron to tunnel through the suppressed potential barrier and be ionized. Perturbation theory is no longer appropriate here as the external electric field is comparable to the Coulomb field and one semi-classical theory developed by Ammosov, Delone, and Krainov (ADK) [31] has been particularly successful in calculating the tunneling ionization rate (see section 2.1).

While the ionization mechanism can be straightforwardly identified using the intensity of the laser, the Keldysh parameter [11] has been widely adopted to quantify the transition from multiphoton ionization to tunneling ionization. The Keldysh parameter (γ) is defined as the ratio of the laser frequency (ω) to the tunneling frequency (ω_T), with the tunneling frequency defined as $\omega_T = E_{\text{laser}}/\sqrt{2IP}$, where IP is the ionization potential energy and E_{laser} is the magnitude of electric field. Then the Keldysh parameter can be expressed as $\gamma = \omega\sqrt{2IP}/E_{\text{laser}}$. When $\gamma \gg 1$, the time scale of the ionization is much larger than the period of optical cycle and the ionization is considered as multiphoton ionization. For $\gamma \ll 1$, the ionization finishes within a single optical cycle and the ionization is classified as tunneling ionization.

All of the works presented in this dissertation are in ultrastrong intensity regime and the laser field in study is adiabatic ($\gamma \ll 1$) thus the ionization mechanism is tunneling ionization.

1.3 Rescattering in Ultrastrong Field

The dynamics of the electron in the continuum after being tunneling ionized are governed by its interaction with the laser electromagnetic field. The periodic change of the electric field can drive the ionized electron back to the parent ion when the tunneling ionization happens within certain phase range of the laser field, a process known as recollision [11-13, 31]. Depending on the energy of the returning electron and the atomic structure of the parent ion, several mechanisms can be triggered during the recollision. Elastic scattering can happen where the electron recollides with the parent ion and then is scattered back into the continuum [32, 33]. The electron might undergo inelastic rescattering with the parent ion which subsequently leads to ionization and excitation [32]. It is also possible that the returning electron is captured by the parent ion and its kinetic energy is released in the form of a photon, a process known as high harmonic generation (HHG) [34]. Figure 1.2 shows the two steps process of the inelastic rescattering ionization mechanism. More details about the rescattering dynamics can be found in section 2.5.

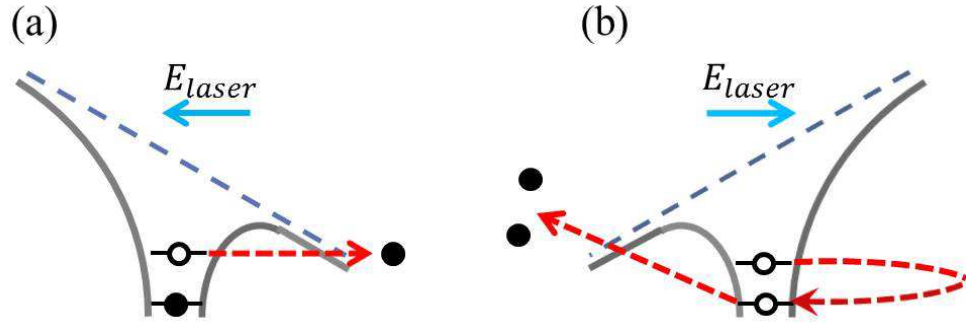


Figure 1.2 Schematic diagram for inelastic rescattering ionization mechanism, (a) first step tunneling ionization and (b) second step recollision. Notice the change of the direction of the electric field.

The rescattering process has been proved to be an important part to the laser-matter interaction physics. B. Walker *et al* [33] first reported the experimental observation of nonsequential ionization (NSI) in He^+ and He^{2+} production. As shown in figure 1.3, He^{2+} is experimentally seen with a significantly large amount of yield even before the saturation of He^+ , creating the ‘knee structure’ in the ion yields plot.

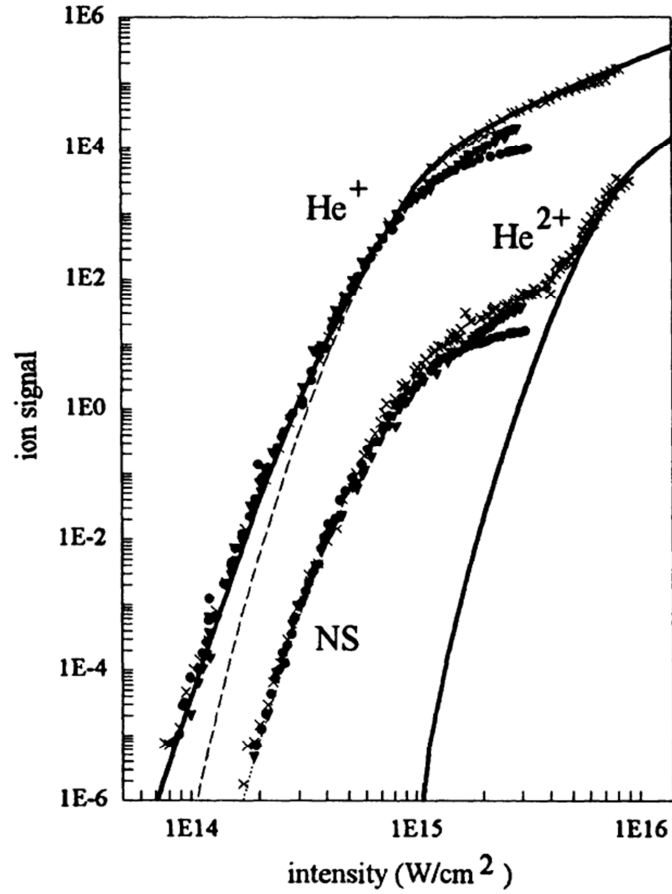


Figure 1.3 Measured He ion yields with the linear polarized laser field of 780 nm and 100 fs. The solid lines are the calculation results of the sequential tunneling ionization. From B. Walker *et al.* Phys. Rev. Lett. **73**, 1227 (1994).

Rescattering is proposed as the mechanism behind the NSI. It explains that even at intensities below the saturation intensity of the first electron (the intensity is not strong enough to tunnel ionize the second electron), the second electron can still be ionized through the recollision between the freed first electron and the parent ion. Recently, the rescattering mechanism has been successfully used to interpret the NSI observed with other multielectron systems [35-37]. It is worthy to note that significant NSI yield requires that the deviation of the returning electron wavepacket respect to the ion parent is small, thus the rescattering process is largely suppressed in the circularly polarized field as observed in [38].

1.4 Dissertation Outline

The intensity (10^{19} W/cm²) in the ultrastrong regime is twenty orders of magnitude larger than the intensity (0.1 W/cm²) of the sunlight on the earth's surface [39, 40]. It's a new regime, approximations that were made in the moderate-strong regime start to break down and interesting yet complicated phenomena not seen before show up. It is our special interest to examine the roles of relativistic effects and that of the magnetic field in the ultraintense laser-matter interaction, including the ionization, dynamics in the continuum and the rescattering process.

Chapter 1 provides a review of the history of understanding the ionization mechanism as the development of laser technology pushes the intensity into the ultrastrong regime. The tunneling ionization, which is the primary tunneling mechanism in this dissertation, and the rescattering process which is another important subject, are introduced as the background.

Chapter 2 breaks down the laser-matter interaction in the ultraintense regime to several major components and provides detailed discussion for each of them.

Specifically, the ADK model to calculate the tunneling ionization rate and the rate equations to calculate the sequential ionization yield are described. Both the beam profile and intensity profile of the laser beam are introduced. The full treatment of the electromagnetic field is described and its significance in the electron dynamics is studied. The three-step model of the relativistic rescattering is discussed with thorough explanations of the exit of the Coulomb Barrier, electron wavepacket dynamics and both elastic and inelastic scattering cross section. Chapter 2 prepares the reader with the necessary theory and methodology to understand the studies presented in the following chapters.

Our first interest is to understand the role of magnetic component of the external field in the bound state ionization in ultrastrong field in chapter 3. Most models, including the ADK model for calculating the tunneling ionization rate, assume the dipole-approximation where the magnetic field is neglected. Studies have been carried out to examine the validity of the dipole-approximation in linearly polarized ultrastrong fields [41], and we would like to extend this study to circularly polarized ultrastrong fields. The classical model is adopted here due to its success in understanding the role of magnetic fields for Rydberg atoms in strong fields [42]. By studying the Poincaré plots and Fourier transform of the trajectory ensembles, it is found that for intensities up to $1 \times 10^{19} \text{ W/cm}^2$ the changes in the trajectory energies are only on the order of a few percent. But the changes from the magnetic component of the field with circularly polarized light can result in a several-fold decrease in the ionization probability when intensities are larger than $1 \times 10^{20} \text{ W/cm}^2$. Our results provide a classical understanding of the relativistic effects in ultrastrong field ionization in circularly polarized field.

Elastic rescattering is another process where the presence of magnetic fields needs to be carefully investigated. Pioneer studies have been carried out to understand the energy transfer from the external field to atomic and ionic systems through the elastic scattering process [22]. In chapter 4, we report photoionization and fully relativistic elastic scattering in ultrastrong fields up to $1 \times 10^{19} \text{ W/cm}^2$. The influence of the relativistic effects and magnetic field on the elastic rescattering is studied with the final photoelectron energy spectrum. Both hydrogenlike and noble gas species with Hartree-Fock scattering potentials show a reduction in elastic rescattering beyond $6 \times 10^{16} \text{ W/cm}^2$ as the Lorentz deflection exceeds the electron wavepacket spread and the elastic rescattering yield deviates from the $1/I^2$ scaling. The calculated noble gas energy spectrum is compared with experimental result and it is found that our three-step model is well suited to modeling scattering in the relativistic, ultrastrong intensity regime.

In chapter 5, we investigate the elastic scattering process as it changes from strong to ultrastrong fields with the photoelectron angular distributions from Ne, Ar, and Xe. It is found that as one increases the returning photoelectron energy, rescattering becomes the dominating mechanism behind the yield distribution as the emission angle for all the species extends from 0° to 90° . The relativistic effects and the magnetic field do not change the angular distribution until one is well into the $\Gamma_r \gg 1$ regime where the Lorentz deflection significantly reduces the yield. As we proceed to the ultrastrong regime, the angular emission range narrows as the mechanism changes over to backscattering into narrow angles along the electric field.

In chapter 6, we investigate the returning fluence and inelastic impact ionization in strong to ultrastrong fields. The studies here address issues related to

wavelength dependence and the matching between returning fluence and cross section on rescattering energies. The ultimate cutoff in the rescattering flux is observed in relativistic regime. This work serves as guidance for experimentalists in searching for detectable inelastic impact ionization yield.

Finally, we conclude the dissertation in chapter 7 with the summary of the major accomplishments and the significance of our research and future research directions.

The detailed source codes are provided in the appendix for reference.

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Chapter 2

SEMI-CLASSICAL THEORETICAL MODELS

2.1 Tunneling Ionization Rate

Landau and Lifshitz [1] first derived the tunneling ionization rate for the ground state of hydrogen atom in a static electric field. By separating the Schrödinger Equation and solving for the asymptotic wave function, the tunneling rate for hydrogen-like ions can be expressed as

$$w_{LL} = 4 \frac{(2|\mathcal{E}_0|)^{5/2}}{E_{\text{laser}}} \exp\left(-\frac{2(2|\mathcal{E}_0|)^{3/2}}{3E_{\text{laser}}}\right) \quad (2.1)$$

where E_{laser} is the electric field of the laser, and $\mathcal{E}_0 = -Z^2/2$ is the ground state ionization potential. This result was then generalized to any asymptotic Coulomb wave function by Smirnov and Chibisov [2]. Later Perelomov, Popov and Terent'ev further generalized the ionization rate to low frequency electromagnetic fields by taking the appropriate time average of the oscillating field [3]. Based on that, Ammosov, Delone and Krainov (ADK) [4] finally obtained a general analytical expression in terms of the effective quantum numbers by considering the semiclassical solutions to the radial wave equation, which enables one to calculate the tunneling ionization rate for complex atoms or atomic ions in arbitrary states in a quasi-static oscillating field. The approximations made in the derivations of the ADK model implicitly requires that the width of the deformed Coulomb potential barrier does not change during the tunneling. In other words, the tunneling time should be much

smaller than the period of the oscillating field. Thus one should only use the ADK model when the Keldysh parameter is $\gamma \ll 1$. The details of the derivations can be found in [4, 5]. In its final form, the rate is expressed as:

$$w_{ADK} = C_{n^*l^*}^2 f(l, m) V_{IP} \left(\frac{3E_{\text{laser}}}{\pi \mathcal{E}_0} \right)^{1/2} \left(\frac{2\mathcal{E}_0}{E_{\text{laser}}} \right)^{2n^* - |m| - 1} \exp\left(-\frac{2\mathcal{E}_0}{3E_{\text{laser}}}\right) \quad (2.2)$$

here

$$C_{n^*l^*}^2 = \frac{2^{2n^*}}{n^* \Gamma(n^* + l^* + 1) \Gamma(n^* - l^*)}$$

$$f(l, m) = \frac{(2l + 1)(l + |m|)!}{2^{|m|} (|m|)! (l + |m|)!}$$

$$n^* = \frac{Z}{\sqrt{2V_{IP}}}$$

$$l^* = n^* - 1$$

$$\mathcal{E}_0 = (2V_{IP})^{3/2}$$

where n^* is the effective principle quantum number, l^* is the effective azimuthal quantum number, l is the azimuthal quantum number, m is the magnetic quantum number, and V_{IP} is the ionization potential energy.

For the ionization of Ar^{8+} ($V_{IP} = 143.5$ eV) in a typical ultrastrong laser field with a wavelength of 800 nm and an intensity of 2×10^{19} W/cm², the Keldysh parameter γ has a value of 0.0077, which is much smaller than unity. And for a Gaussian laser pulse, the time interval of our calculations has a typical value of 0.007 fs so that the instantaneous electric field can be safely considered as constant during the tunneling. Therefore we choose the ADK model to calculate the tunneling

ionization rate due to the quasi-static nature ($\gamma \ll 1$) of the laser fields in the works presented in this dissertation, as well as the excellent agreement between the calculated ADK ionization rate and the experimentally observed ion yields [6 - 11].

2.2 Sequential Ionization Ion Population

A typical laser pulse has a temporal profile as shown in Figure 2.1. As the electric field increases, the barrier of the deformed Coulomb potential is further suppressed, allowing the tunneling ionization for the tighter bound electrons. The process of successively removing electrons from an outer-most shell to an inner-most shell in a time-dependent electric field is known as sequential ionization (SI).

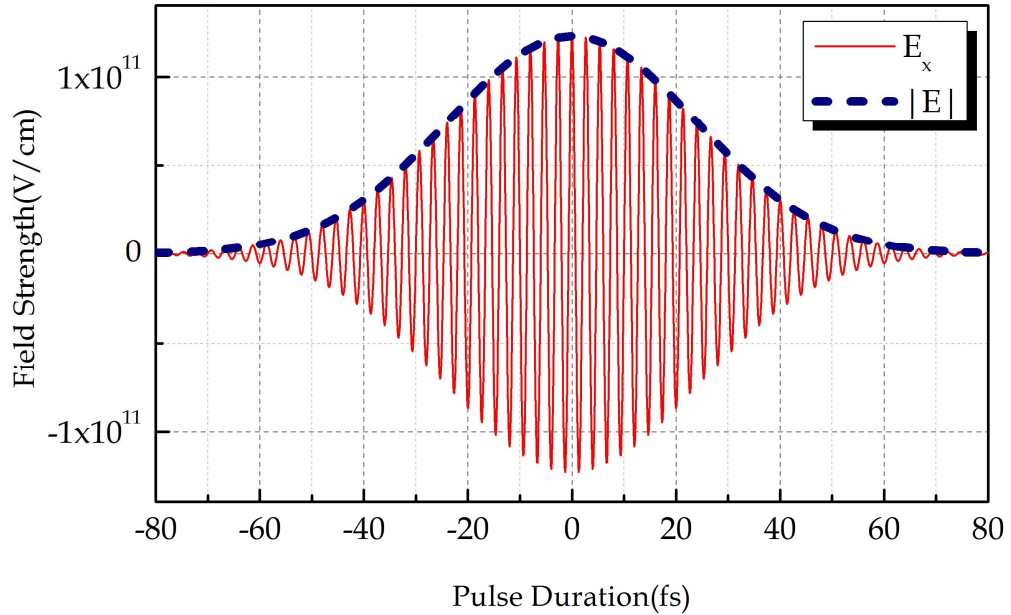


Figure 2.1 The temporal profile of the electric field of a typical Gaussian laser pulse with peak intensity of $2 \times 10^{19} \text{ W/cm}^2$, wavelength at 800 nm and full width at half maximum (FWHM) equal to 40 fs.

During the sequential ionization process, the population of each charge state at any given time can be calculated with the rate equations [12], as given by

$$\begin{aligned}
\frac{dp_0}{dt} &= -\Gamma_0 p_0 \\
\frac{dp_1}{dt} &= -\Gamma_1 p_1 + \Gamma_0 p_0 \\
&\dots \\
\frac{dp_k}{dt} &= -\Gamma_k p_k + \Gamma_{k-1} p_{k-1} \\
&\dots \\
\frac{dp_n}{dt} &= \Gamma_{n-1} p_{n-1}
\end{aligned}$$

where p_k is the population of the k th charge state (specifically p_0 is the population of the neutral atom and p_n is the population of the highest ionized state) and Γ_k is the tunneling ionization rate for the k th charge state calculated with the ADK model (Equation 2.2). The solutions to the above rate equations are given as

$$\begin{aligned}
p_0(t) &= \exp(-\phi_0(t)) \\
p_1(t) &= \exp(-\phi_1(t)) \int_{-\infty}^t \exp(\phi_1(s)) \Gamma_0(s) p_0(s) ds \\
&\dots \\
p_k(t) &= \exp(-\phi_k(t)) \int_{-\infty}^t \exp(\phi_k(s)) \Gamma_{k-1}(s) p_{k-1}(s) ds \\
&\dots \\
p_n(t) &= \int_{-\infty}^t \Gamma_{n-1}(s) p_{n-1}(s) ds
\end{aligned}$$

where $\phi_k(s)$ is the running integral of ADK rate defined as

$$\phi_k(s) = \int_{-\infty}^s \Gamma_k(s') ds'$$

By taking a reasonable small time interval dt (e.g. 0.007 fs), a good convergence can be obtained in the conservation of the sum of all charged states population. Figure 2.2 shows the population evolution of Neon charge states as a function of the laser pulse time, where the laser pulse has an intensity of $2 \times 10^{19} \text{ W/cm}^2$, a wavelength at 800 nm and a FWHM of 40 fs (Figure 2.1). As the electric field is increasing to its peak value, deeper and deeper charge states successively become ionized and then saturated. The whole $n = 2$ valence shell is removed 40 fs before the arrival of the peak of the laser pulse. The final ion state is Ne^{8+} since $2 \times 10^{19} \text{ W/cm}^2$ is not sufficient to ionize the 1s electron ($V_{IP} = 1362 \text{ eV}$). For any time interval (dt), the ionization probability of the k th charge state is calculated as $p_k(t + dt) - p_k(t)$. The total population of all charge states at any time is well conserved.

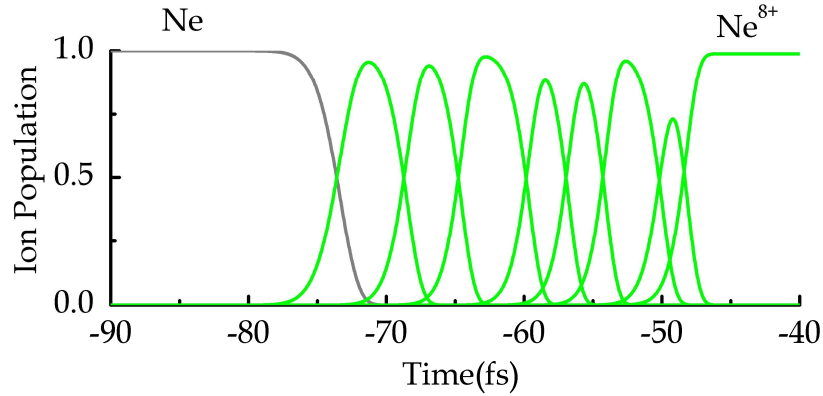


Figure 2.2 Population evolution for Neon charge states from neutral Ne to Ne^{8+} as a function of the laser pulse time.

2.3 Ultrastrong Laser Field

2.3.1 Beam Spatial Profile

The laser beam is collimated and focused to a region which is typically only a few micrometers in diameter in order to produce ultra-high intensity in the experiments. Figure 2.3 shows the spatial profile of a Gaussian laser beam (TEM_{00}) propagating along z direction near its focal region. The beam converges to a minimum diameter known as the beam waist, w_0 , at $z = 0$. Its value is defined as the width at which the transverse intensity decreased by a factor of $1/e^2$ of its maximum value [13] given by

$$w_0 = \left(\frac{4}{\pi}\right) \frac{f\lambda}{D} \quad (2.3)$$

where f is the collimating optics' focal length ($f = 2$ is adopted for the works presented in this dissertation), D is the $1/e^2$ diameter of the collimated incident beam, and λ is the wavelength of the laser beam. The radius of the beam at a distance z from the beam waist is defined as

$$w(z) = w_0 \sqrt{1 + \left(\frac{z}{z_R}\right)^2} \quad (2.4)$$

where z_R is known as the Rayleigh length, which is the distance along the propagation direction of the beam from the beam waist to the place where the area of the cross section is doubled (or diameter increased by a factor of $\sqrt{2}$), and can be calculated as

$$z_R = \frac{\pi w_0^2}{\lambda} \quad (2.5)$$

The beam width has been used to gauge the excursion of the electron at the focal region. It is also used in the calculation of photoelectron trajectory to determine whether the photoelectron is out of the laser's focal region.

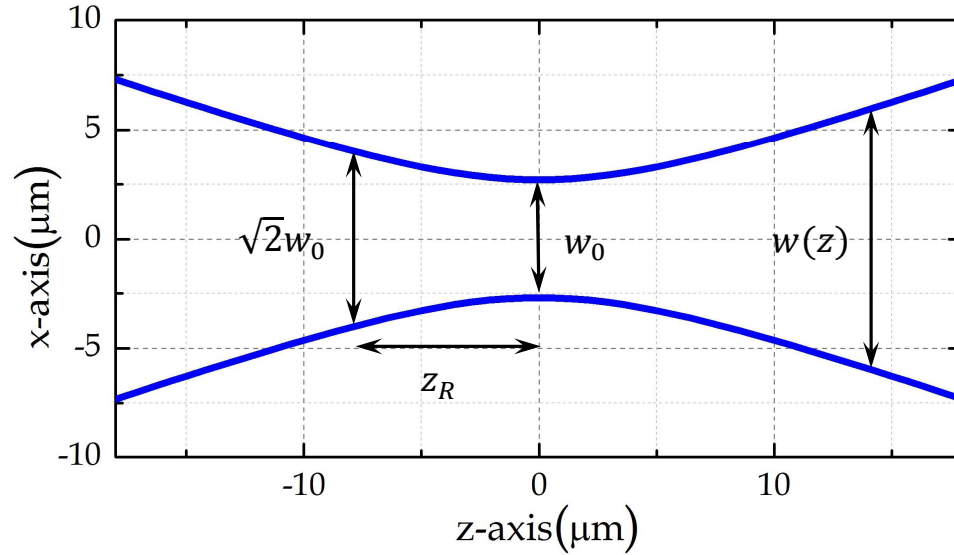


Figure 2.3 Spatial profile of Gaussian laser beam. The transverse beam width, w (blue solid) as a function of the axial distance, z .

2.3.2 Spatial Intensity Profile

The experimental collection of the photoelectron yield is a result of averaging over the laser's focal region. In order to mimic that in the calculation, one needs to understand the spatial intensity profile of the laser beam in order to perform the appropriate focal volume integration. The intensity has dependences on both the space and time with the form

$$I(r, z, t) = I_0(t) \frac{w_0^2}{w(z)^2} \exp\left(-\frac{2r^2}{w(z)^2}\right) \quad (2.6)$$

where $I_0(t)$ is the peak intensity at t (Figure 2.1), $w(z)$ is the beam diameter defined in equation (2.4) and $r = \sqrt{x^2 + y^2}$ as the transverse distance from the propagation axis (z). Figure 2.4 shows a typical contour plot of the intensity spatial profile with a peak intensity at $2 \times 10^{19} \text{ W/cm}^2$. The asymptotic iso-intensity boundary follows the divergence of the beam width (Figure 2.3) along both the propagation and transverse directions. It should be noted that the intensity drops quickly from 10^{19} W/cm^2 at the center to 10^{17} W/cm^2 within only $3 \mu\text{m}$ at $z = 0$ in the transverse direction, which is characteristic for ultrastrong laser fields. This dramatic change in the gradient of the laser field has a huge impact on the released photoelectron dynamics in the continuum. The volume of an iso-intensity profile at I can be shown as [14]

$$V(I) = \frac{\pi w_0^4}{\lambda} \left(\frac{2}{9} \xi^3 + \frac{4}{3} \xi - \frac{4}{3} \arctan(\xi) \right) \quad (2.7)$$

where $\xi = \sqrt{I_0/I - 1}$ and I_0 is the peak intensity at the focus.

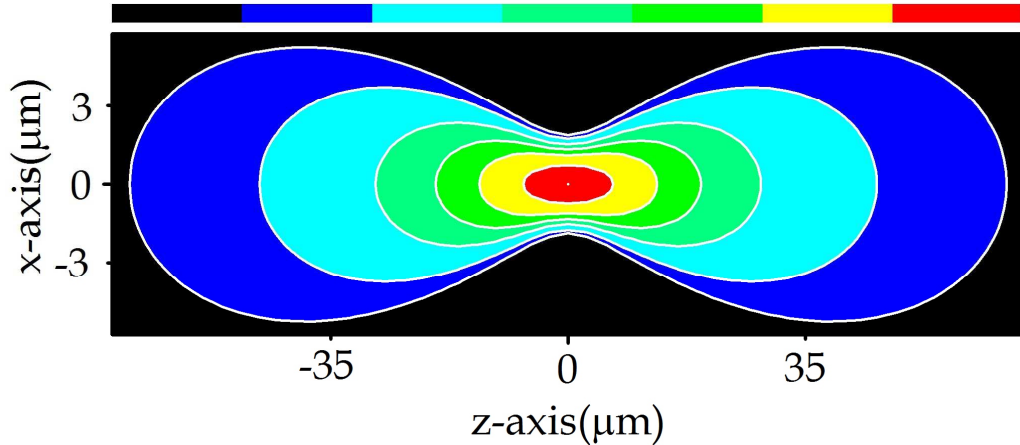


Figure 2.4 Contour plot ($y = 0$) of the intensity spatial profile at $I_0(t) = 2 \times 10^{19} \text{ W/cm}^2$ near the laser beam focus. The intensity values are shown on a logarithmic 7 colors scale span from $1 \times 10^{17} \text{ W/cm}^2$ (Black) to $1 \times 10^{19} \text{ W/cm}^2$ (Red).

In real photoelectron experiments, a skimmed gas beam (with a typical density of 10^{11} atoms/cm³) of the atom species of interest is produced by a gas jet to intersect with the laser beam at its focal region at a constant speed [6]. The gas beam has a typical half-density width of 0.9 mm, which is about 10^3 times larger than the beam waist of the laser. In calculation, we mimic the experimental set up by allocating atoms within a volume defined by an iso-intensity profile at a certain intensity ($I_{\text{threshold}}$), which essentially truncates the theoretically infinitely large Gaussian iso-intensity spatial volume to a finite one. The atoms are distributed uniformly within the volume to ensure the constant gas density. Choosing an appropriate $I_{\text{threshold}}$ requires careful considerations. A lower threshold intensity means larger integration volume and better simulation of the experiments. But lower threshold intensity also requires more atoms to be allocated and subsequently longer computation time and more memory for data storage. In our studies, the iso-intensity spatial volume at 1% of the peak intensity has been proven to yield convergent and accurate calculation results as well as consumes a reasonable amount of computation resources, as shown in Figure 2.5. As shown in Figure 2.5(b) for the photoelectron energy spectrum of Neon, the iso-intensity profiles with large values (e.g. 20%) generally underestimate the yield of photoelectrons with low kinetic energy. A difference of two orders of magnitude in the photoelectron yield is shown by including the contributions from the 1% iso-intensity profile. Good convergence is achieved by expanding the integration volume to the iso-intensity profile at 1% (2×10^{17} W/cm²) of the peak intensity. In the calculations for the photoelectron yield experiment, a typical number of 70,000 atoms are uniformly allocated within the 1% iso-intensity spatial volume. The whole simulation process

can be finished in a typical length of 8 hours with the help of parallel computation on a Unix server. The details of computation scheme can be found in section 2.6.

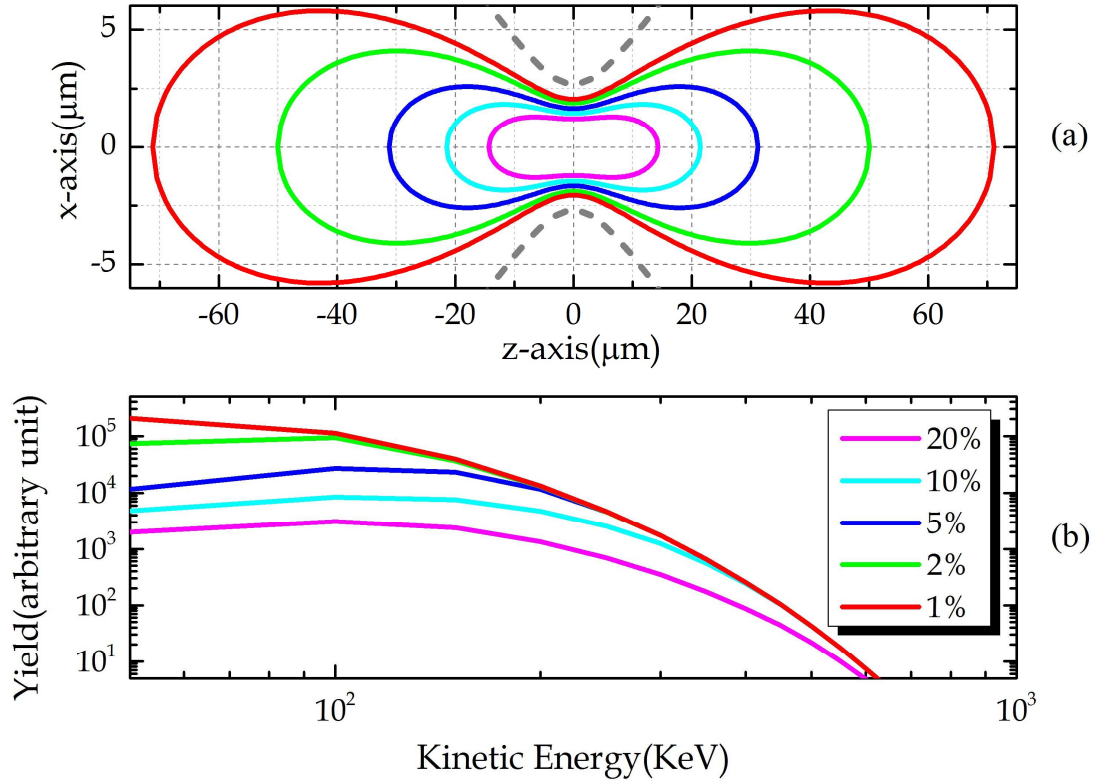


Figure 2.5 (a) The iso-intensity spatial profile at the focal region of a Gaussian laser beam. The intensities are 20%, 10%, 5%, 2% and 1% of the peak intensity from inner most (purple solid) to outer most (red solid). The grey dashed line specifies the beam width in the same focal region. (b) The photoelectron energy spectrum of Neon for polar angle of 70° obtained with different iso-intensity spatial volume specified in (a). In both (a) and (b), the laser pulse has an intensity of 2×10^{19} W/cm², wavelength of 800nm, and FWHM of 40 fs.

2.3.3 Electromagnetic Field

In moderate-strong intensity physics, the plane-wave approximation is widely adopted due to its simplification and accuracy in calculations. The plane-wave electromagnetic field is transverse to the propagation axis (z) of the laser field and there are no longitudinal (z) components for both the $\vec{E}$ and $\vec{B}$ fields. Mathematically, the plane-wave solution is a result of the paraxial approximation which itself is not compatible with the exact Maxwell equations by violating the divergence condition [15]. The validity of paraxial approximation in moderate strong field physics is provided by the fact that the laser beam width is much larger than the wavelength of the laser.

As one moves to the ultrastrong field regime, the laser beam is so tightly focused such that the scale of the laser beam waist is comparable to the wavelength of the laser field (see Figure 2.4, the transverse diameter of the iso-intensity profile at $1 \times 10^{19} \text{ W/cm}^2$ and $z = 0$ is only about 500 nm comparing to the 800 nm wavelength). In such a scenario, the paraxial approximation is no longer valid and the usually neglected longitudinal component of the electric field becomes non-trivial. As shown in Figure 2.6, the value of the E_z component at the laser focus can be as large as $1.3 \times 10^{12} \text{ V/m}$, which is more than 10% of the peak value of E_x at $1.2 \times 10^{13} \text{ V/m}$. Exact treatment of the electromagnetic field is needed.

Lax, Louisell and McKnight [16] developed the exact solutions for both the transverse and longitudinal components by expanding the fields as a power series in the ratio of beam waist to diffraction length, which is defined as $\varepsilon = w_0/l$, where l is the diffraction length defined as

$$l = \frac{2\pi w_0^2}{\lambda} \quad (2.8)$$

By performing Finite-difference time-domain numerical analysis to validate the Maxwell equations, good accuracy is achieved by adopting the third order expansion solution. Below are the expressions for all the field components of the third order and the details of the derivation can be found in [17, 18].

$$\begin{aligned}
E_x &= E(r, z) \left\{ \cos(\phi + \alpha) + \varepsilon^2 \left[\cos(\phi + 3\alpha) \frac{2x^2 + r^2}{s(z)^2} - \cos(\phi + 4\alpha) \frac{r^4}{s(z)^3 s_0} \right] \right\} \\
E_y &= E(r, z) \varepsilon^2 \cos(\phi + 3\alpha) \frac{2xy}{s(z)^2} \\
E_z &= -2E(r, z) \varepsilon \frac{x}{s(z)} \left\{ \sin(\phi + 2\alpha) \right. \\
&\quad \left. + \varepsilon^2 \frac{r^2}{s(z)^2} \left[3 \sin(\phi + 4\alpha) - \cos(\phi + 5\alpha) \frac{r^2}{s(z) s_0} \right] \right\} \\
B_x &= E_y \\
B_y &= E(r, z) \left\{ \cos(\phi + \alpha) + \varepsilon^2 \left[\cos(\phi + 3\alpha) \frac{2y^2 + r^2}{s(z)^2} - \cos(\phi + 4\alpha) \frac{r^4}{s(z)^3 s_0} \right] \right\} \\
B_z &= E_z \frac{y}{x}
\end{aligned} \tag{2.9}$$

where

$$\begin{aligned}
E(r, z) &= E_0 \frac{w_0}{w(z)} \exp \left(-\frac{r^2}{w(z)^2} \right) \\
r^2 &= x^2 + y^2 \\
\phi &= w_0 t - kz - \frac{zr^2}{z_R w(z)^2} \\
\alpha &= \arctan \left(\frac{z}{z_R} \right)
\end{aligned} \tag{2.10}$$

where w_0 , $w(z)$, and z_R are defined in equation (2.3), (2.4) and (2.5). The above solutions are for a linearly polarized laser field. The solutions for a circularly polarized field can be obtained as the superposition of two linearly polarized fields with polarization directions perpendicular to each other and with a phase difference of $\pi/2$. The inclusion of the longitudinal components significantly changes the electron dynamics in the continuum as will be detailed in section 2.4. It has also been shown that the longitudinal components yield significant impact on the photoelectron angular spectrum in the final state [6].

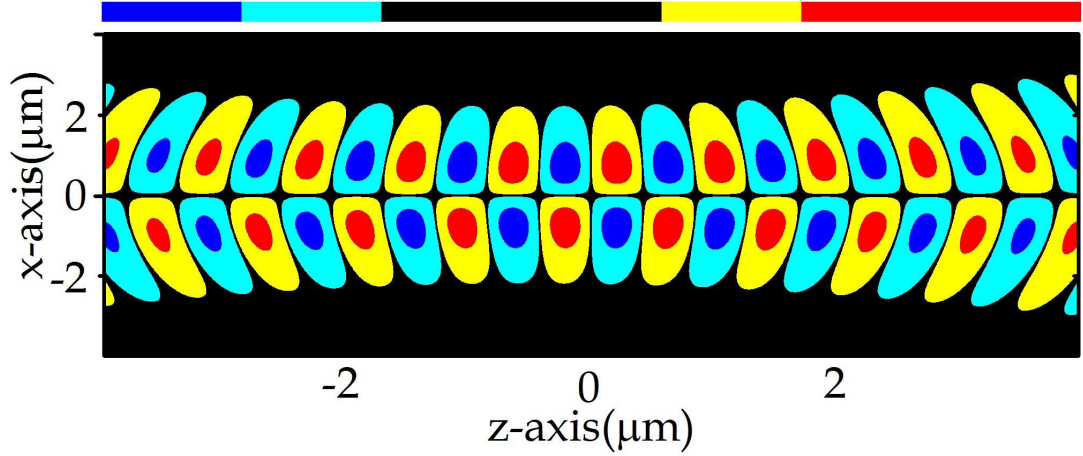


Figure 2.6 Contour plot ($y = 0$) of the spatial profile of the electric field longitudinal component (E_z) at $I_0(t) = 2 \times 10^{19} \text{ W/cm}^2$ near the laser beam focus. The electric field values are shown on a logarithmic 5 colors scale span from $-8 \times 10^{11} \text{ W/m}$ (black) to $8 \times 10^{11} \text{ W/m}$ (red). The longitudinal component is diagonally symmetric respect to the origin. As a reference, the beam waist has a size of $3 \mu\text{m}$.

2.4 Electron Dynamics in Continuum

The electron dynamics in the continuum is subject to its interaction with the laser field. The electric field with plane-wave approximation is described as $\mathbf{E} = \mathbf{E}_0 \cos(\omega t)$. In moderate to strong laser fields ($< 3 \times 10^{16} \text{ W/cm}^2$), the motion of the electron is non-relativistic ($v/c \approx 0$) and the Lorentz force due to the magnetic component of the laser field can be safely neglected, which is known as the dipole-approximation. Under the plane-wave approximation and the dipole approximation, the velocity of the electron can be obtained using Newton's second law as

$$\mathbf{v}(t) = \mathbf{v}_0 - \frac{e}{m_e \omega} \mathbf{E}_0 (\sin(\omega t) - \sin(\omega t_0)) \quad (2.11)$$

where ω is the angular frequency of the oscillation field, m_e is the mass of the electron, e is the charge of the electron, t_0 and $\mathbf{v}_0$ are the initial time and velocity respectively. As can be seen in equation (2.11), the motion of the electron is confined to be one dimensional. The cycle averaged kinetic energy of the electron in the oscillating field is known as the ponderomotive energy [19], given as

$$U_p = \frac{e^2 |\mathbf{E}_0|^2}{4m_e \omega^2} \quad (2.12)$$

As one moves to the ultrastrong intensity regime, the above two approximations are no longer valid. Firstly, both the Gaussian intensity profile with drastically changing gradient and the longitudinal components have to be adopted, which has been described in section 2.3. Secondly, the motion of the electron becomes relativistic as the velocity becomes comparable to the speed of the light. Under the condition of $v/c \approx 1$, the Lorentz force (defined as $\mathbf{v} \times \mathbf{B}$) becomes non-negligible. The electron dynamics become fully relativistic and three-dimensional and the closed

form analytic solution is no longer available. The relativistic pondermotive energy now can be expressed with the non-relativistic U_p as [20]

$$U_p^{Relativistic} = \sqrt{m_e^2 c^4 + 2m_e c^2 \cdot U_p} - m_e c^2 \quad (2.13)$$

Under this circumstance, the electron trajectory in the continuum is solved by solving the relativistic equations of motion numerically

$$\begin{aligned} \frac{dp_x}{dt} &= e \left(E_x + \frac{p_y}{\gamma m_e} B_z - \frac{p_z}{\gamma m_e} B_y \right) \\ \frac{dp_y}{dt} &= e \left(E_y + \frac{p_z}{\gamma m_e} B_x - \frac{p_x}{\gamma m_e} B_z \right) \\ \frac{dp_z}{dt} &= e \left(E_z + \frac{p_x}{\gamma m_e} B_y - \frac{p_y}{\gamma m_e} B_x \right) \\ \frac{dx}{dt} &= \frac{p_x}{\gamma m_e} \\ \frac{dy}{dt} &= \frac{p_y}{\gamma m_e} \\ \frac{dz}{dt} &= \frac{p_z}{\gamma m_e} \end{aligned} \quad (2.14)$$

where γ is the well-known Lorentz factor defined as

$$\gamma = \sqrt{1 + \left(\frac{|p|}{m_e c} \right)^2} \quad (2.15)$$

and $|p| = \sqrt{p_x^2 + p_y^2 + p_z^2}$. The $\vec{E}$ and $\vec{B}$ field components are calculated using the previously defined equations (2.9) and (2.10). The equations are then numerically

integrated with the Runge-Kutta method (see details in section 2.6). With both the accuracy and computation efficiency of the calculation into considerations, a typical value of $1/100$ of the oscillating field period is used. At each step the momentum and energy of the electron are updated and the final energy spectrum and angular spectrum can be obtained for the photoelectron experiment.

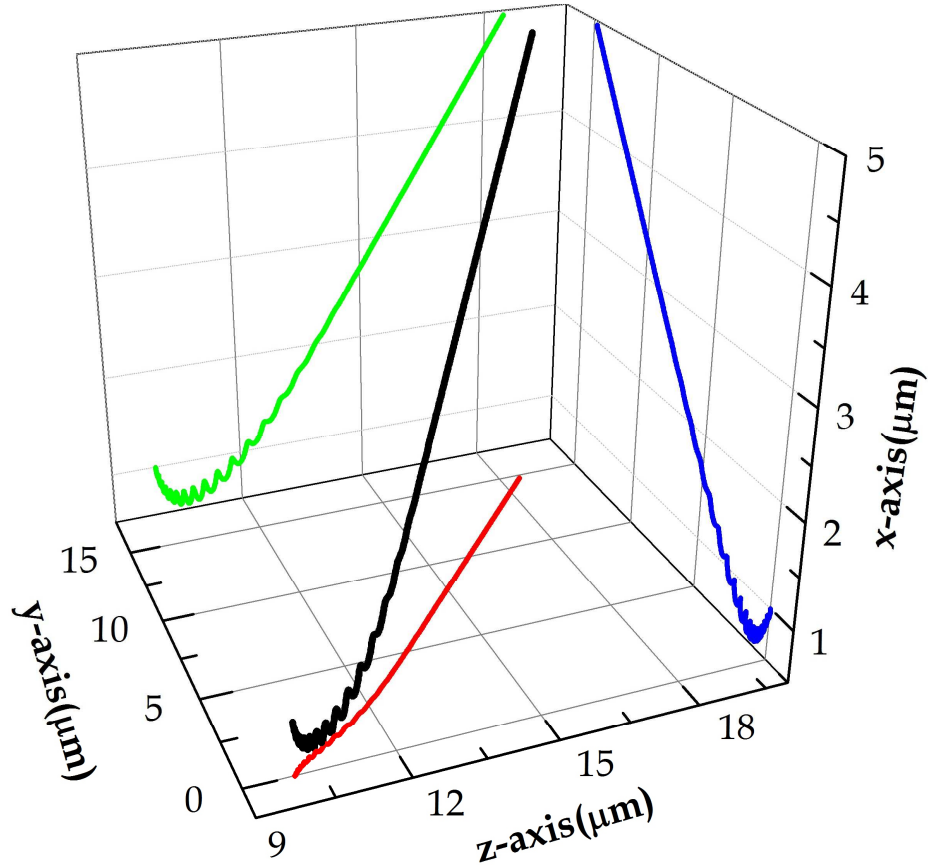


Figure 2.7 Plot of trajectory evolution of an electron released spatially at $(1 \mu\text{m}, 0 \mu\text{m}, 10 \mu\text{m})$ and temporally at -0.242 fs in a laser pulse with profile parameters defined in figure 2.1 and figure 2.4, shown with 3D trajectory (black) and trajectory projections in x - y (blue), y - z (red) and x - z (green).

A sample electron trajectory is shown in figure 2.7. The electron is released at the peak of the laser pulse (-0.242 fs) and located at the 1×10^{19} W/cm² iso-intensity volume ($1 \mu\text{m}$, $0 \mu\text{m}$, $10 \mu\text{m}$) (see figure 2.5). The electron stays within the focal region for several cycles of periods before the drastically changing gradient of the electric field at the Gaussian focal region ejects the electron out of the laser beam with a trajectory in a nearly straight line fashion. The signature of the longitudinal component of the electric field is clearly seen in the x - z plane projection as the E_z pushes the electron in the laser propagation direction.

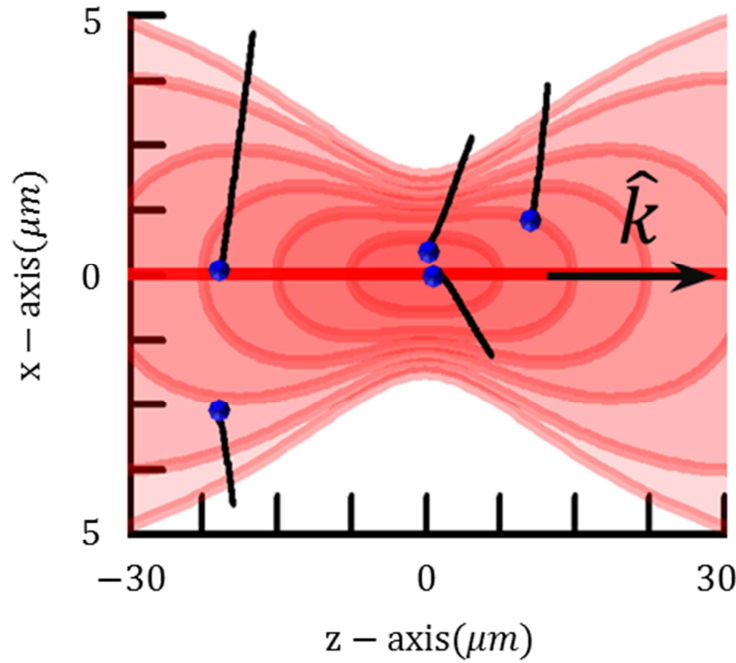


Figure 2.8 Plot of sample electron trajectories, calculated at $(x, y, z) = (0, 0.5 \mu\text{m}, 0)$, $(0, 0, 0.5 \mu\text{m})$, $(-3 \mu\text{m}, 0, 20 \mu\text{m})$, $(1 \mu\text{m}, 0, 10 \mu\text{m})$, and $(-1.5 \mu\text{m}, 0, -20 \mu\text{m})$ over the time periods (initial, final) = $(-2.5$ fs, 14.5 fs), $(-2.5$ fs, 20 fs), $(-60$ fs, -30 fs), $(-100$ fs, 30 fs), and $(-60$ fs, -5 fs), respectively. The shown Gaussian focus intensity contours are at 0.5, 0.2, 0.1, 0.05, 0.02 and 0.01 times the peak intensity of 2×10^{19} W/cm².

More sample trajectories at the ultrastrong Gaussian laser beam are shown in figure 2.8. Electrons exit the laser beam by following a path almost perpendicular to the iso-intensity profiles. The forward angle with respect to the propagation axis due to the longitudinal component is also clearly observed.

It has been mentioned in section 2.3 that the ultrastrong Gaussian field with longitudinal component significantly changes the electron dynamics in a plane-wave field. The Lorentz factor of the electron in the relativistic field is defined in equation 2.15. The γ factor can be used to track the energy gained of the electron from the external electric field. Figure 2.9 compares the γ factor evolution of the electron in a plane-wave approximation field (figure 2.9 (b)) and in an exactly treated Gaussian field (figure 2.9 (c)) where the longitudinal components and magnetic field are both included. For the plane-wave field, the electron's kinetic energy gained from the external field for the full pulse duration is zero, as the electric field changes its direction within each oscillation cycle and the total integration of the electron's acceleration is zero. This process is well seen in figure 2.9 (b) as the electron loses all the energy previously gained from the field and remains at rest ($\gamma = 1$) after the whole pulse passes. However, when the gradient of the Gaussian electric field, the longitudinal components and the magnetic field are included, it's a whole new picture for the electron dynamics. The Lorentz force $\mathbf{v}_x \times \mathbf{B}_z$ now has a nonvanishing cycle average which causes the drift in y direction because B_z is almost in phase with v_x . The integration of acceleration is no longer zero as the electric field is now inhomogeneous in space and as the electron explores the gradient of the electric field, it accumulates a net energy gain. As a result, the electron can exit the laser pulse with a significant amount of net kinetic energy in an ultrastrong field. As clearly shown in

figure 2.9 (c), the electron is ejected out of the Gaussian laser pulse before the pulse reaches its peak at $t = 0$ and becomes essentially free in the space with a relativistic velocity. A good reference about the theory of electron ultraintense laser interaction can be found at [18].

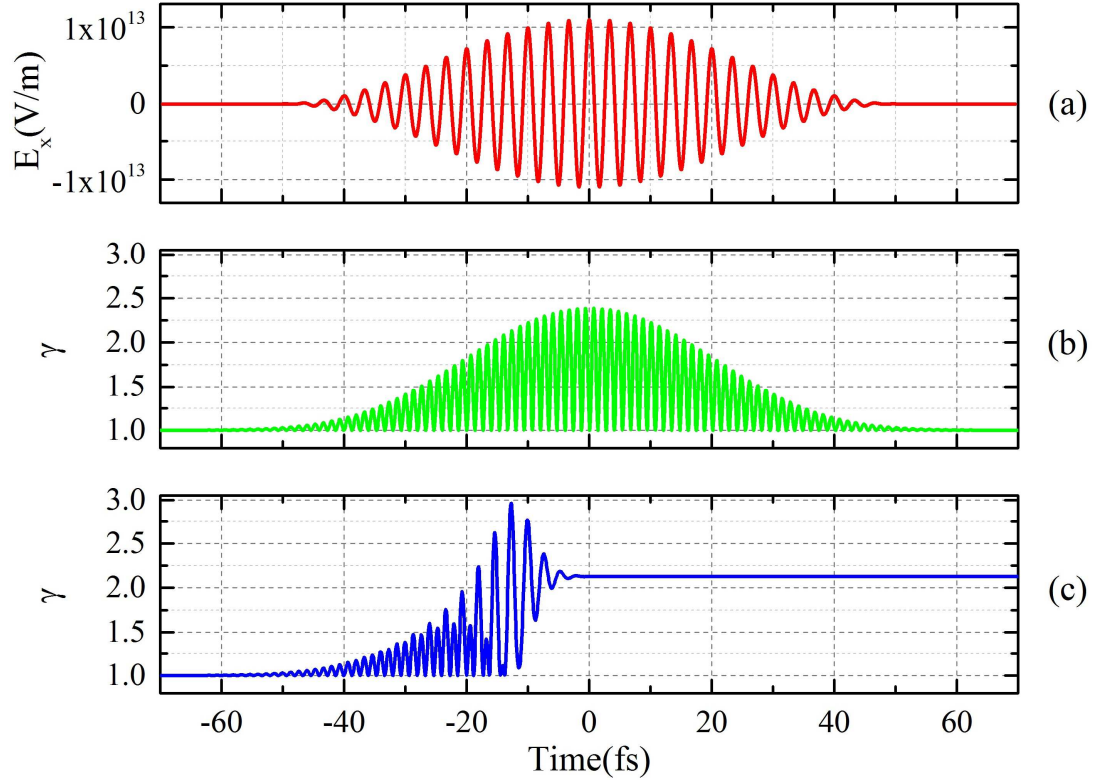


Figure 2.9 Electron γ factor evolution as a function of the pulse time for (b) plane-wave approximation and (c) Gaussian field with longitudinal component and magnetic field included. The pulse (a) has a peak intensity of 1.59×10^{19} W/cm², wavelength of 1 μ m and FWHM of 40 fs.

Closed form analytical solutions are no longer available when the laser field is exactly treated Gaussian field. And the dynamics of the electron become complicated as its final state is extremely sensitive to the initial spatial and temporal conditions of

the laser pulse. Thus numerical simulation suits well in this situation due to its simplicity and accuracy. More details regarding the Monte Carlo trajectory ensemble method will be covered in section 2.6.

2.5 Relativistic Rescattering Dynamics

The rescattering is an important and complex process in the laser-matter interaction physics as be described in section 1.3. To understand it, the whole process is broken down to three steps: (1) electron tunneling through the potential barrier, (2) electron moves in the continuum, (3) electron revisits and interacts with the parent ion. The rescattering mechanisms in the third step in this dissertation are limited to elastic scattering and inelastic rescattering ionization.

2.5.1 Exit of Coulomb Barrier

As shown in figure 1.2(c), the electron tunnels through the Coulomb barrier along the instantaneous polarization direction. The exit point can be numerically calculated by solving the Schrödinger equation with the external electric field included in the parabolic coordinates. The details of the calculation can be found in [5]. In most situations where the external laser field is adiabatic enough, a simple one-dimensional version can be used instead to quickly calculate the exit point, specifically by solving

$$-\frac{Z}{|x|} + E_{\text{laser}} \cdot x = V_{IP} \quad (2.16)$$

where Z is the parent ion charge, E_{laser} is the laser electric field, V_{IP} is the ionization potential and the laser polarization direction is along the x -axis. The electron emerges in the opposition direction with respect to the electric field direction. Attention should be paid that equation (2.16) is only for tunneling ionization and there is no real

solution when the Coulomb barrier is suppressed by the external field to a level below the ionization potential.

Right after the tunneling ionization, an electron wavepacket is formed at the y - z plane which is perpendicular to the polarization axis. The wavepacket has a momentum distribution which is of a normal distribution [21] described as

$$\omega(p) = \frac{1}{\sqrt{2\pi}\sigma_p} \exp\left(-\frac{p^2}{2\sigma_p^2}\right) \quad (2.17)$$

where the standard deviation is a function of the electric field and the ionization potential given as

$$\sigma_p = \sqrt{\frac{E_{\text{laser}}}{2\sqrt{2V_{IP}}}} \quad (2.18)$$

The spatial distribution of the electron wavepacket can be subsequently calculated using the Heisenberg uncertainty principle. The spatial distribution also follows a two-dimensional normal distribution.

In order to mimic the quantum wavepacket, the electron trajectory ensemble method is adopted. Certain numbers of electrons (typical value of 1,000) are launched with their values of initial momentum and position randomly generated from the distribution defined in equation (2.17). In this way, the trajectory of each electron in the ensemble can be easily calculated using the equation (2.14) all the way until it revisits the parent ion, and the statistics of the returning electron wavepacket will then be obtained. A sample contour plot for the electron density of the initial electron wavepacket of He^+ is shown in figure 2.10.

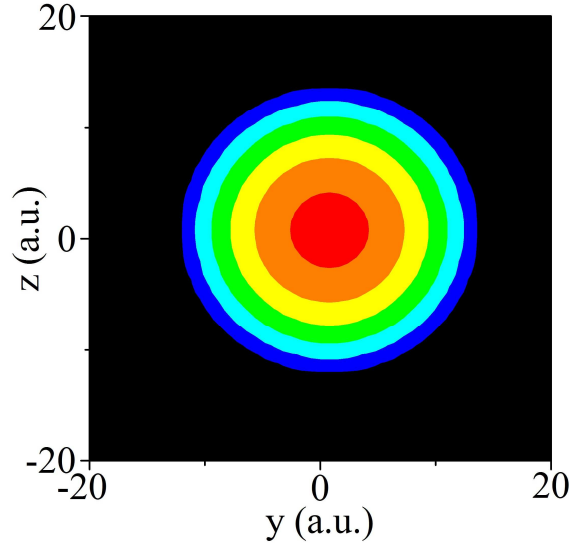


Figure 2.10 Contour plot of He^+ initial wavepacket in the y - z plane. Color scale in logarithm from 10^{-2} (red) to 10^{-7} (blue). The wavepacket is formed near the peak of a laser pulse with intensity of $2.4 \times 10^{15} \text{ W/cm}^2$.

2.5.2 Electron Wavepacket Dynamics

The motion of each electron is calculated with the equation (2.14). In order to understand the dynamics of the electron wavepacket in the continuum, a single electron trajectory is investigated at first. Figure 2.11 shows the elapsed time τ_{elapse} (the duration from the birth time to the return time) and returning kinetic energy K_{return} of the electron as a function of the birth time t_{birth} . By comparing with the temporal profile of the electric field (figure 2.11(a)), it is found that the rescattering can only happen within the $\pi/2$ to π and $3\pi/2$ to 2π ranges of one cycle. Long trajectories are those released with birth phase at $\pi/2$ or $3\pi/2$, which can take up to one full oscillation period before returning back to the parent ion. On the other hand, the long trajectories return with zero kinetic energy because the integration of the acceleration for one full cycle is zero. The maximum returning kinetic energy can be obtained by releasing the electron at 1.8 or 4.9 rad. For the field intensity of $1 \times$

10^{17} W/cm² and wavelength of 800 nm here, the maximum returning kinetic energy is 689 a.u. and the ponderomotive energy (see equation 2.12) U_p has a value of 219 a.u.. This is perfectly consistent with the observation that the maximum returning kinetic energy is 3.17 times the ponderomotive energy found in [22].

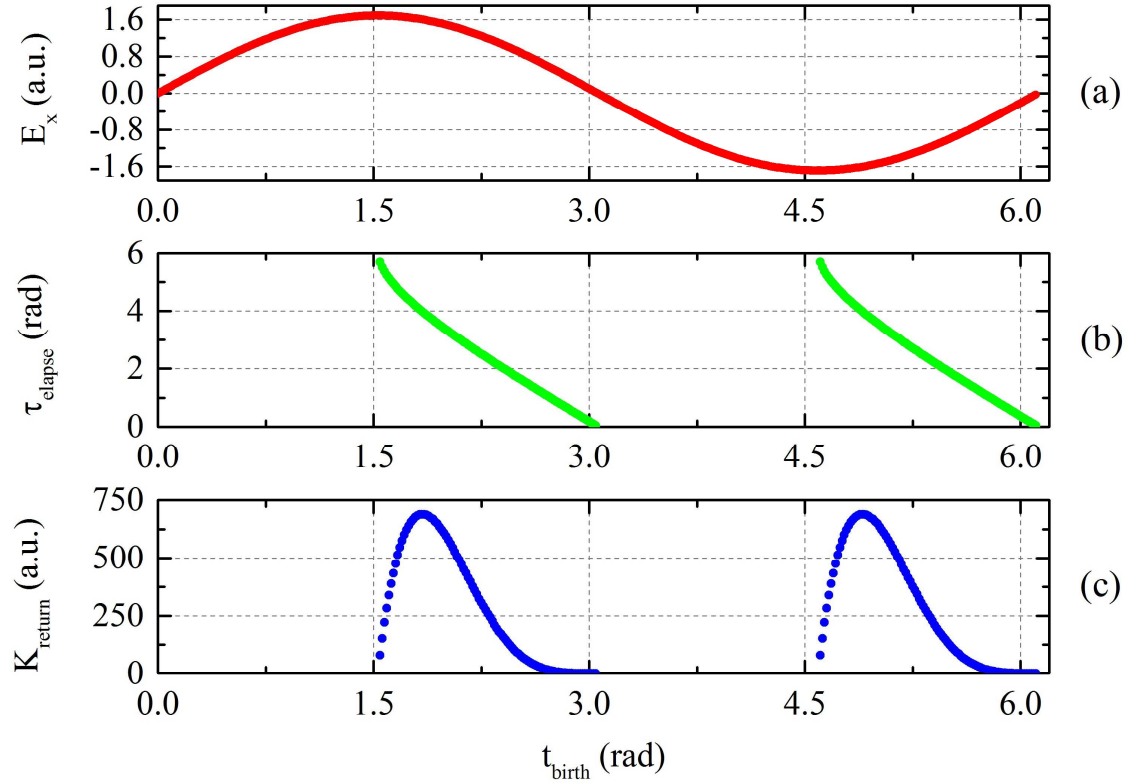


Figure 2.11 Elapsed time ($t_{\text{return}} - t_{\text{birth}}$) (b) and returning kinetic energy as a function of birth phase (t_{birth}). The electric field (a) has intensity of 1×10^{17} W/cm² and wavelength of 800 nm.

The wavepacket width as function of birth phase is shown in figure 2.12. The intensity for each ion species is chosen to have 10% ionization. For each ion specie, it is obvious that those ‘long trajectories’ released near $\pi/2$ return to the parent ion with

largest wavepacket spread due to their longest elapsed time (see figure 2.11(b)). Among the four species, Ar^{9+} has the largest spread because of the huge Lorentz deflection due to its ultrastrong intensity of $1.6 \times 10^{18} \text{ W/cm}^2$.

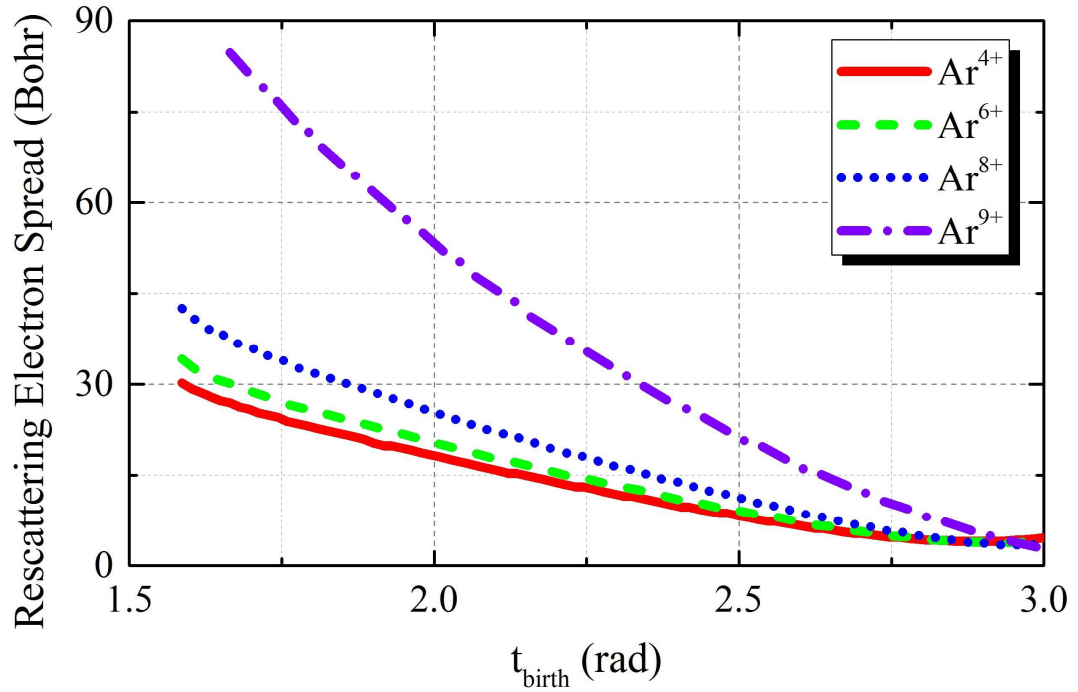


Figure 2.12 Returning electron wavepacket spread as function of birth phase (t_{birth}). The intensity for each species: Ar^{4+} (red solid) at $4.6 \times 10^{15} \text{ W/cm}^2$, Ar^{6+} (green dash) at $1.1 \times 10^{16} \text{ W/cm}^2$, Ar^{8+} (blue dot) at $4.2 \times 10^{16} \text{ W/cm}^2$, Ar^{9+} (purple dash dot) at $1.6 \times 10^{18} \text{ W/cm}^2$.

In the single trajectory study in figure 2.11, the electron is released at the origin with a zero momentum. For the electron trajectory ensemble, each electron has a non-origin position and a non-zero momentum due to the Heisenberg uncertainty (equation 2.17). The evolution of the electron wavepacket can be obtained by tracking all electron trajectories in the ensemble. Shown in figure 2.13 is an example of

electron wavepacket evolution for Ar^{8+} as a function of the elapsed time. The wavepacket spreads out quickly as time passes. The wiggle of the wavepacket is due to the oscillation of the external electric field. And it is evident that the Lorentz force due to the magnetic field manages to displace the wavepacket by ~ 50 a.u. along z . As the intensity increases, the Lorentz deflection of the returning wavepacket gets even larger and significantly reduces any rescattering ionization yield [7]. By compiling the position of each trajectory in the electron trajectory ensemble, we can statistically calculate out the width and position of the returning wavepacket and then find out the magnitude of the returning flux [11].

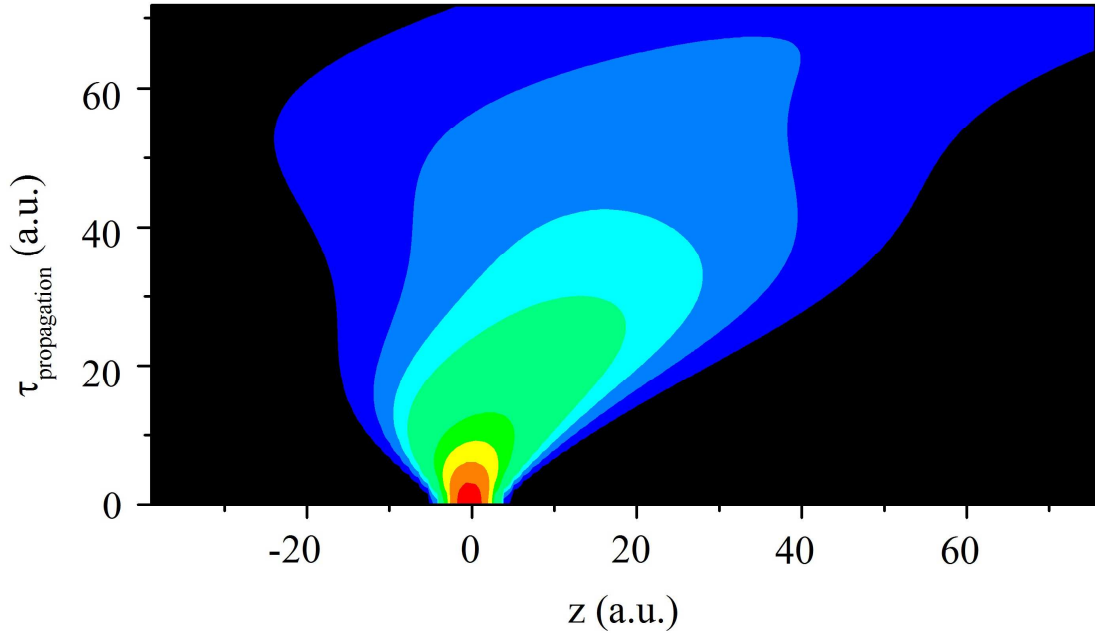


Figure 2.13 Evolution of electron wavepacket in the laser propagation direction (z axis) as a function of elapsed time (since birth phase). The laser intensity is $5.2 \times 10^{16} \text{ W/cm}^2$ and wavelength is 800 nm. The electron is released at -116 a.u. comparing to $\text{FWHM} = 1653$ a.u..

2.5.3 Elastic Scattering

Rutherford scattering was firstly discovered in 1911. It is a process of elastic scattering of charged particles by the Coulomb potential. It has been well studied for hydrogen like species, by using the Yukawa potential [23] to describe the screened Coulomb potential

$$V(r) = \frac{Ze^{-\mu_0 r}}{r} \quad (2.19)$$

where μ_0 is the screening parameter, Z is the charge of the ion. The returning wavepacket can be safely treated as a plane wave packet and the elastic crossing section in atomic unit ($\hbar = m_e = 1$) can be derived with Born Approximation [24]

$$\frac{d\sigma}{d\Omega} = \frac{4Z^2}{(\mu_0^2 + 8K\sin(\theta/2)^2)^2} \quad (2.20)$$

where K is the incident energy of the charged particle and θ is the deflected angle (see figure 2.14). The screening parameter μ_0 can be found by fitting the formula to experimental data and the classical Rutherford scattering formula works well for hydrogen like species when returning energy is larger than a few hundred eV.

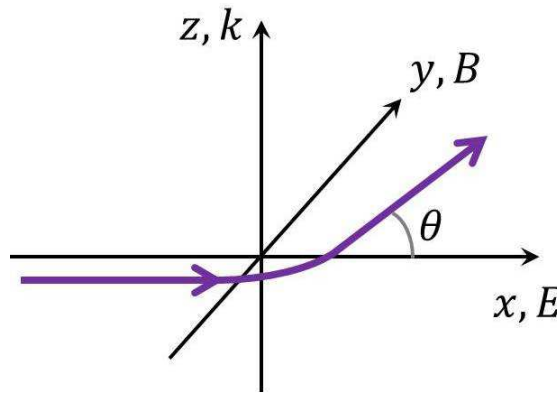


Figure 2.14 Coordinate system for scattering with parent ion (origin).

For real species such as noble gases, the quantum partial-wave calculations are carried out to calculate the elastic scattering cross section where the Coulomb phase shift needs to be numerically calculated [25, 26]. And instead of using the classical Yukawa potential, Hartree-Fock density is employed to describe the screening of the nucleus with the electron density being expressed as [27]

$$\rho_e = \frac{Z}{4\pi r} \sum_{i=1}^3 D_i d_i^2 \exp(-d_i r) \quad (2.21)$$

where D_i and d_i are fitting coefficients calculated using the ELSEPA routine [27]. The calculated ELSEPA elastic scattering cross sections for Ar^{8+} at different energies are shown in figure 2.15. It has been reported that the final electron kinetic energy can be up to 10 times ponderomotive energy when electron is back scattered by the Coulomb potential [26].

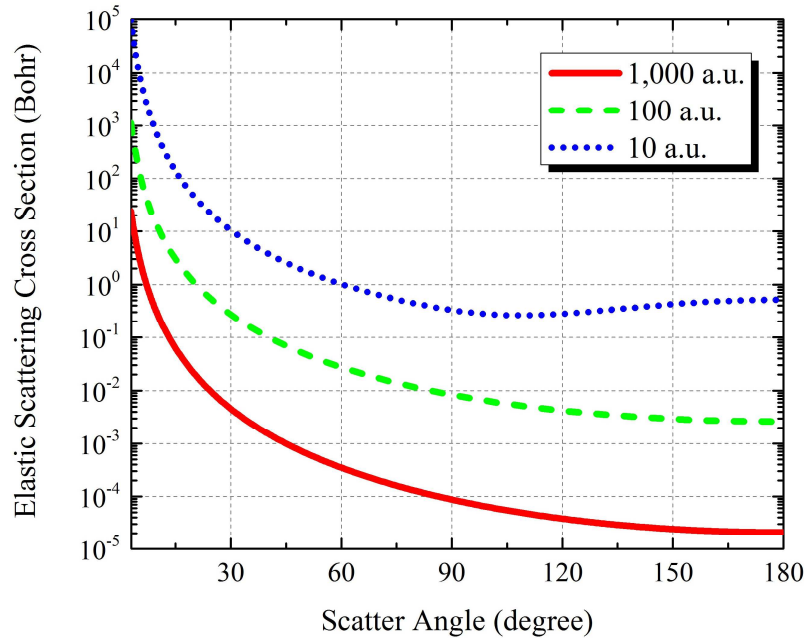


Figure 2.15 Elastic scattering cross section calculated with ELSEPA for Ar^{8+} at 1,000 a.u. (red solid), 100 a.u. (green dash) and 10 a.u.(blue dot).

2.5.4 Inelastic Scattering

As for the inelastic scattering impact ionization cross section, the most successful empirical formula is the one proposed by Lotz [28]

$$\sigma_{\text{Lotz}} = c_1 \ln\left(\frac{K}{V_{IP}}\right) \times \frac{\left(1 - c_2 \exp\left(-c_3 \left(\frac{K}{V_{IP}} - 1\right)\right)\right)}{K \cdot V_{IP}} \quad (2.22)$$

where c_i are coefficients obtained by fitting to experimental cross section data. Lotz's formula has been used in [11] and [7] to calculate the nonsequential ionization yield due to inelastic rescattering.

For special situations where hydrogenic ions are considered, a formula proposed by Clark [29] provides a straightforward way to calculate the cross section,

$$\sigma_{\text{Clark}} = \frac{\pi}{V_{IP}} \left(\frac{n}{Z}\right)^2 r_{nl} \sigma_{nl}^{\text{HR}}(u) \quad (2.23)$$

Where u is the impact electron energy in threshold units and $\sigma_{nl}^{\text{HR}}(u)$ is the reduced hydrogenic cross section defined in [29]. Figure 2.16 compares the cross section calculated with equation (2.23) to experimental data for the $\text{Ne}^{9+} + e \rightarrow \text{Ne}^{10+} + 2e$ process and good agreement is obtained.

2.6 Computation Methods and Model Validity

In the simulation for photoelectron energy spectrum, a typical number of 70,000 atoms are uniformly allocated within the 1% iso-intensity spatial volume (see figure 2.5). Each atom experiences the full duration of the laser pulse. The full duration is chopped into intervals each with a typical value of 1/100 of the oscillation period (see figure 2.1). At each time step, the tunneling ionization rate for the atom is

calculated with the ADK model (see equation 2.2). When the calculated ionization yield is larger than a preset threshold (a typical value of 10^{-4} a.u.), an electron ensemble is launched at the calculated exit point (see section 2.5.1) with calculated spread width (see equation 2.17). The electron ensemble typically has 1,000 electrons. For each electron, its trajectory is followed by solving the equations of motion defined in equation (2.14) where the electromagnetic field is treated exactly with the components given in equation (2.10). We use the RKSUITE routine to numerically solve the equations of motion. The trajectory is followed until it exits the beam region and its final energy and momentum are recorded as raw data.

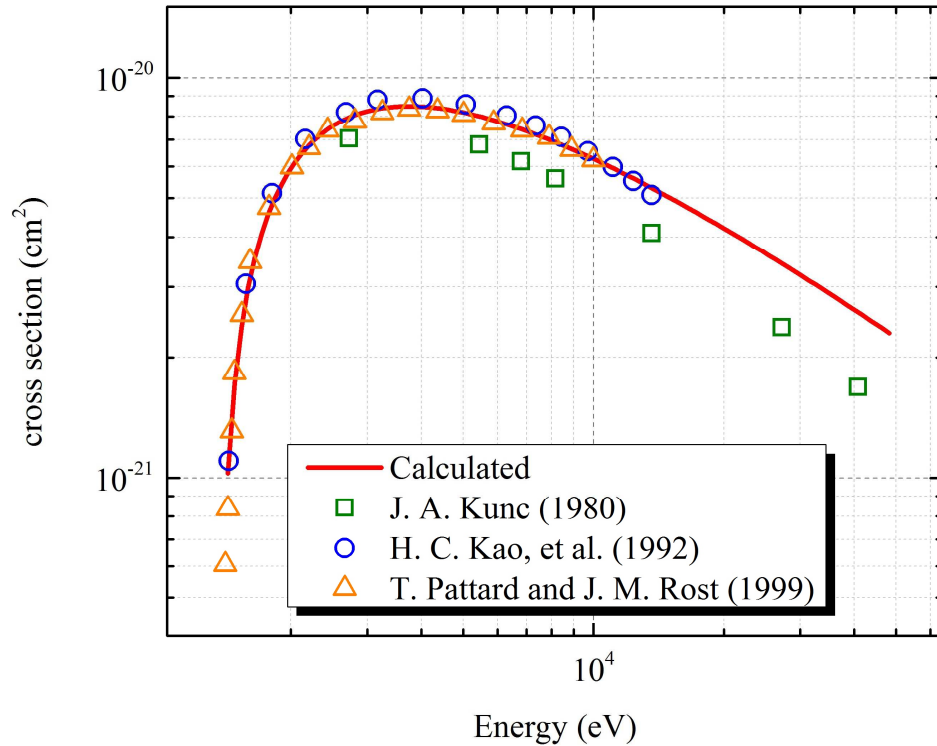


Figure 2.16 Impact ionization cross section for $\text{Ne}^{9+} + e \rightarrow \text{Ne}^{10+} + 2e$.

As for the elastic scattering study, we use the ELSEPA routine [25] to calculate the elastic cross section when each electron trajectory in the electron ensemble revisits the parent ion, and subsequently the elastic scattering yield, which can be obtained by multiplying the cross section with the return flux. Instead of calling the ELSEPA routine repeatedly for each electron, we can optimize this process by generating a list of incident energy and corresponding cross section at the start. Therefore given the energy of each electron, we can quickly retrieve its cross section by applying binary search algorithm.

All the calculations are carried on the mills.hpc.udel.edu cluster at University of Delaware. Unix shell scripts are written to facilitate parallel computation. Adopting the famous Conquer-and-Divide ideology, the main program generates the raw data of the simulation while separate programs are written for data analysis (energy spectrum, angular spectrum, contour plot, etc.). This enables us to generate raw data once, which is the most time consuming process, and perform different analysis on the raw data later on. Also by separating the two parts, we reduce the possibility of having code bugs ruining the main program. With the parallel computation and optimizations, one iteration of the simulation has been reduced to couple hours, a huge productivity boost compared to legacy programs [30].

The validity of our developed model has been examined by comparing with experimental collections and its successes can be found in the publications [6-11]. And simulations have been performed before the conduct of experiments to determine the optimal angle for photoelectron detector.

Here we show a sample pseudo code and the real codes can be found in appendix,

```

*pseudocode for non-sequential ionization ion yield simulation
configure parameters for intensity, wavelength, ion potential...
for each atom allocated in the 1% iso-intensity volume
    for each time step of the pulse laser
        calculate tunneling rate with ADK model
        if rate larger than threshold then wavepacket released
        for each electron in ensemble
            calculate electron trajectory with rksuite subroutine
            calculate returning fluence of wavepacket
            calculate cross-section and non-sequential ionization yield
            data record
        data record
    data record

```

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Chapter 3

CLASSICAL STUDY OF ATOMIC BOUND STATE DYNAMICS IN CIRCULARLY POLARIZED ULTRASTRONG FIELDS

3.1 Introduction

The electric dipole approximation for light has facilitated a physical understanding of light–matter interactions across an expansive number of systems. For light with wavelengths from x-rays [1] to microwaves [2] at intensities from a single photon to 10^{17} W/cm², the dipole approximation is used ubiquitously in quantum and classical treatments with success in atoms, molecules, clusters, and solids. The key question on whether the dipole approximation has begun to fail can be gauged by how the dynamics and observations [3] are explained by considering only the electric field of the light, E_{laser} . In this ultraintense field study, we are particularly interested in the dynamics, energy, and ionization of the bound states as they are affected not only by the laser electric field, E_{laser} , but also by the magnetic field component of the field, B_{laser} . We present the result for when the interaction is primarily with the bound state and ionization is negligible, i.e. less than a few per cent, and when the system is highly ionizing, i.e. between 10% and 100%.

Ultraintense fields are often described as those where the classical nonlinearity parameter, $a_0 = e|E_{\text{laser}}|\lambda/(2\pi mc^2)$, is greater than one in atomic units for an electron charge, e , mass, m , in a field magnitude, $|E_{\text{laser}}|$ at a wavelength λ . When a_0 is greater than one the electron motion in the continuum becomes relativistic within

one laser period [4]. Typically this occurs at optical frequencies for fields of order 10^{18} W/cm² and greater. In ultraintense fields, non-dipole effects from B_{laser} can affect the basic photoionization process [5, 6]. The current understanding of this breakdown is limited to the propagation of the photoelectron in an ultra-intense continuum [7] where B_{laser} has been calculated to deflect the electron by nanometers during the first optical cycle subsequent to ionization [8]. There is growing evidence [9] that rescattering processes (where the photoelectron is driven back into the parent ion by the strong external light field and can give rise to non-sequential ionization, multi-electron excitations, high-harmonic radiation, and attosecond pulses) will begin to shut down due to the B_{laser} deflection [10–13].

Theory efforts are now addressing how the bound state dynamics and ionization process are affected by the ultrastrong field. The methods include semiclassical and classical approaches [14, 15], such as those used in this work, and relativistic quantum treatments [15, 16]. Similar to the B_{laser} continuum deflection, the bound electron has been calculated to undergo a B_{laser} deflection as it ionizes. Classically, this is understood as the Lorentz deflection of the electron by the external B_{laser} field as it makes the final pass in its trajectory by the ion core and out to the critical radius as it ionizes [14]. This deflection is understood quantum mechanically [17] to extend to under the barrier dynamics. As one moves into the interaction of the ultrastrong field with the bound state, questions remain on the role of B_{laser} and an intuitive physical picture has yet to be reached on single- or multi-electron physics. This atomic response to ultrahigh laser fields is an important initial condition for complex phenomena found in plasmas [18], x-ray generation [19], and laser based particle acceleration [20]. At optical frequencies, laser plasma physics [21, 22] and

radiation [23] from ultrastrong field interactions are now being reported for intensities from 10^{19} to 10^{20} W/cm² making the need for such insight more urgent.

Our earlier study [14] focused on the linear polarized (LP) light case. Here we follow this work and test the dipole approximation in ultrastrong, circularly polarized (CP) fields. With CP light, new strong field effects can occur depending on the rotation of the field with respect to the angular momentum of the bound state [24, 25] and the role of B_{laser} may be more prominent since the magnitude of the field is constant as it rotates every cycle rather than oscillating through zero. Perhaps most important is the fact recollision is less of a factor with CP light. The resulting simplification of possible excitation pathways for the atomic system greatly aids in the interpretation and modelling for the pioneering experimental studies [3] in this new, ultrahigh intensity regime.

The choice of a classical model is motivated by the success of such models for strong field physics at describing important rescattering [26], harmonic emission [27], and multi-electron [28] physics in strong fields. Classical models have also been instrumental for understanding multi-electron excitations [29, 30], relativistic effects [8], and the role of magnetic fields for Rydberg atoms in strong fields [31]. The classical approach compliments ongoing work to address the quantum aspects of the ultrastrong field. If B_{laser} can be shown to have no significant perturbation on the ground state, the insights gained from strong field rescattering and electron collision studies on atoms and ions at accelerators may be directly applied to multi-electron excitation in the ultrastrong field. New challenges to address in ultrastrong field, multi-electron excitations include recollision energies greater than 100 eV and the excitation of four or even six electrons, and inner shell excitations. While much of the

focus has been on extremes of the ultrastrong field (where $a_0 > 2$, intensities greater than 10^{19} W/cm² at 800 nm) there is a significant amount of unknown atomic physics in the region where the photoelectron energy scale is from 5×10^3 to 5×10^5 eV, $0.2 < a_0 < 2$, (or 10^{17} to 10^{19} W/cm² at 800 nm) and classical models may be very reliable.

3.2 Classical Model

The method used in these studies has been described in detail [14]. Briefly, the atom is treated classically as a relativistic, single-electron, hydrogen-like system. Bound states are given the energies $E_n = -Z^2/2n^2$ where n is the ‘principal quantum number’ and Z is the atomic number. The angular momentum is varied from 0 to $n\hbar$. The Kepler orbits with a period, $\tau_K \cong 2\pi(n^3/Z^2)$, are numerically integrated. For the initial position and momentum of the electrons we follow the method given in [32]. The Kepler orbits (figure 1(a)) are defined by a set of five parameters: inclination of the orbit (i), which is the polar angle from z (synonymously k_{laser}) into the normal for the orbit plane $\mathbf{n}$; the line of nodes longitude (Ω) from x to the intersection of the orbit plane with the $x - y$ plane containing E_{laser} and B_{laser} . Additional initial coordinates include the mean anomaly (α); orbit eccentricity (ϵ); and angle ω_A from the line of nodes to the periapsis. These parameters are generated randomly with equal probability to create three-dimensional (3D) ensembles for the electron and can take on the values,

$$0 \leq \alpha \leq 2\pi, 0 \leq \epsilon^2 \leq 1, -1 \leq \cos i \leq 1,$$

$$0 \leq \Omega \leq 2\pi, 0 \leq \omega_A \leq 2\pi.$$

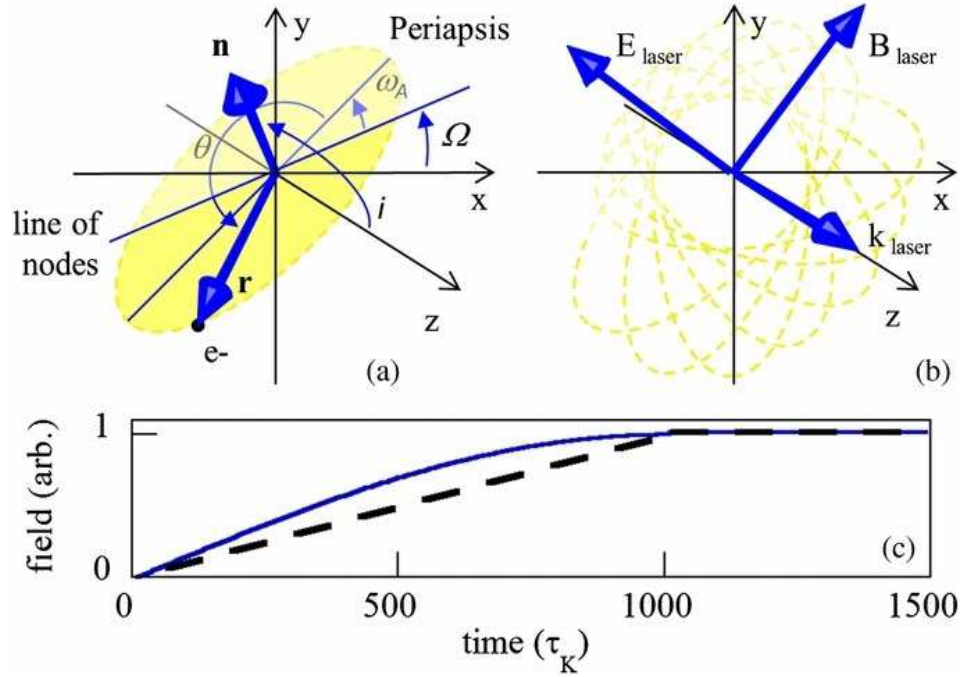


Figure 3.1 Definition of Kepler orbit initial coordinates (a) for a single orbit in a plane perpendicular to $\mathbf{n}$. The inclination of the orbit from the z -axis (also the direction of $\mathbf{k}_{\text{laser}}$ for these studies) is the polar angle i . The node line from the intersection of the orbit plane and the x - y plane is given by the azimuthal angle Ω , ω_A is the rotation of the periastris from the node line and the angle from the periastris to $\mathbf{r}$, where $\mathbf{r}$ is from the nucleus to the electron, is given by θ . E_{laser} and B_{laser} are in the x - y plane in these studies with $\mathbf{k}_{\text{laser}}$ along positive z . The light is circularly polarized. Example directions for E_{laser} and B_{laser} ramps used for the pulse envelope of the E_{laser} , B_{laser} fields are shown in (c) for a linear ramp (dash) and sinusoidal (solid).

In these calculations the initial position within the orbit, θ , or initial angle to the periastris, ω_A , do not affect the dynamics; the field changes slowly over many Kepler orbits and the importance of the launch phase in the orbit is diminished. As the electron interacts with a soft core potential [33, 34], $-Ze/\sqrt{r^2 + \delta}$, and external field,

E_{laser} , B_{laser} , the values of position and momenta as functions of time are generated by integrating Hamilton's equations of motion:

$$\frac{dp_x}{dt} = \frac{-Ze^2x}{(r^2 + \delta)^{3/2}} - eE_{\text{laser}}(\vec{r}, t)_x \left[1 - \frac{p_z}{\sqrt{p^2 + m_0^2 c^2}} \right] \quad (3.1)$$

$$\frac{dp_y}{dt} = \frac{-Ze^2y}{(r^2 + \delta)^{3/2}} - eE_{\text{laser}}(\vec{r}, t)_y \left[1 - \frac{p_z}{\sqrt{p^2 + m_0^2 c^2}} \right] \quad (3.2)$$

$$\frac{dp_z}{dt} = \frac{-Ze^2z}{(r^2 + \delta)^{3/2}} - \frac{eE_{\text{laser}}(\vec{r}, t)_x p_x + eE_{\text{laser}}(\vec{r}, t)_y p_y}{\sqrt{p^2 + m_0^2 c^2}} \quad (3.3)$$

$$\frac{dx}{dt} = \frac{p_x c}{\sqrt{p^2 + m_0^2 c^2}} \quad (3.4)$$

$$\frac{dy}{dt} = \frac{p_y c}{\sqrt{p^2 + m_0^2 c^2}} \quad (3.5)$$

$$\frac{dz}{dt} = \frac{p_z c}{\sqrt{p^2 + m_0^2 c^2}} \quad (3.6)$$

where c is the speed of light, δ is our soft core parameter, Z is the atomic number, m_0 is the rest mass, $r = \sqrt{x^2 + y^2 + z^2}$ and $p = \sqrt{p_x^2 + p_y^2 + p_z^2}$. When only the dipole approximation is considered, the Lorentz force terms from B_{laser} are zero, i.e. $p_z/\sqrt{p^2 + m_0^2 c^2}$ in equations (1) and (2), and $E_{\text{laser}}(\vec{r}, t)$ terms in equation (3) are dropped. The value of δ is chosen to keep the energy of the electron less than 5 MeV at $r = 0$. We also verified using a Coulomb potential with at $r = 0$ so their energy would exceed 5 MeV) does not change the results presented here. The envelope for the pulsed field used in these studies is shown in figure 3.1(c). The two shapes of the ramp shown (linear and 0 to $\Pi/2$ of a sin function) has no effect on the results presented

here. Spin is neglected as it does not affect ionization at the level of 5% until $Z = 60$ [35].

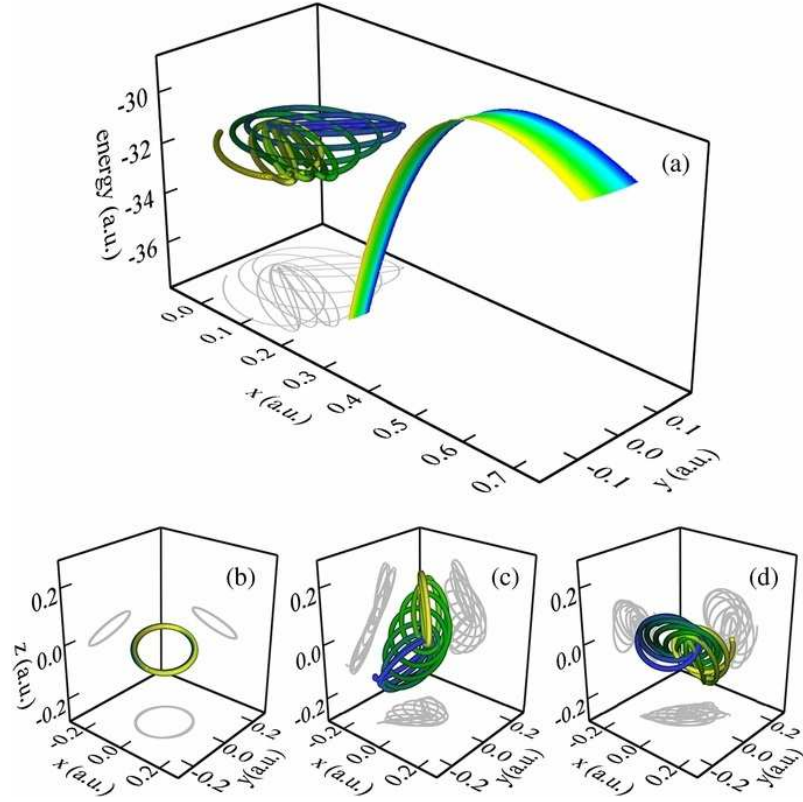


Figure 3.2 Trajectory plot (a) of $Z = 8$ ($IP = 32$ au) for ten Kepler orbits projected in the x - y plane shown with the minimum in the effective Coulomb plus external circularly polarized electric potential. The laser field is rotating in the x - y plane with a magnitude $|E| = 16$ au and wavelength of 7370 au (0.9×10^{19} W/cm² and 390 nm, respectively). Time is mapped in the plot with the light yellow ($t = 0$) to dark blue ($t = 1$ au) color scale. An example of the influence of the external field on ten Kepler orbits ($Z = 8$, $n = 1$) is shown in x , y , z spatial trajectory plots for the field free case (b), and ultrastrong field case ($|E| = 30$ au and wavelength of 274 au) with linear polarization along the x -axis (c), and circular polarization (d). The orbits shown are after the system has been allowed to interact with the field for four optical cycles. Shadow projections are shown.

The accuracy of the solutions can be gauged by the conservation of energy in the absence of the external radiation field. In this case, the energy deviates by less than 10^{-11} after 200 Kepler orbits for any state used in the calculation. Our studies here are limited to the ‘quasistatic’ regime where the laser field period is 100 times the Kepler orbit time and the laser field ‘slowly’ rotates around the nucleus. For reference, we also give a few examples in the ‘static’ field case where E_{laser} is fixed along x and B_{laser} is fixed along y . For intensities of order 10^{19} W/cm² ionizing states are typically bound by ~ 500 eV with Kepler orbit times (e.g. $n = 2$, $Z = 12$) of $\sim 1.3 \times 10^{-18}$ s. A laser field in this ‘100 $\times$ ’ quasistatic limit would then have a wavelength longer than 39 nm. In general, we find the results presented here are equally valid for a range of frequencies $20 < \omega_{\text{Kepler}}/\omega_{\text{laser}} < \infty$.

3.3 Results

At the point in space where laser and Coulomb fields are equal, an effective barrier forms for the bound electron, the minimum of which is along E_{laser} . The minimum in this barrier is illustrated in figure 3.2(a) for a laser field ($\lambda = 390$ nm) as it rotates around a $Z = 8$ nuclear charge in the $x - y$ plane over a time period of ten Kepler orbits, which are also shown projected into the $x - y$ plane. From the figure, one can see the rotation is negligible for a single orbit, but may be comparable to the orbit precession. The rotation in the field seen in Figure 3.2(a) over ten Kepler orbits is 0.12 Rad. In figure 3.2, time is shown by the color scale imposed on the ten orbits and the effective barrier. Examples of ten orbits in 3D, (x, y, z) space are shown in figure 3.2(b)–(d). Figure 3.2(b) is the trajectory plot with no external laser field. The precession due to relativity is very small for the given circular orbit; ten orbits appear

as essentially one that is fixed in space. Figure 3.2(c) is the same initial trajectory but with a linearly polarized E_{laser} field. While the external field is essentially static, the changes in the direction of the orbit are significant from orbit to orbit. Nevertheless, aspects of the original orbit are still discernible in the $x - y$, $y - z$, and $x - z$ projections. The impact of the circularly polarized field can be seen in figure 3.2(d). The constantly changing direction of the circular polarization leads to a complicated trajectory. To help reveal more exactly how the fields change the dynamics and determine the role of B_{laser} we use Poincaré plots (figures 3.3, 3.4), Fourier transforms of the trajectory (figure 3.5) and the intensity dependence for classical ionization with CP light (figure 3.6). We begin when the interaction is primarily with the bound state and ionization is less than a few per cent, e.g. 1.8×10^{18} to 6.0×10^{19} W/cm² for a $Z = 10$, $n = 1$ state. We then proceed to the impact of B_{laser} in the strongly ionizing case, e.g. 4.9×10^{20} W/cm² for the same $Z = 10$, $n = 1$ state.

Poincaré plots preserve essential properties of periodic or quasiperiodic orbits while reducing the dimensionality of the analysis. In the Poincaré plot, a point is plotted every time a periodic orbit returns to a specified plane. They have proven useful in the past, for example, with the analysis of chaotic motion in Rydberg orbits when an external magnetic field is introduced [36]. The onset of this chaotic motion is associated with a loss of clear repeatable orbits in the Poincaré plots with ‘stochastic’ motion indicated by a uniform and random distribution within the allowable Poincaré region. The two Poincaré plot dimensions (p_x , x recorded in the $z = 0$ plane and p_z , z recorded in the $x = 0$ plane) presented here are chosen because E_{laser} is along x and the Lorentz force from B_{laser} is along z . We begin in figure 3.3 with the (p_x , x) Poincaré plot of five trajectories ($Z = 10$, $n = 1$) with varying angular momentum

shown over 1000 Kepler orbits and no external field (figure 3.3(a)). The angular momentum of the five different orbits is shown with the colour scale. The periodic nature of the orbit can be seen as the trajectory retraces the closed forms in figure 3.3(a) over the 1000 orbits. In the following we consider the Poincaré plots with a static external field ($\omega_{\text{Kepler}}/\omega_{\text{laser}} \rightarrow \infty$, figure 3.3(b), (d), (f)) and a quasistatic field ($\omega_{\text{Kepler}}/\omega_{\text{laser}} = 100$, figure 3.3(c), (e), (g)).

In figure 3.3(b), a static external field of strength $E_{\text{laser}} = 41$ au (6×10^{19} W/cm² or $\sim 44\%$ of the critical field) is added to the same states used in figure 3.3(a). The 1000 orbits shown are in the flat portion of the pulse envelope (figure 3.1(c)) after the ramp. The addition of the field forces the electron trajectory towards $-x$ as expected and destroys the clear periodicity in the Poincaré plot. The only remaining character from the initial trajectory is the angular momentum. The addition of the static B_{laser} , shown in figure 3.3(d), reveals an additional shift to $-x$ (highlighted in figure 3.3(d) that has also been seen in configuration space studies and described as an increase in the polarization of the bound state. Overall a comparison of the Poincaré with E_{laser} in figure 3.3(b) and the full E_{laser} and B_{laser} field in figure 3.3(d) shows many similarities. The reason behind this is revealed in figure 3.3(f) with only B_{laser} applied to the atom (i.e. $E_{\text{laser}} = 0$). The changes from the external ‘field-free’ Poincaré plot (figure 3.3(a)) are only slight as the Lorentz force from the external magnetic field perturbs, but does not change the orbits to the point of being unrecognizable and chaotic as seen in figure 3.3(b).

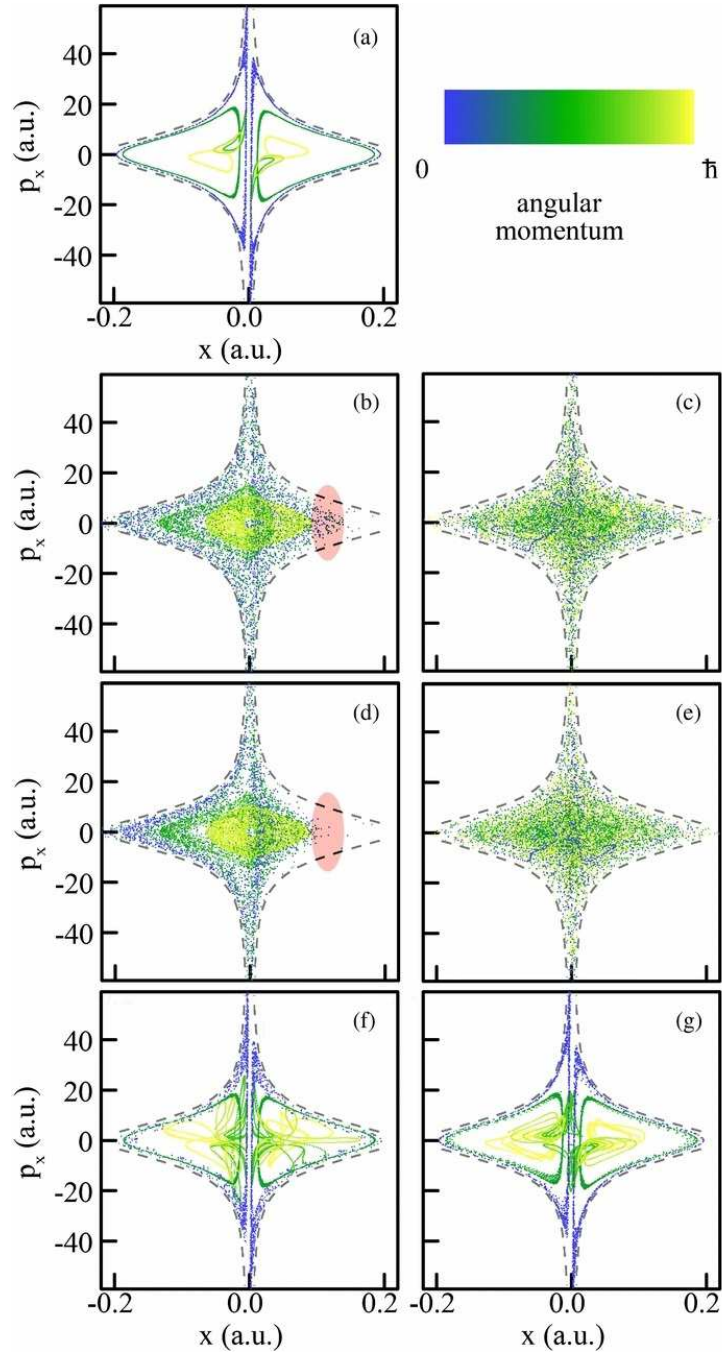


Figure 3.3 Poincaré (p_x, x) plots for five sample trajectories of the $Z = 10, n = 1$ state ($IP = 1360$ eV, angular momentum $0.27\hbar, 0.50\hbar, 0.62\hbar, 0.76\hbar, 0.82\hbar$) shown for 1000 Kepler orbits. The state with no external field ($E_{\text{laser}} = B_{\text{laser}} = 0$) is shown in (a). The angular momentum for the orbit is indicated by the false color scale. The Poincaré plots with an intense, electric field ($E_{\text{laser}} = 41$ au along x , 6×10^{19} W/cm²) are shown in (b) for a static field and for a laser frequency $1/100$ that of the Kepler orbit frequency (c). The case with both E_{laser} and B_{laser} at 6×10^{19} W/cm² is shown in (d) for the static case and in (e) for $\omega_{\text{Kepler}}/\omega_{\text{laser}} = 100$. The Poincaré plots with only B_{laser} in the static and quasistatic case ($\omega_{\text{Kepler}}/\omega_{\text{laser}} = 100$) are shown in (f) and (g), respectively. Two highlighted regions in (b) and (d) at $(p_x = 0, x = 0.1)$ show where differences in the trajectories with and without B_{laser} can be seen. A dashed line is superimposed on the graphs to show the maximum extent of the possible values in the Poincaré plot.

In the quasistatic case ($\omega_{\text{Kepler}}/\omega_{\text{laser}} = 100$ shown in figure 3.3(c, e), the E_{laser} field significantly changes the trajectory into a motion on the Poincaré plot traditionally characterized as chaotic. The limits to the trajectories (indicated with dashed lines on figure 3) are clearly explored with five trajectories over 1000 orbits. For perspective with the current ultrastrong laser experiments [37], one laser period of a Ti:sapphire laser at $\lambda = 800$ nm is 1750 Kepler orbits for the $Z = 10, n = 1$ state. The inclusion of B_{laser} (Fig. 3.3(e)) only modifies the trajectories seen with E_{laser} , figure 3.3(c). In figure 3.3(g) we show the Poincaré plot with only B_{laser} . The changes per orbit for the CP, quasistatic case are even smaller than for the static B_{laser} field. One may surmise this is due to the fact the field direction reverses itself every half cycle, undoing the change in the trajectory from the previous half cycle. The initial structure seen in figure 3.3(a) is still recognizable in the orbits of figure 3.3(g).

In figure 3.4 we show the companion (p_z, z) Poincaré plots of the five orbits shown in figure 3.3. Similar trends as a function of the external field are observed in figure 3.3 and figure 3.4. The addition of the B_{laser} Lorentz force gives the changes shown in figure 3.4(d), (e). Like figure 3.3(d), (e) they are noticeable only on close inspection of the figure. In the (p_z, z) figure 3.4(f), (g) plots, the impact of the Lorentz force from the CP B_{laser} is more noticeable than in figure 3.3(f), (g) and leads to larger changes to the trajectories.

To gain insight into the energy changes on the bound trajectories due to the E_{laser} and B_{laser} external fields and the chaotic motion, we have chosen to Fourier transform the trajectory ensembles. The initial random launch conditions for the trajectory ensembles are the same with or without the field to allow the best comparisons. The Fourier transforms of $r(t)$ are done on 4000 trajectories over a time period of four optical cycles (512 Kepler orbits). When viewed as an ensemble of the 4000 trajectories analyzed, one gains a broader view of the dynamics.

We first show the sum average of the spectral amplitude from the Fourier transform for two static electric fields of 7.4 and 41 au in figure 3.5(a) with a $Z = 10$, $n = 1$ state. Without the external field the Fourier transform is a single frequency at the Kepler orbit of 15.9 au. With the static external field the frequency of the trajectories is split into two peaks with the magnitude of the splitting dependent on the laser intensity. As shown in the figure, the frequency splitting can be several atomic units even for a 7.4 au field, which corresponds to an intensity only 1% of the critical intensity to classically ionize a $Z = 10$, $n = 1$ bound state.

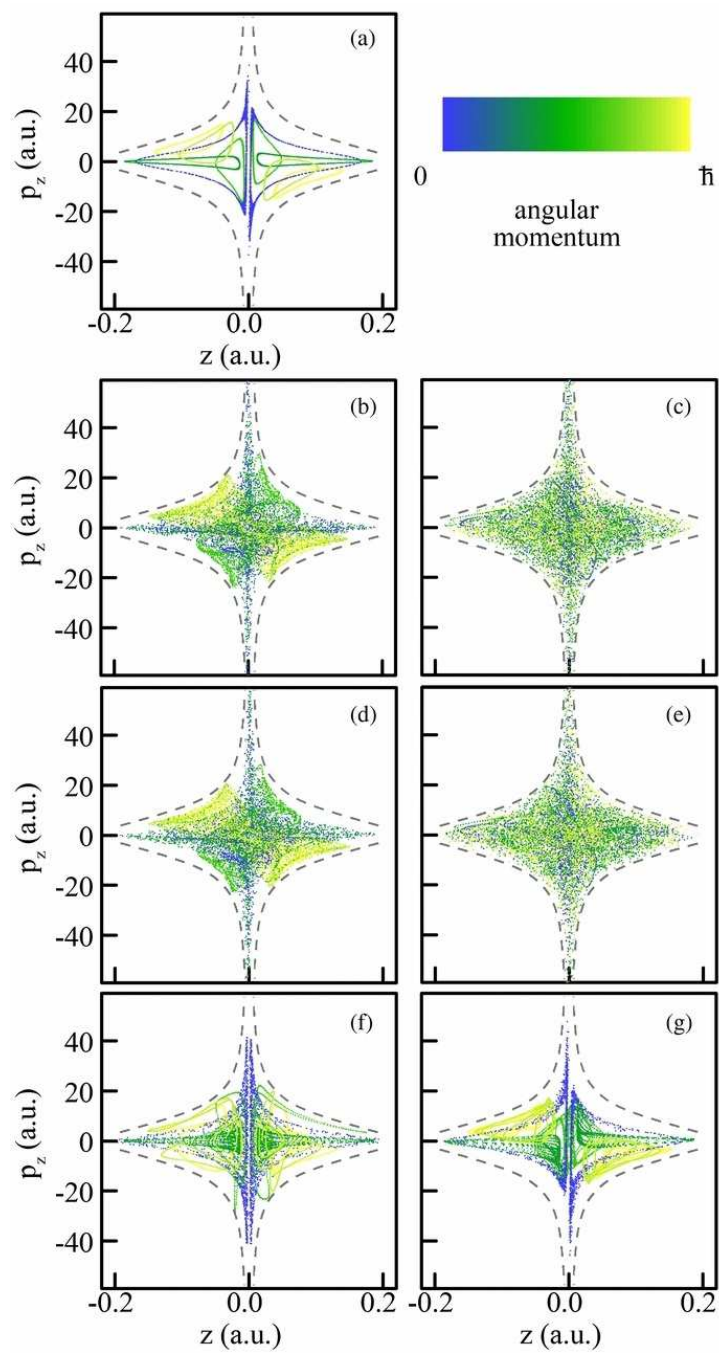


Figure 3.4 Poincaré (p_z, z) plots for five sample trajectories of the $Z = 10, IP = 1360$ eV (angular momentum $0.27\hbar, 0.50\hbar, 0.62\hbar, 0.76\hbar, 0.82\hbar$) are shown in over 1000 Kepler orbits. The Poincaré plots with an intense, electric field (6×10^{19} W/cm²) are shown in (b) for a static field and in (c) for a laser frequency 1/100 that of the Kepler orbit frequency, E_{laser} is along x . The case with both E_{laser} and B_{laser} at 6×10^{19} W/cm² is shown in (d) for the static case and in (e) for $\omega_{\text{Kepler}}/\omega_{\text{laser}} = 100$. The Poincaré plots with only B_{laser} in the static case and $\omega_{\text{Kepler}}/\omega_{\text{laser}} = 100$ case are shown in (f) and (g), respectively. A dashed line is superimposed on the graphs to show the maximum extent of the possible values in the Poincaré plot.

The introduction of the ultrastrong laser field, E_{laser} , moves the orbit from simple to chaotic. The net result is that any one trajectory over several hundred Kepler orbits will explore very different regions of space and have a widely varying $r(t)$ function. Consistent with figures 3.3, 3.4, inspection of the orbits reveals changes in the FT spectral content can be seen even between adjacent sequences of consecutive orbits as the trajectory is affected by E_{laser} . An analogy for this change can be compared to coherence, where the disruption of the motion by the ultrastrong field creates the spectral width in the FT of the Kepler orbit, $r(t)$. A case by case inspection of the trajectories reveals what sets the coherence in the trajectory is a small impact parameter, hard collision with the nucleus. The introduction of the magnetic field does not significantly alter the frequency spectrum for the trajectories as can be seen in figure 3.5(b), (c) where the results with and without B_{laser} overlap at 1.9×10^{18} W/cm² (b) and 6×10^{19} W/cm² (c).

The exact structure of the spectrum for the FT also depends on which component of the motion relative to the field is analyzed, e.g. $x(t), z(t), r(t)$. We have chosen $r(t)$ for this presentation.

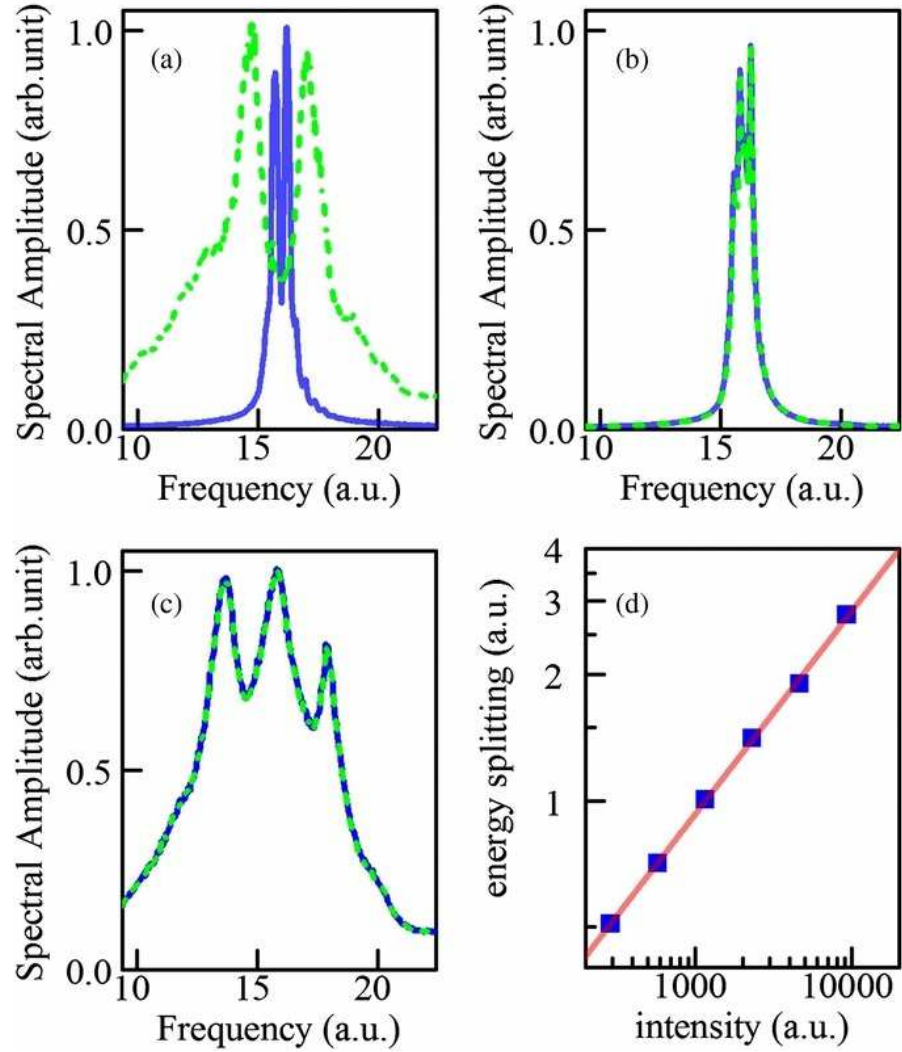


Figure 3.5 Spectral amplitude from the Fourier transform of the trajectories, i.e. $|r(t)|$. Shown in (a) are the results for two static electric fields of 7.3 au of field (solid blue) and 41 au of field (dash green), $1.9 \times 10^{18} \text{ W/cm}^2$ and $6 \times 10^{19} \text{ W/cm}^2$, respectively. The results for the circular polarized field with the electric field only (solid blue) and both the electric and magnetic fields (dash green) are shown in (b) at $1.9 \times 10^{18} \text{ W/cm}^2$ and in (c) at $6 \times 10^{19} \text{ W/cm}^2$. The energy splitting of the $Z = 10$, $n = 1$ bound state (taken from the analyzed Fourier transforms) as a function of the external field intensity is shown in (d) with a linear fit, slope 0.49.

The width of the peak splitting is consistent between the different components although the shape differs. For CP light, the splitting for an analysis of $r(t)$ shows three peaks from the orientation of the electric dipole relative to the rotating field. The shift of the upper and lower peak was analyzed and accurately converted to a binding energy with a $1/|r|$ potential. This energy splitting for the $Z = 10, n = 1$ bound state as a function of intensity is shown in figure 3.5(d) with a linear fit where the energy split equals 0.49 times the intensity. The FT of the trajectory indicates that while B_{laser} may deflect the electron in the bound state, it does not significantly affect the energy of the bound states.

When dealing with thousands of trajectories in ensembles, an analysis of the motion can be approached with configuration space plots [14]. In the configuration space plots, the occurrence of a trajectory at a space coordinate is summed over the ensemble. When divided by the total number of trajectories, the technique gives a classical particle density that can be compared to the quantum electron probability density [38]. In linearly polarized fields, earlier work with configuration space plots show B_{laser} can slightly increase the induced dipole. The final analysis of the role of the ultrastrong external magnetic field is the ionization probability with and without the magnetic field and any associated changes to the bound state configuration space. Figure 3.6(a) gives the ionization probability for four states that ionize from 10^{14} W/cm² (for $Z = 1, n = 1$) to 10^{22} W/cm² (for $Z = 20, n = 1$). In the nonrelativistic regime $< 10^{16}$ W/cm² (e.g. $Z = 1, n = 1$) there is no difference in the atomic ionization response with or without the magnetic field component of the external radiation field; the ionization rates from the electric field only (solid line in figure 3.6(a)) are unchanged when including the magnetic field (dash line in figure 3.6(a)).

As one exceeds 10^{18} W/cm^2 , the ionization rates begin to be affected by a CP B_{laser} field, most notably in figure 3.6(a) when the interaction is in the strongly ionizing case where ionization is between 10% and 100%. Ionization with only the electric field increases smoothly from threshold at the critical field, where the laser field equals the Coulomb field, to complete ionization within the laser pulse. Including the laser magnetic field was found to (1) reduce ionization and (2) introduce a two-step aspect to the ionization.

In the two-step process, initially at lower fields where the total amount of ionization is less than a few per cent, ionization with E_{laser} and B_{laser} is the same as that with E_{laser} only. When the interaction becomes strongly ionizing with ionization $> 10\%$ ionization with B_{laser} then ‘stalls’ until eventually, at a field two to three times that required for E_{laser} only, ionization resumes to saturation (100% ionization). This feature is robust from 10^{19} to 10^{22} W/cm^2 in the calculations. Looking at the ionization within a single pulse reveals these two steps can also be identified as a function of time as ionization occurring early in the pulse, where the intensity is lower and there is no impact to including B_{laser} , and then later in the pulse where there is more ionization with E_{laser} only.

We have highlighted in grey one such region in figure 3.6(a) where the E_{laser} only dipole response is 81% ionized and the response with B_{laser} is suppressed and only 50% ionized at the end of the pulse. The configuration space plot for the bound trajectories in the E_{laser} only case is shown in figure 3.6(b) for a pulse with a peak intensity of $4.9 \times 10^{20} \text{ W/cm}^2$, the highlighted region in figure 3.6(a). The configuration plot with E_{laser} only is shown with the ground state configuration plot subtracted out so that only changes are shown in figure 3.6(b) (the spatial integration

of figure 3.6(b) is zero). The configuration space plot is a snapshot of the trajectory ensemble ‘classical probability’ over 1 Kepler orbit time. The instantaneous CP E_{laser} direction when the configuration space plots were evaluated is superimposed on the $x - y$ plane. In terms of the pulse (figure 3.1(c)), this snapshot was taken at 450 Kepler orbits, which is about half-way in the intensity on the rising edge of the pulse. This is the time in the pulse when the ionization occurring early in the pulse is greatest. The snapshot configuration space plot under these conditions reveals the trajectories that ionize early in the pulse at lower intensity. Including the B_{laser} field has no effect on configuration space snapshot for this early ionization shown in figure 3.6(b).

Inspecting first the $x - y$ plane in figure 3.6(b), one can see the ring of depletion for trajectories near edge of the potential, $r \sim 0.2$ au, due to ionization near the barrier. In addition, the instantaneous force on the electron away from the electric field can be seen in the slight displacement towards positive x , negative y at this phase of the field. In the $x - z$, $y - z$ planes one also sees the ring of depletion and instantaneous nudge towards $+x$, $-y$ and a new feature along z . As the field rotates in the $x - y$ plane, trajectories aligned in this plane are depleted. This well-known effect is understood quantum mechanically as the preferential ionization of $m = 0$ states by tunneling. The states oriented out of the plane of ionization ionize at higher fields and can be seen to persist even in the electric field only response of figure 3.6(b).

The configuration plot for the full E_{laser} and B_{laser} field is shown in figure 3.6(c). This snapshot of the configuration space is taken for 1 Kepler orbit at the peak in the pulse, a time of 1250 Kepler orbits (see figure 3.1(c)). The instantaneous CP E_{laser} and B_{laser} direction, which is the same CP field phase as for figure 3.6(b), is superimposed on the $x - y$ plane.

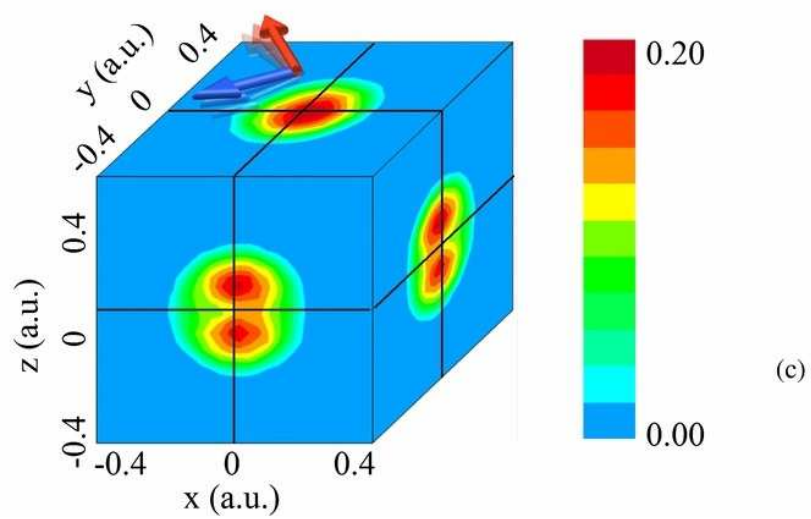
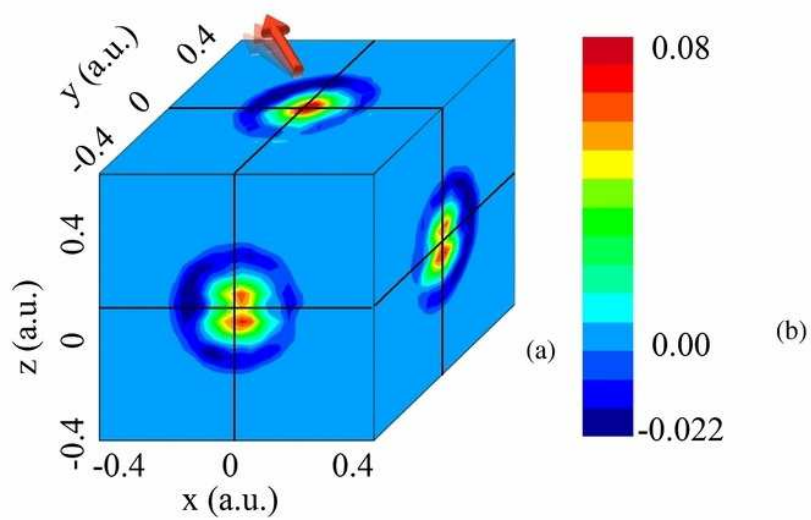
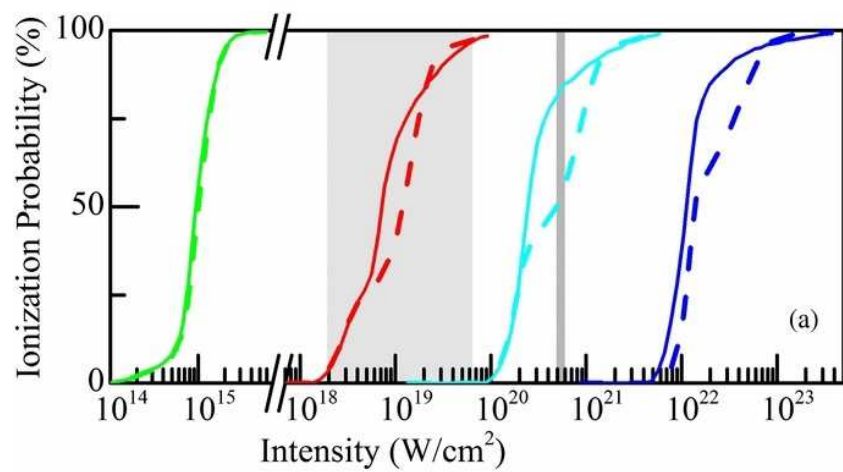


Figure 3.6 Ionization probability as a function of intensity (a) for circular polarized light with the electric field only (solid) and the full electric and magnetic field (dash). From left to right (low to high intensity) the states ionized are Z, n : 1, 1 (green); 5, 1 (red); 10, 1 (cyan); 20, 1 (blue). The region studied in figures 3–5 where ionization of the $Z = 10, n = 1$ is weak, i.e. less than 3%, is highlighted in light grey from $1.9 \times 10^{18} \text{ W/cm}^2$ to $6 \times 10^{19} \text{ W/cm}^2$. The configuration space plot of the trajectories for the $Z = 10, n = 1$ state with the electric field only at $4.9 \times 10^{20} \text{ W/cm}^2$ (the highly ionized region highlighted in dark grey for part (a)) is shown in (b) projected into the x - y , y - z , z - x planes. To show the changes due to the electric field, the field free ground state population has been subtracted out, showing a depletion of the trajectories near the ionization barrier that is rotating in the x - y plane. The instantaneous electric field value for the configuration plot shown is indicated with a red arrow. The configuration space plot with both electric and magnetic fields for the same 10,1 state at $4.9 \times 10^{20} \text{ W/cm}^2$ is shown in (c). The electric field only configuration space has been subtracted to clarify the role of the magnetic field. The field directions for the electric field (red) and magnetic field (blue) are indicated for the time of the configuration space plot shown. The false color shown is normalized to one for the central peak in the field free distribution.

This time is at the second step in the ionization during the pulse. Here B_{laser} has a clear impact on the ionization. When the pulse is finished, this difference is an ionization state of 81% with E_{laser} and 50% with the full $E_{\text{laser}}, B_{\text{laser}}$ field. The configuration plot with E_{laser} and B_{laser} is shown with the E_{laser} only configuration plot subtracted out. The overall positive value of the configuration space plot in figure 3.6(c) is a result of the 50% to 81% difference in the ionization shown in figure 3.6(a); with E_{laser} and B_{laser} the bound state population is greater since it is less likely to ionize. The spatially integrated configuration space of figure 3.6(c) equals the difference between the 50% to 81% curves in the highlighted region of figure 3.6(a). By inspecting figure 3.6(c), one can see the inclusion of B_{laser} increases the number of bound state trajectories out of the $x - y$ plane. The increase in trajectories aligned

along z makes ionization more difficult when CP B_{laser} is included. The alignment gives the difference between the E_{laser} and B_{laser} cases observed in figure 3.6(a). The net impact of the Lorentz force with a CP, ultraintense field then appears to be an alignment of the trajectories along z and a stabilization against ionization.

3.4 Conclusion

We present theory results for the ionization of atoms in strong to ultrastrong circular polarized (CP) fields. A relativistic, three-dimensional trajectory ensemble method is used to account for the role of the laser magnetic field. The results show trajectories in ultrastrong fields are best characterized as chaotic when analyzed by Poincaré plots. Including the laser magnetic field can be observed to slightly change the distributions within these plots. A Fourier analysis of the trajectories does not reveal any significant change in the energy of the bound states due to the laser magnetic field as the energy shifts are linear in intensity well into the ultrastrong field regime. A result of the magnetic field was a stabilization of the atomic bound state as the Lorentz force preferentially aligned trajectories along z , perpendicular to the rotating $x - y$ plane of the field where ionization occurs. The results indicate relativistic effects in ultrastrong field ionization may most easily seen with CP fields in experiments with ultrahigh intensities and occur at lower intensities than expected for LP light.

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Chapter 4

PHOTOELECTRON ENERGY SPECTRA FROM ELASTIC RESCATTERING IN ULTRA STRONG LASER FIELDS: A RELATIVISTIC EXTENSION OF THE THREE-STEP MODEL

4.1 Introduction

High strength laser fields can ionize the outer, least tightly bound electron from atoms and molecules by overcoming the binding nuclear Coulomb field. Fields of this strength (0.17 a.u., intensities of 10^{15} W/cm²) also dominate photoelectron dynamics and the oscillating laser field can force the photoelectron to return and “rescatter” with the parent ion [1]. Over the past twenty years, strong field ionization and rescattering has been used to measure electron dynamics [2], collisionally excite multiple electrons [3], generate coherent attosecond x-ray light [4], and perform molecular tomography [5]. For optical frequency lasers, these phenomena occur on energy scales (e.g., the ponderomotive [1] or “quiver” energy $U_p = e^2|E|^2/(4m\omega^2)$ for an electron charge $-e$ in an oscillating electric field E , frequency ω) that are less than 1% of the electron rest mass m . Hence, the interaction is safely described nonrelativistically using the dipole approximation $\vec{E} \cdot \vec{r}$ in the length gauge. Models for these interactions [6–11] range from fully quantum one-electron [12] or multielectron treatments [13] to insightful one-electron [14] and multielectron classical theories [15,16].

When the laser field is increased, more tightly bound electrons ionize; up to 26 electrons have been ionized for 24 a.u. laser fields (2×10^{19} W/cm²) [17]. In these ultrastrong fields, relativistic dynamics [18] are important and photoelectron energies

can exceed several times the electron rest mass [19]. The laser field may no longer be simply approximated and the laser magnetic field B is required [20]. Research with mid-IR wavelength lasers and keV energy, attosecond XUV pulses are also beginning to venture into the ultrastrong field frontier [21]. Theoretical underpinnings common to strong field models fail in ultrastrong fields. New approaches are required to overcome the numerous challenges such as three-dimensional spatial dynamics that extend relativistically from an atomic unit of length to that of an optical wavelength in a femtosecond. Theory treatments have ranged from one-electron time-dependent Dirac and Klein-Gordon solutions [22] to fully classical [23–26]. Recent calculations have addressed the fundamental physics including the role of electron spin [27]. At this time, the theoretical approaches have reached a point where it is possible to compare with experimental results in a quantitative way. Such comparisons will make it possible to identify complex dynamics and multielectron physics.

4.2 Relativistic, Three-step Recollision Model

An emerging technique which accurately captures much of the physics and can be useful when comparing to experimental results involves treating interactions such as ionization [10] or radiation [28] quantum mechanically and propagation of the photoelectron in the field classically when the electron de-Broglie wavelength is much smaller than the drive wavelength. Ionization and propagation components of this model compare favorably with recent ultrastrong field experiments [17]. Monte Carlo trajectory ensembles in the model capture essential quantum aspects of the electron [23,24] and such semiclassical approaches have been compared to full quantum solutions with the Dirac equation [29]. Adding elastic rescattering is a natural extension of the model and the approach has advantages in its connection to the well-

known three-step model [30]. Perhaps more important is the ability to include temporal and spatial integrated experimental conditions. Relativistic dynamics and a focal geometry inherent to all ultrastrong field experiments lead to complicated field accelerations that depend on the position and time in the laser field. Rendering a result for comparison to experimental result has involved, for example [17], integration over 10^{-3} m distances and 10^{-13} s. There is also a natural extension of the technique to plasma physics in ultrastrong fields, which utilizes classical particle-in-cell methods.

We report photoionization and fully relativistic elastic scattering [31] in ultrastrong fields. Key questions addressed include the final photoelectron energies as they are affected by elastic rescattering [32], atomic scattering potentials, the laser magnetic field, and relativistic effects. The work helps quantify the changes in rescattering as one moves from the strong field to ultrastrong field. After the ionization process itself, elastic scattering is the primary mechanism by which the field energy is transferred to atomic and ion systems. Using hydrogenlike and screened atomic scattering potentials for noble gas species, we are able to model elastic rescattering as a function of intensity. Magnetic deflection effects [20] are observed beyond 6×10^{16} W/cm² when the rescattering parameter [33] $[\Gamma_r = U_p^{\frac{3}{2}} V_{IP}^{\frac{1}{2}} / (3c^2 \omega)]$ for ionization from a binding energy V_{IP} indicates that the Lorentz deflection of the photoelectron equals its wave function spread. Relativistic scattering enhancements are observed for intensities beyond 2×10^{18} W/cm² where the classical field nonlinearity parameter $[a_0 = e|E|/(\omega mc)]$ signifies that the electron motion is relativistic within a single field cycle. The extreme relativistic regime ($a_0 > 10$) [18] lies beyond the scope of this work. Lasers that promise to achieve extreme relativistic intensities [34] are under construction. Atomic units are used throughout the work

except as noted where conventional units (e.g., W/cm²) are used for comparison to other work.

4.2.1 Ionization

Our calculations use linearly polarized light, $\vec{E} = E_0 \sin(kz - \omega t) \exp[-(t - z/c)^2/\sigma^2] \hat{x}$ with a pulse duration $\sigma = 34$ fs and carrier wavelength $\lambda = \frac{2\pi}{k} = 800$ nm. When considering the full field $\vec{B} = |E|/c \hat{y}$. In the dipole approximation we set $\vec{B} = 0$. This plane wave is used for all cases except as noted for comparison with data at 10^{19} W/cm² where we adopt the experimental focus. Ionization is calculated using the Ammosov-Delone-Krainov (ADK) rate [35] for hydrogenlike 1s states and the least tightly bound electron for the noble gas ions. The ionization curves for He⁺ and Ar⁸⁺ are shown in Fig. 1 along with the laser electric field. To keep the comparisons across species similar, E_0 is chosen so that ionization reaches 90% by the end of the pulse. Such a treatment of the ionization rate is believed to be accurate within a factor of 2. Relativistic and Coulomb factors [36] lead to corrections in the rate of less than 25% for the cases presented here.

4.2.2 Continuum Dynamics

After ionization, a Gaussian Monte Carlo ensemble electron “wave packet” is launched in the continuum with a quantum spread from the initial ionization width [33] and subsequent propagation. The deflection due to B is calculated using the Lorentz force on the photoelectron, $\vec{F} = -e\vec{E} - e\vec{v} \times \vec{B}$. As the electron interacts with a soft core potential, $Ze/\sqrt{r^2 + \delta}$, and external field, the position and momenta for the trajectories within the Monte Carlo ensembles are generated by integrating Hamilton’s equations of motion:

$$\frac{dp_x}{dt} = \frac{-Ze^2x}{(r^2 + \delta)^{3/2}} - eE_x \left[1 - \frac{p_z}{\sqrt{p^2 + m^2c^2}} \right] \quad (4.1)$$

$$\frac{dp_y}{dt} = \frac{-Ze^2y}{(r^2 + \delta)^{3/2}} \quad (4.2)$$

$$\frac{dp_z}{dt} = \frac{-Ze^2z}{(r^2 + \delta)^{3/2}} - \frac{eE_x p_x}{\sqrt{p^2 + m^2c^2}} \quad (4.3)$$

$$\frac{dx}{dt} = \frac{p_x c}{\sqrt{p^2 + m^2c^2}} \quad (4.4)$$

$$\frac{dy}{dt} = \frac{p_y c}{\sqrt{p^2 + m^2c^2}} \quad (4.5)$$

$$\frac{dz}{dt} = \frac{p_z c}{\sqrt{p^2 + m^2c^2}} \quad (4.6)$$

where c is the speed of light, δ is the soft-core parameter (typically $\delta = 0.5$), Z is the atomic number, $r = \sqrt{x^2 + y^2 + z^2}$, and $p = \sqrt{p_x^2 + p_y^2 + p_z^2}$. When only the dipole approximation is considered, the Lorentz force terms from B are zero, resulting in $p_z/\sqrt{p^2 + m^2c^2}$ in Eq. (1) and E in Eq. (3) being dropped. For most calculations, the soft-core potential term is set to zero since, as we will show, it does not affect the results presented here.

Also shown in Figs. 4.1(c) and 4.1(d) is an example rescattering flux “snapshot” from a collection of electron trajectories for He^+ and Ar^{8+} with the ionization and return scattering time indicated in Figs. 4.1(a) and 4.1(b). Strong-field rescattering [Fig. 4.1(c)] shows the traditional spreading and return of the electron after ionization while for ultrastrong field rescattering [Fig. 4.1(d)] the Lorentz deflection acts to displace the electron by ~ 50 a.u. along z . The > 20 a.u. spatial extent of the returning electron justifies a plane-wave approximation.

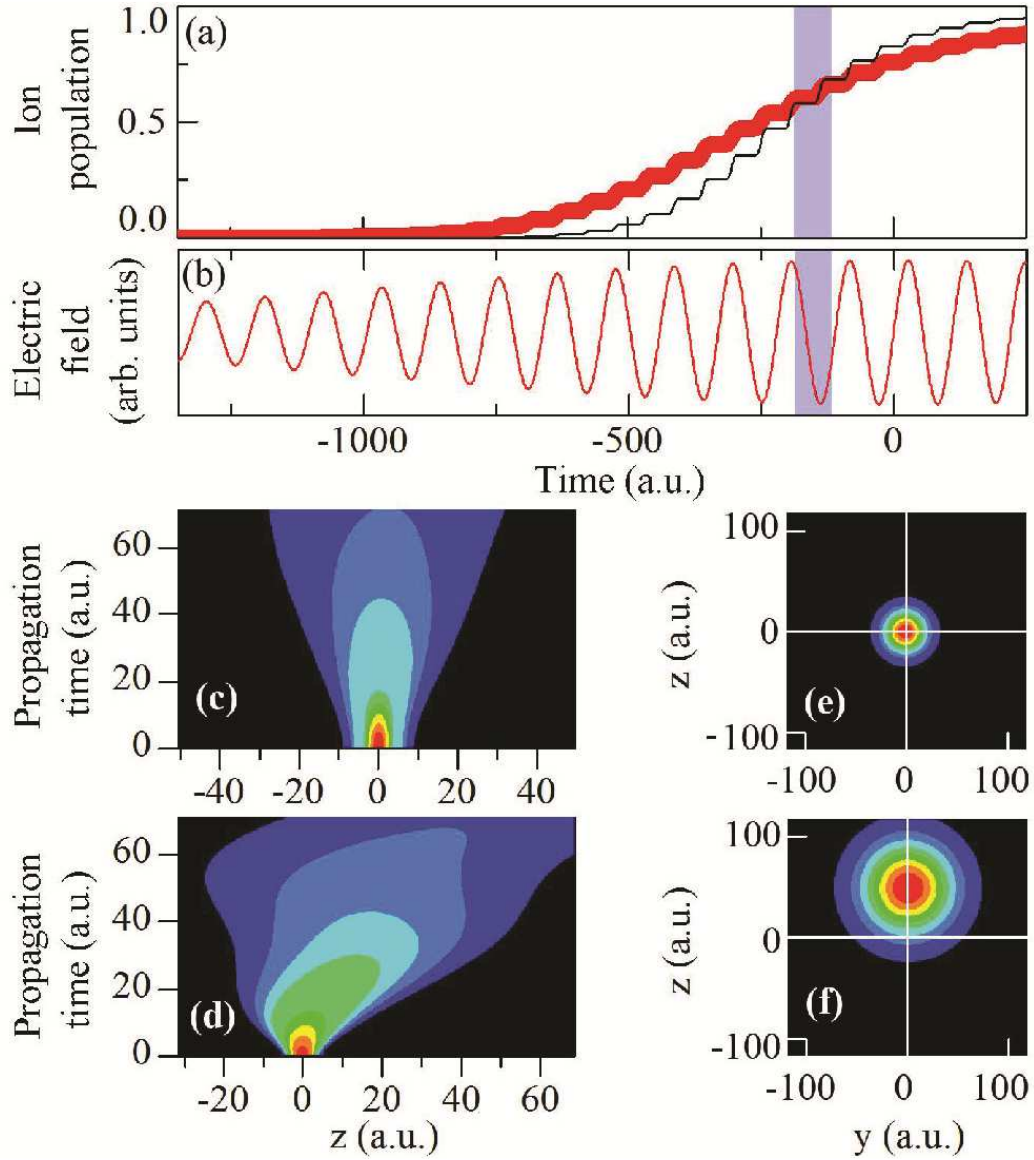


Figure 4.1 He^+ (bold) and Ar^{8+} (thin) population (a) as a function of time in the laser field (b) whose peak intensity is $2.4 \times 10^{15} \text{ W/cm}^2$ for He^+ and $5.2 \times 10^{16} \text{ W/cm}^2$ for Ar^{8+} . The shaded region from -188 a.u. to -116 a.u. Panels (a) and (b) indicate the time from ionization to scattering return for (c) and (d). The continuum electron density along z during this 72-a.u. window is shown for He^+ (c) and Ar^{8+} (d). The y - z flux profile at return is shown for He^+ (e) and Ar^{8+} (f).

Compared to ionization of neutrals or molecules, the plane-wave approximation becomes more accurate for ionization in ultrastrong fields. The smaller extent of more tightly bound states results in a greater spreading of the ionized electron due to the uncertainty principle. This increase in spreading is evident when comparing the ionization of Ar^{8+} in Fig. 4.1(f) to that of He^+ in Fig. 4.1(c). With regard to the use of trajectory ensembles in the continuum, the de Broglie wavelength of the continuum electron is typically 0.5 to 0.01 atomic units of length.

4.2.3 Elastic Rescattering

Upon revisiting the parent ion, elastic scattering is calculated using a full partial-wave calculation [37]. Elastic scattering (Fig. 4.2) is calculated for hydrogenlike species using a bare nucleus, Coulomb potential, $V(r) = Ze/r$. Low-energy scattering with unphysically large impact parameters (given the finite extent of the electron) is avoided by eliminating scattering energies below $0.3U_p$. Neglecting these energies does not affect final-state results above $0.3U_p$. For noble gases, we use Hartree-Fock screening of the nucleus with a screening charge density given by $\rho_e(r) = \frac{Ze}{4\pi r} \sum_{i=1}^3 D_i d_i^2 \exp^{-d_i r}$, with the D_i and d_i coefficients calculated using the ELSEPA routine [37]. The charge distribution ρ_e is used to obtain the screening potential from which scattering is calculated. While the scattering charge for hydrogenlike species is independent of r , atomic species have an effective charge that depends on the distance from the nucleus due to screening. The effective charge for scattering with Xe^{8+} is 8 for relatively large $r = 4$ a.u., increasing to 20 for an interaction at $r = 1$ a.u., and to 47 (nearly the full value of the bare nucleus) at $r = 0.1$ a.u.. For $\theta = \pi/2$ scattering, the incident energies corresponding to these impact parameter distances are shown in the Fig. 4.2(b) top x axis.

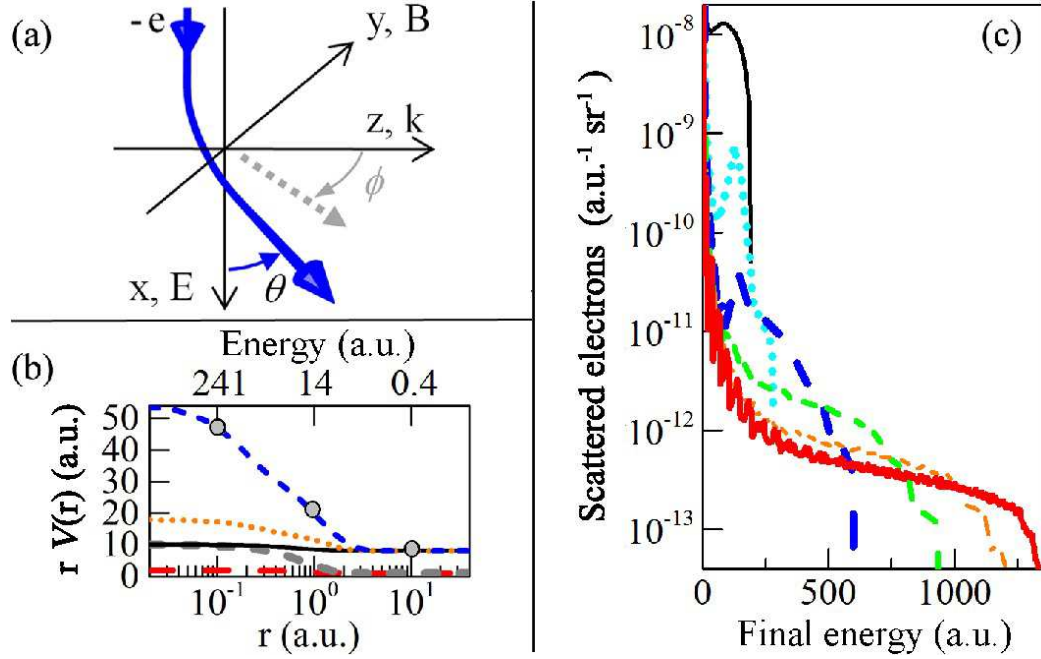


Figure 4.2 Coordinate system (a) for scattering with the parent ion (origin). The scattering potentials [plotted as effective charge in atomic units $rV(r)$ as a function of r] are shown in (b) for Xe⁸⁺ (dash, blue), Ar⁸⁺ (dotted, orange), Ne⁸⁺ (solid, black), Ne⁺ (thick dash, gray), and He⁺ (long dash, red). For Xe⁸⁺ the energy corresponding to the minimum r and effective charge for $\theta = \pi/2$ scattering is indicated by the top axis and on the Xe⁸⁺ potential curve (circle, filled gray). Energy and angle (θ) resolved scattering (c) from hydrogenlike ($Z = 3$) at 6×10^{16} W/cm² for $0.05 < \theta < 0.5$ (solid black), $0.5 < \theta < 1.0$ (dotted, light blue), $1.0 < \theta < 1.55$ (long dash, blue), $1.55 < \theta < 2.1$ (dash, green), $2.1 < \theta < 2.6$ (short dash, orange), and $2.6 < \theta < 3.14$ (thick solid, red).

Impact parameter distances of $r = 1$ a.u. have an incident energy of 14 a.u. for $\theta = \pi/2$ scattering. At the intensity where Xe⁸⁺ ionization is 90%, the maximum 3.2 U_p return energies of 116 hartree probe deep into the Xe⁸⁺ screened potential experiencing effective charges in the range $rV(r) \approx 30$. The screening potentials used for Ne⁺, He⁺, Ar⁸⁺, and Ne⁸⁺ are shown in Fig. 4.2(d). As is well known, potentials are

most accurately known for neutrals and single charge ions where experimental measurements have been done. The Ne^{8+} , Ar^{8+} , and Xe^{8+} ion potentials shown in Figs. 4.2(b) and 4.2(d) are sufficiently accurate for this work. Scattering is calculated for all ϕ and for θ between 0.05 to π radian forward to backscattering, respectively. An example of the angle and energy resolved scattering is shown in Fig. 4.2(c). The angle and energy integrated result from Fig. 4.2(c) is the total elastic scattering, which expressed as a ratio of the scattering to ionization is 2×10^{-4} for the example in Fig. 4.2(c).

4.3 Results

4.3.1 Total Elastic Scattering

To determine the intensity dependence for angle and energy integrated scattering, we calculated the total elastic scattering for multiple species from 10^{15} to 10^{19} W/cm^2 . Let us first direct attention to hydrogenlike species. These are the simplest to interpret as a function of intensity I , since a single parameter (Z) is changed as ionization proceeds from $Z = 2$ at $1.4 \times 10^{16} \text{ W/cm}^2$ to $Z = 7$ at $1.6 \times 10^{19} \text{ W/cm}^2$. Three different calculations are shown for the ionization, first is the nonrelativistic case where the laser field is treated in the dipole approximation ($B = 0$). The calculated electron scattering as a fraction of the total ionization at the end of the laser pulse decreases from 10^{-2} in a 10^{15} W/cm^2 strong field to 10^{-11} in the ultrastrong field at 10^{19} W/cm^2 . This tremendous reduction in the rescattering efficiency is consistent with the energy scaling in Rutherford scattering. A I^{-2} fit shown in Fig. 3 is in excellent agreement with the nonrelativistic $B = 0$ case, due to a nonrelativistic recollision energy U_p that is linear in intensity. Second, we calculated

the rescattering including relativistic effects in the continuum dynamics while maintaining the dipole approximation, i.e., $B = 0$. The results are similar to the nonrelativistic calculations but there is an increase by a factor of 3 at $2 \times 10^{19} \text{ W/cm}^2$ due to the relativistic mass shift limiting the excursion. The onset of the relativistic enhancement effects coincide with a classical field nonlinearity parameter $a_0 > 1$. Finally we included the full interaction with relativity and B . The results follow closely the nonrelativistic and relativistic cases until $6 \times 10^{16} \text{ W/cm}^2$ where scattering begins to drop. By $2 \times 10^{17} \text{ W/cm}^2$ the scattering is an order of magnitude smaller than the $1/I^2$ scaling. When the intensity reaches the value of $a_0 = 1$ at $2 \times 10^{18} \text{ W/cm}^2$ the yield is reduced by seven orders of magnitude. This reduction in the scattering is due to the Lorentz deflection (Fig. 4.1) from B [20,38] and consistent with the relativistic rescattering parameter, $\Gamma_r > 1$. The regime where Γ_r is much less than 1 may be considered in the nonrelativistic, $B = 0$ limit.

Included in Fig. 4.3 is scattering for noble gas ions with relativity and B . For clarity, these data points are labeled in the graph. As is indicated in Fig. 4.2(b), a screened potential gives greater scattering. With scattering scaling as Z^2 , the yield from an atom such as xenon can be significantly greater than a bare nucleus of the same ion charge. We begin with traditional strong field ionization of the first charge state for Ne^+ ($1 \times 10^{15} \text{ W/cm}^2$, $U_p = 2.2 \text{ a.u.}$) and He^+ ($2 \times 10^{15} \text{ W/cm}^2$, $U_p = 4.4 \text{ a.u.}$). The calculated total scattering is within a factor of 2 times the hydrogenlike result for the Ne^+ scattering, due to the low scattering energy. The calculated scattering for He^+ is near the hydrogenlike results as well, due in addition to the small nuclear charge of 2. Next we examine the scattering from Xe^{8+} ($2 \times 10^{16} \text{ W/cm}^2$) and Ar^{8+} ($5 \times 10^{16} \text{ W/cm}^2$).

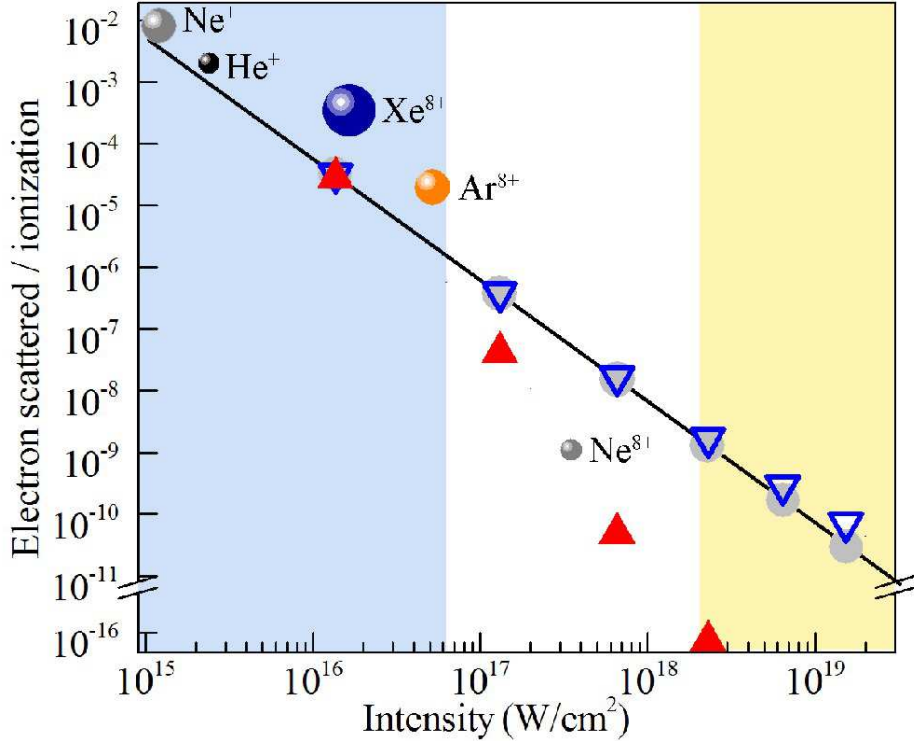


Figure 4.3 Ratio of total scattered photoionization (all angles, energies greater than $0.3U_p$) to photoionization as a function of intensity for hydrogenlike species with a nonrelativistic dipole response with $2 \leq Z \leq 7$ (circle, gray), relativistic dipole response with $2 \leq Z \leq 7$ (inverted triangle, blue), and relativistic full E, B field response with $2 \leq Z \leq 5$ (filled triangle, red). A $1/I^2$ line (solid, black) is added. Noble gas scattering (sphere) is shown for Ne^+ (gray), He^+ (small, black), Xe^{8+} (large, blue), Ar^{8+} (orange), and Ne^{8+} (small, gray). Two regions are highlighted for $\Gamma_r < 1$ (light blue) and $a_0 > 1$ (light orange).

The scattering yield for Xe^{8+} and Ar^{8+} is an order of magnitude larger than the simple hydrogenlike, Coulomb ion result due to the large screened nuclear charge. Both are at $\Gamma_r < 1$ intensities. Last is an excellent test case for scattering in ultrastrong fields. Ne^{8+} at $3 \times 10^{17} \text{ W/cm}^2$ has photoelectron energies on the order of $U_p = 660$ a.u. With $\Gamma_r = 15.6$, the rescattering is expected to be strongly affected by B . Looking to Fig. 4.3 we can see the amount of scattering for Ne^{8+} is 60 times smaller than the

nonrelativistic $B = 0$ hydrogenlike case and greater than the expected relativistic hydrogenlike case with B . A result consistent with a reduction from the Lorentz deflection and slight enhancement from a screened nuclear charge of 10. Experiments are underway to verify the drastic reduction in rescattering by nine orders of magnitude over an intensity change of only a factor of 6.

4.3.2 Photoelectron energy spectra

Elastically scattered electrons are critical to understanding photoelectron final states. The maximum energy without scattering is $2U_p$ while with scattering energies [39] can reach $10U_p$. We begin the analysis of the photoelectron final energies for the three cases used previously: a traditional nonrelativistic strong-field analysis with $B = 0$, including relativistic effects while setting $B = 0$, and the full field with relativistic dynamics. We begin in Fig. 4 with hydrogenlike ions and the portion of the spectrum resulting from elastic scattering with the parent ion at $2 \times 10^{16} \text{ W/cm}^2$ ($\Gamma_r = 0.14$, $a_0 = 0.1$), $6 \times 10^{16} \text{ W/cm}^2$ ($\Gamma_r = 0.94$, $a_0 = 0.17$), $2 \times 10^{17} \text{ W/cm}^2$ ($\Gamma_r = 6.54$, $a_0 = 0.31$), and $2 \times 10^{18} \text{ W/cm}^2$ ($\Gamma_r = 311$, $a_0 = 0.97$).

As we progress from Fig. 4.4(a) to Fig. 4.4(d) we see the evolution of the spectra and the impact of relativity and the Lorentz deflection. The agreement between all cases [Fig. 4.4(a)] is consistent with a nonrelativistic, dipole interaction. With increasing intensity, the overall decrease in the contribution of elastic scattering to the photoelectron final-state energy spectrum is quantified in Fig. 4.4(b) for a $\Gamma_r = 0.94$ where scattering is beginning to be suppressed as B deflects the returning electron. By the intensity of $2 \times 10^{17} \text{ W/cm}^2$, $\Gamma_r = 6.54$ in Fig. 4.4(c) nearly all elastic scattering has been suppressed with the highest energy photoelectrons most strongly affected.

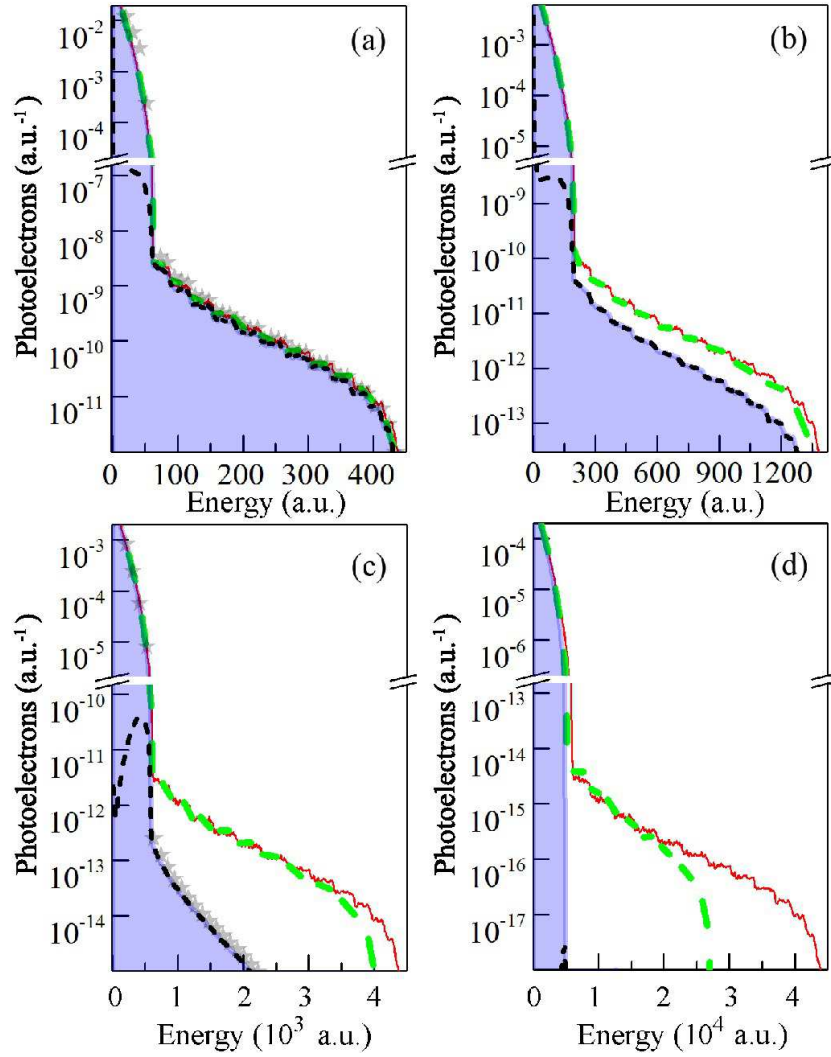


Figure 4.4 Photoelectron energy spectrum for hydrogenlike species with a nonrelativistic dipole response (thin, red), relativistic dipole response (dash, green), and relativity with B (filled, blue) at 2×10^{16} W/cm² (a), 6.3×10^{16} W/cm² (b), 2×10^{17} W/cm² (c), and 2×10^{18} W/cm² (d). The scattering component is shown (dotted, dark blue). To aid in (a)–(d) comparison the energy scale is $0 - 10.5U_p$. The results when including Coulomb focusing (gray star symbol) for the full field, relativistic calculation are shown in (a) and (c).

Finally, at $a_0 = 1$ in Fig. 4.4(d) one may infer that elastic scattering in the ultrastrong field does not occur, or at least is not observable at the level of 10^{-18} electrons per hartree and steradian. For the sake of completeness we also show the spectra without the Lorentz deflection but including the relativistic mass shift. The effect of relativistic continuum dynamics is to decrease the maximum kinetic energies attained from the field and elastic scattering.

Where the excursion of the electron is comparable to the ion potential, the force from the ion can affect the photoelectron in the continuum in a process known as Coulomb focusing [40]. One might at first suppose the large parent ion charges in ultrastrong fields could lead to strong Coulomb focusing effects. However, due to the large excursion that places the photoelectron far away from the parent ion and the high momentum gained from acceleration in the external field, Coulomb focusing plays a smaller role in ultrastrong fields. The results of Figs. 4.4(a) and 4.4(c) have included the Coulomb focusing with the soft-core potential described in Eqs. (1)–(3). The increase in the rescattering can be seen in Fig. 4 but is only a fraction of the displayed symbol size.

Our last calculation is for noble gases, relativistically with full E, B fields and the scattering potentials described in Fig. 4.2. This work is intended to help bridge the gap between theoretical work and experimental efforts to quantify new ultrastrong field physics. To begin we connect to earlier, nonrelativistic strong-field observations. Experimental data [39] is plotted in Fig. 4.5(a) along with our results for Ne^+ . Our calculations are consistent with the well-known strong-field response and previous results [39]. In Fig. 4.5(b) the photoelectron energy spectrum for Ar^{8+} is shown. With a $\Gamma_r = 0.87$ Ar^{8+} is beginning to be effected by B .

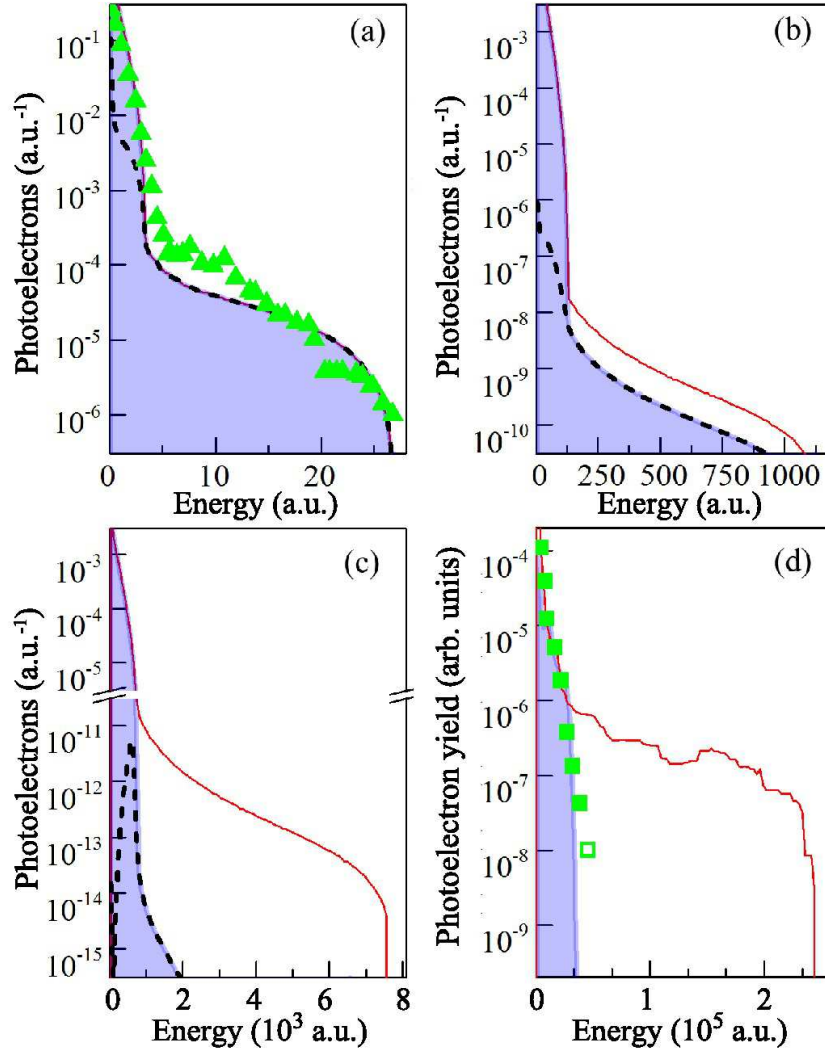


Figure 4.5 Energy resolved photoelectron spectrum for Ne^+ (a), Ar^{8+} (b), Ne^{8+} (c), Xe^+ and Xe^{26+} (d), and a nonrelativistic dipole (thin, red), relativistic, E , B response (filled, blue) with the partial yield from rescattering (dotted, dark blue). Experimental data is shown (triangle) [39] and (square) [41]. For (d) the nonrelativistic response (thin, red) has been multiplied by 120 from the calculated value (dotted, red) to compare with the data. The open square data point in (d) is the limit of the signal to noise for that experimental energy.

The scattering reduction seen in Fig. 4.5(b) is consistent with the Lorentz rescattering parameter factor of $\exp(-\Gamma_r)$. In Fig. 4.5(c) the Ne^{8+} calculation reveals that at $3 \times 10^{17} \text{ W/cm}^2$ for $\Gamma_r = 15.6$, scattering may be neglected since its occurrence is at the level of 10^{-14} electron per hartree and steradian). The highest intensity presented is $1.2 \times 10^{19} \text{ W/cm}^2$ for the ionization of Xe at $\Gamma_r = 7480$, $a_0 = 2.4$. To compare with experiments, Fig. 4.5(d) has been modeled using the experimental focus, spatial volume, energy resolution, angular acceptance [17,41], and multiple charge state distribution expected as ionization proceeds from neutral Xe to Xe^{26+} by the end of the pulse. Comparing the data with our calculation, one can see that the high-energy rescattering expected nonrelativistically is absent.

4.4 Conclusion

A three-step model is extended into the relativistic, ultrastrong field regime ($\Gamma_r > 1$ and $a_0 < 10$). Continuum dynamics are treated semiclassically with Monte Carlo trajectory ensembles to account for relativistic and B effects while ionization and rescattering is treated quantum mechanically. Studies of scattering in hydrogenlike systems show elastic rescattering generally obeys a $1/I^2$ scaling when the Lorentz deflection is small, i.e., $\Gamma_r < 1$. Elastic scattering decreases roughly as a function of $\exp(-\Gamma_r)$ until becoming undetectable. Relativistic mass effects are noted but play a smaller role, contributing for intensities beyond 10^{18} W/cm^2 . In addition to work with fundamental hydrogenlike species, we calculated how the elastic scattering would be observed for noble gas species with screened atomic potentials. The results compare favorably with experimental data.

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Chapter 5

ELASTIC RESCATTERING PHOTOELECTRON DISTRIBUTIONS ENTERING THE RELATIVISTIC REGIME

5.1 Introduction

High-strength laser fields can exceed the binding nuclear Coulomb field for atoms and molecules and ionize the outer, least tightly bound electron. Fields of this strength (0.17 a.u., intensities of 10^{15} W/cm²) dominate photoelectron dynamics and the oscillating laser field can force the photoelectron to return and ‘rescatter’ with the parent ion[1]. Strong field ionization and rescattering has been used to measure electron dynamics[2], collisionally excite multiple electrons[3], generate coherent attosecond x-ray light[4], and perform molecular tomography[5]. Photoelectron angular distributions are a key to understanding the physics of elastic rescattering in strong laser fields. Collaborative theoretical and experimental efforts[6] disentangled the many possible excitation pathways. These studies clarified the role of the short-range (recollision near the parent ion) and long-range (recollision away from the parent ion core) elastic rescattering.

The three-step model of ionization [7,8] has provided a context by which many strong field processes, including elastic scattering, can be clearly understood. The three-step analysis is traditionally limited to nonrelativistic, dipole interactions where the energy scale of the interaction [e.g., the ponderomotive energy $U_p = e^2|E|^2/(4m\omega^2)$ for an electron charge $-e$ oscillating in an electric field E at a frequency ω] is far less than the electron rest mass m . As the intensity increases to “ultrastrong”

fields [9] nonrelativistic and dipole approximations are no longer accurate. The external magnetic field B can deflect the photoelectron rescattering and cause it to miss the parent ion [10]. The ratio of the Lorentz deflection distance to the spatial width of the returning electron wave is indicated by a Lorentz deflection parameter [11,12], $\Gamma_r = U_p^{3/2} V_{IP} / (3c^2 \omega)$ for ionization from a binding energy V_{IP} . When $\Gamma_r = 1$, the deflection of the returning electron is equal to its spatial extent. It follows that for $\Gamma_r \gg 1$ rescattering will be reduced to the point of shutdown. At even higher ‘extreme’ fields[13], relativistic effects, radiation processes, and the external B field affect both bound and continuum electrons.

The purpose of this work is to understand the elastic scattering process as it changes from strong to ultrastrong fields. Elastic scattering is a primary mechanism by which the field converts energy into particle motion, a process that is critical to realizing many long-term goals in science including laser fusion. As was the case for pioneering studies in the strong field[14,15], the complexity of the ultrastrong field frontier requires theoretical models than can accurately capture experimental observations. In ultrastrong fields the electron can quickly become relativistic and traverse a large portion, or even exit, the laser focus during a femtosecond laser pulse. Spatial and temporal integration of the interaction region can be an integral part of understanding the forces experienced by the photoelectron on the way to the detector and the science that underlies the measurements [16]. We report on the photoelectron angular distributions from Ne, Ar, and Xe across strong and ultrastrong fields. The results show how the Lorentz deflection affects the rescattering and can reduce it to a negligible level as one advances into the ultrastrong field regime. Comparisons of these angular distributions to future experiments will be needed to quantify whether

several assumptions in the models are correct, such as a lack of multielectron excitation. The flexibility of the model should allow for valid elastic scattering calculations with most atoms and ions at field interactions where $\Gamma_r \sim 1$ and the interaction is not yet into the extreme relativistic regime.

5.2 Model

The model used represents an emerging technique that accurately captures ultrastrong field physics. Interactions (such as ionization[17] or radiation reaction[18]) are treated quantum mechanically and propagation in the field is handled classically when the electron deBroglie wavelength is much smaller than the drive wavelength. Properly applied the technique has advantages in its extension physically of the three step model[7] to ultrastrong fields and insight from its intuitive treatment of ionization, propagation, and recollision. The full reasons behind the validity of this three-step parsing are complex but lie within the different approximations valid for each step. For the first step of tunneling ionization, the energy scales are of order 10 to 30 a.u. While the external field does affect the ionizing bound state near the nucleus, it does not generally change the bound state wave function or ionization rate by more than a factor of 2[19,20]. In this study, we use the low-frequency, nonrelativistic tunneling ionization rate[17] also referred to as the Ammosov, Delone, Krainov rate[21]. The electric field in the studies is a $\sigma = 34$ fs pulsed, $\lambda = 800$ nm carrier wavelength, plane wave,

$$\mathbf{E} = E_0 \sin(2\pi/\lambda z - \omega t) \exp\left(-\left(t - \frac{z}{c}\right)^2 / \sigma^2\right) \hat{\mathbf{x}} \quad (5.1)$$

when considering the full field, $\mathbf{B} = |\mathbf{E}|/c \hat{\mathbf{y}}$, where $\hat{\mathbf{x}}, \hat{\mathbf{y}}, \hat{\mathbf{z}}$ are the unit vectors in cartesian coordinates. In the dipole approximation we set $\mathbf{B} = 0$. For all the results

presented in this work we adjust E_0 such that the atom or ion has reached 90% ionization by the end of the pulse.

In the second step, the external field accelerates the electron to energies that can exceed 10^3 Hartree. Quantum aspects in the continuum are arguably captured using a Monte Carlo trajectory ensemble with uncertainties in momentum and position determined by tunneling ionization. The semiclassical trajectory ensemble method used has been described previously [22]. Briefly, for each time step we propagate on the order of 10^4 trajectories with a weight determined by the ionization to represent the quantum photoelectron in the continuum. The trajectories are generated by integrating Hamilton's equations of motion with the external field $\mathbf{E}$ and soft core ion potential ($\delta \sim 0.2$)

$$\frac{dp_x}{dt} = \frac{-Ze^2x}{(r^2 + \delta)^{3/2}} - e|\mathbf{E}| \left[1 - \frac{p_z}{\sqrt{p^2 + m^2c^2}} \right] \quad (5.2)$$

$$\frac{dp_y}{dt} = \frac{-Ze^2y}{(r^2 + \delta)^{3/2}} \quad (5.3)$$

$$\frac{dp_z}{dt} = \frac{-Ze^2z}{(r^2 + \delta)^{3/2}} - e|\mathbf{E}| \frac{p_x}{\sqrt{p^2 + m^2c^2}} \quad (5.4)$$

$$\frac{dx}{dt} = \frac{p_x c}{\sqrt{p^2 + m^2c^2}} \quad (5.5)$$

$$\frac{dy}{dt} = \frac{p_y c}{\sqrt{p^2 + m^2c^2}} \quad (5.6)$$

$$\frac{dz}{dt} = \frac{p_z c}{\sqrt{p^2 + m^2c^2}} \quad (5.7)$$

where p_x, p_y, p_z are the momenta along the cartesian coordinates; Z is the ion charge and t is time. An example snapshot of a trajectory ensemble from ionization just after the peak of the optical cycle at an intensity of $1.3 \times 10^{17} \text{ W/cm}^2$ is shown in Fig. 5.1. In Fig. 5.1 we follow an example of 1000 trajectories for ionization at $z = 0, t = -188 \text{ a.u.}$ in a pulse with a peak intensity of $1.3 \times 10^{17} \text{ W/cm}^2$ (i.e., $\mathbf{E} = -1.7\hat{x}$ in atomic units) to the return at $z = 0, t = -118 \text{ a.u.}$, ($\mathbf{E} = 0.6\hat{x}$). The rapid spreading of the initial ionization is clear in Fig. 5.1 as well as the Lorentz deflection of the photoelectron by approximately 50 a.u. in the direction of z when it returns 70 a.u. of time later. For this case ($\Gamma_r = 3.2$), the resulting rescattering flux that revisits the core is 4% of the peak value [i.e., $\exp(-3.2)$]. Fortunately, to a high level of accuracy, the returning electron can be treated as a plane wave since the rescattering electron wave at $\sim 200 \text{ a.u.}$ wide is much larger than the $\sim 1 \text{ a.u.}$ length scale of the scattering potential.

Upon the return of the photoelectron to the core, we calculate elastic scattering with the parent ion. Elastic scattering in ultrastrong fields is affected by new aspects when compared with scattering in strong fields. The Lorentz deflection in the continuum, as ionization is driven by the external field, is addressed by the trajectory ensemble method described previously. The relativistic continuum and rescattering process with the ion potential must be treated accurately since ultrastrong fields present large recollision energies; scattering probes effective charges very near the full nuclear charge. The potentials used were calculated in the Hartree–Fock approximation using ELSEPA[23]. For clarity, we plot in Fig. 5.1(c) the potentials’ effective charge, i.e., $rV(r)$, as a function of distance from the nucleus.

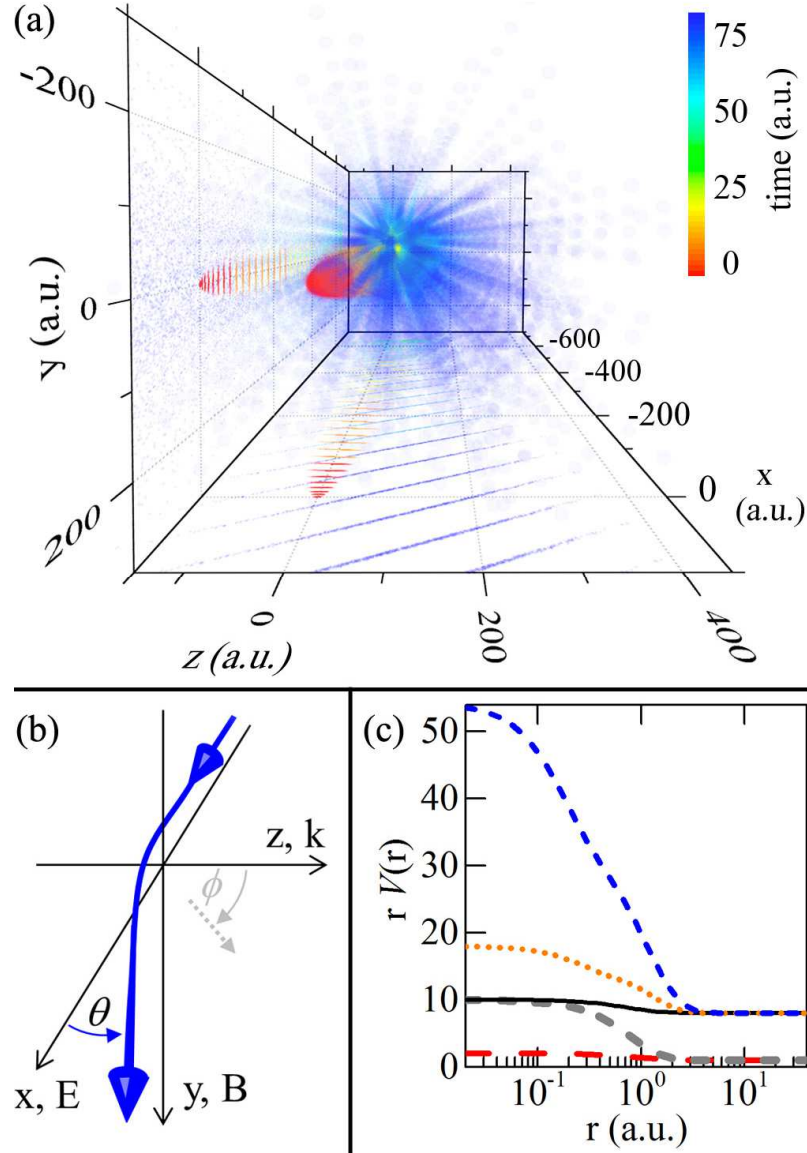


Figure 5.1 Monte Carlo ensemble for 10^3 trajectories: (a) from ionization at an intensity of 1.3×10^{17} W/cm² and its return to the core 70 a.u. later. Symbol is plotted for every 1.1 a.u. time step from ionization at $t = 0$. Color mapping used for propagation time after ionization. The x - z and x - y plane projections show the rapid spreading of the electron from ionization at the origin. In addition, the increasing distance between the 1.1 a.u. time steps in the plane projections indicate the electron acceleration in the field and the tilt of the electron wave front from the Lorentz force (x - z projection); (b) coordinate system for the elastic scattering from the nucleus shown in (a); (c) scattering potentials for Ne^+ , Ne^{8+} , Ar^{8+} , and Xe^{8+} . Atomic units (a.u.) are used.

Viewed in this way, one can more easily see the screening provided by the ion core. After elastic scattering the photoelectron continues to interact with the field until the laser pulse has passed, typically two pulse durations (2σ) after the peak.

5.3 Results

We display the calculated final photoelectron energy spectrum in Fig. 5.2. The angle integrated photoelectron yields for Ne^+ , Xe^{8+} , Ar^{8+} , and Ne^{8+} are plotted for nonrelativistic, dipole calculations, and full fields with relativity. For ease of comparison, energy units of U_p were chosen in Fig. 5.2. We present the scattering normalized to the amount of ionization, i.e., the integration of the photoelectron energy spectrum over energy gives a value of 1. We begin with Ne^+ , which can be compared to experiments in the strong field [24]. The yield shows the characteristic low energy ionization from 0 to $2U_p$ and the high energy plateau from rescattering with the parent ion that stretches from $2U_p$ to $10U_p$. There is, as expected, no difference between the nonrelativistic dipole calculations and those including relativity and the $\mathbf{B}$ field for Ne^+ . Figure 5.2 also displays ionization for Xe^{8+} at $2 \times 10^{16} \text{ W/cm}^2$, Ar^{8+} at $5 \times 10^{16} \text{ W/cm}^2$, and Ne^{8+} at $3 \times 10^{17} \text{ W/cm}^2$. These species extend into the ultrastrong field and two primary factors are responsible for the decrease in rescattering as the intensity increases. The first, as in Rutherford scattering, is the inverse square energy dependence of the scattering process. Since the recollision energy (i.e., U_p) scales with the intensity, one can expect a quadratic drop in the scattering yield with increasing intensity. The second is the Lorentz deflection shown in Fig. 5.1. The decrease in electron yield from $2U_p$ to $10U_p$ for Xe^{8+} is a result of the energy dependence of elastic scattering.

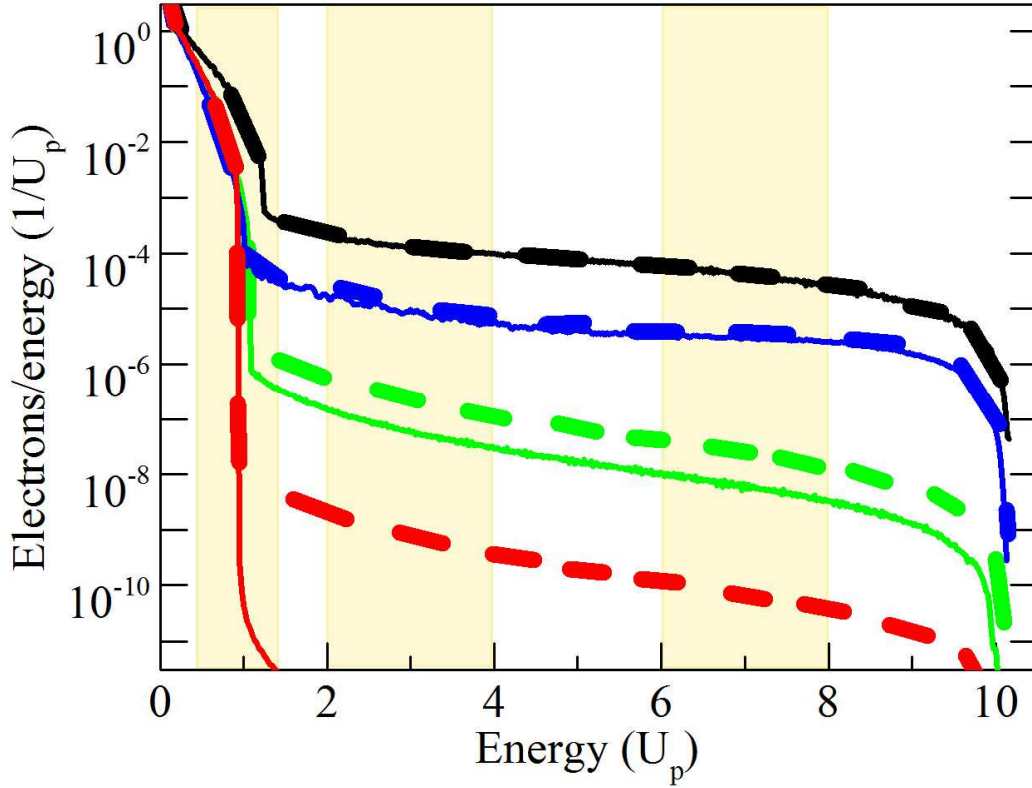


Figure 5.2 Angle-integrated photoelectron energy distributions for Ne^+ (black, $U_p = 2.6$ a.u.), Xe^{8+} (blue, $U_p = 38$ a.u.), Ar^{8+} (green $U_p = 115$ a.u.), and Ne^{8+} (red, $U_p = 770$ a.u.) as a function of the final photoelectron energy. Yield is given in electrons per unit U_p energy. For each species, we show the nonrelativistic dipole (thick, dash) and the relativistic full field (solid) yields. Energy integration regions for the angular distributions shown in Fig. 3 are highlighted.

Despite the large nuclear charge for xenon, the higher intensity decreases the elastic scattering yield by 1 order of magnitude compared to Ne^+ . The ionization of Ar^{8+} occurs at a $\Gamma_r = 1$ so the expected reduction from the Lorentz factor is 0.37, i.e., $\exp(-\Gamma_r)$. Nevertheless, comparing the nonrelativistic, dipole calculation for Ar^{8+} to the relativistic full field result shows in this case, the nearly 3 order of magnitude drop in rescattering compared to Ne^+ is due primarily to the higher energies in ultrastrong

fields. Progressing to Ne^{8+} at $3 \times 10^{17} \text{ W/cm}^2$, the mechanism behind the reduction in the rescattering changes abruptly to Lorentz deflection. For Ne^{8+} the Lorentz deflection parameter is $\Gamma_r = 15.6$. The new role of the Lorentz reduction in rescattering can be clearly seen in Fig. 5.2. While the intensity has increased only by a factor of 6 from Ar^{8+} to Ne^{8+} ($5 \times 10^{16} \text{ W/cm}^2$ to $3 \times 10^{17} \text{ W/cm}^2$) the rescattering in Ne^{8+} is absent at a level of 10^{-11} of the photoionization yield.

In Fig. 5.3 we show the calculated angular distributions at the photoelectron energies ($\pm$ energy integration range) of $U_p(\pm 0.5U_p)$, $3U_p(\pm U_p)$, and $7U_p(\pm U_p)$ in the nonrelativistic, dipole case [Figs. 5.3(a)–5.3(c)] and in the fully relativistic case with the laser magnetic field [Figs. 5.3(d)–5.3(f)]. The authors note the relativistic, $\mathbf{B}$ field yields for Ne^{8+} are vanishingly small and do not appear in Fig. 5.2. For reference, these are for Ne^{8+} at $3U_p$, 6.1×10^{-14} electrons/energy ($1/U_p$), and at $7U_p$, 4.8×10^{-19} electrons/energy ($1/U_p$). The angular distributions are presented as an angle θ from the electric field axis (Fig. 5.1). We plot the different species together to better understand the contributing mechanisms and expected changes in the angular distributions going into the ultrastrong field. All yields are normalized to aid in comparison. One may retrieve the actual yield at any energy for these species by combining the angular distribution with the angle integrated results of Fig. 5.2.

Ionization at energies of U_p in Figs 5.3(a) and 5.3(c) have an angular emission, $\theta = \arctan\left(\left(p_y^2 + p_z^2\right)^{1/2}/p_x\right)$, that is dominated by the initial momentum from ionization and drift energy from the field without scattering. The angular width is narrow ($\theta < 15^\circ$) for ionization by a plane wave. Due to the acceleration and momentum along the laser field direction, p_x increases quickly when going to ultrastrong fields. Since the transverse momentum (p_y, p_z) determined by the atomic

bound state does not change by more than a factor of 3 across these species, the distributions become more aligned with the field as one proceeds from Ne^+ to Xe^{8+} to Ar^{8+} to Ne^{8+} .

As one increases to $3U_p$, rescattering is the mechanism behind the yield and the emission angle for all the species is at its broadest extending from 0° to 90° from the laser electric field. The combination of the returning energy and scattering potential gives the observed structure and ‘scattering rings’ at large angles first observed in xenon [25]. What is important to observe from Figs. 5.3(b) and 5.3(e) is the $\mathbf{B}$ field and relativistic effects do not change the angular distribution until one is well into the $\Gamma_r \gg 1$ regime where Lorentz deflection has greatly reduced the yield. For Ar^{8+} , where $\Gamma_r \cong 1$, there is no significant difference in the angular distributions of Figs. 5.3(b) and 5.3(d). Proceeding to the highest energy, $7U_p$ distributions in Fig. 5.3(c), the angular emission range narrows as the mechanism changes over to backscattering into narrow angles along the electric field. The smaller impact parameters for the scattering process give these higher energies a greater sensitivity to the Lorentz deflection. In Figs. 5.3(c) and 5.3(f), one can see backscattering ($\theta = 0$) for both Ar^{8+} and Ne^{8+} is lower in Fig. 5.3(f) with the external laser $\mathbf{B}$ field compared to Fig. 5.3(c).

As the intensity increases, the Lorentz deflection that increases Γ_r and causes the rescattering electron to miss the parent ion also forward-deflects the overall photoionization yield [22]. In a plane wave this forward deflection is described by [26]

$$\tan \theta = \sqrt{2mc^2/E_{\text{kinetic}}} \quad (5.8)$$

At intensities of $3 \times 10^{17} \text{ W/cm}^2$ and below, this is not a concern in our context for the angular distributions since the forward-deflection angle from this field momentum is less than 5° .

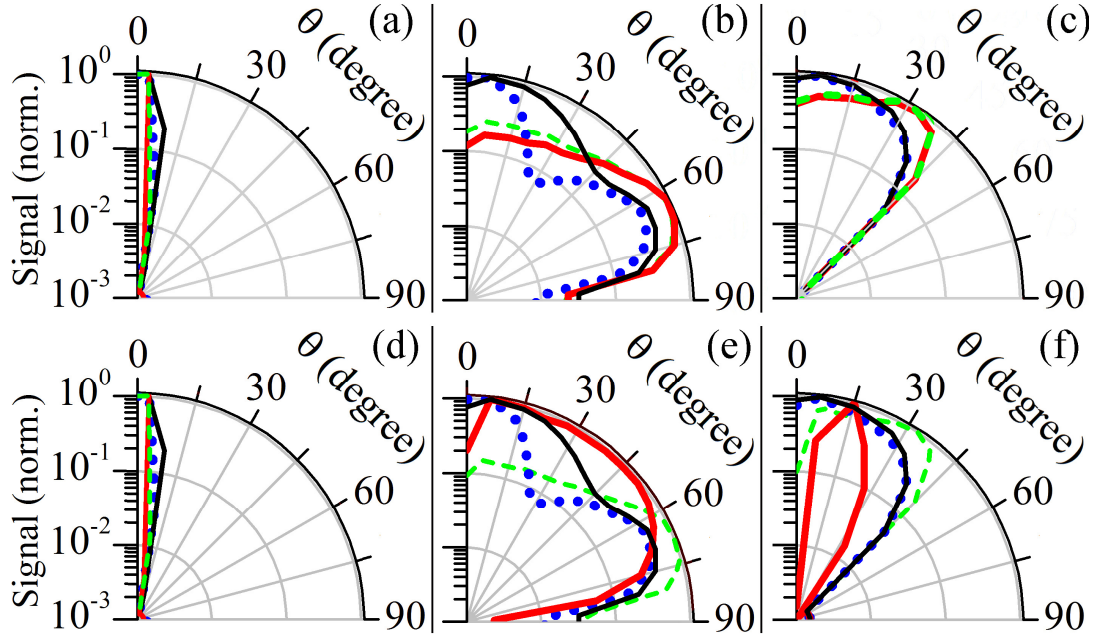


Figure 5.3 Photoelectron angular distributions calculated for $B = 0$, for Ne^+ (solid, black), Xe^{8+} (dotted, blue), Ar^{8+} (dash, green), and Ne^{8+} (thick solid, red): (a)–(c) nonrelativistically; (d)–(f) full field, relativistically; (a), (d) for energies $U_p \pm 0.5U_p$; (b), (e) for energies $3U_p \pm U_p$; and (c), (f) for energies $7U_p \pm U_p$. Yields are normalized to the peak value at that energy (Fig. 5.2).

5.4 Conclusion

In conclusion, we use a relativistic extension of a three-step recollision model with Hartree–Fock scattering potentials to calculate photoelectron energy spectra and angular distributions for ionization with elastic scattering from strong to ultrastrong fields up to $3 \times 10^{17} \text{ W/cm}^2$. The yield of the noble gas species shows a decrease in

the rescattering yield due to the inverse square dependence of the elastic scattering on energy. As the laser magnetic field affects the photoionization, there is a sharp reduction in elastic rescattering when $\Gamma_r > 1$. The onset of the reduction from 5×10^{16} to 3×10^{17} W/cm² decreases the rescattering yield by $\sim 10^5$ over a change of intensity by a factor of 6. The angular distributions of the photoelectrons are not drastically changed during the range $0 < \Gamma_r < 1$. For $\Gamma_r > 1$, elastic backscattering at large energies is most strongly effected, corresponding to short-range collisions between the parent ion and the returning photoelectron. The relativistic extension of a three-step recollision model with accurate atomic potentials is well-suited to comparison with experiments in the ultrastrong intensity regime that lies between traditional strong fields and extreme relativistic interactions.

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Chapter 6

THE ULTIMATE CUTOFF IN RESCATTERING FLUX

6.1 Introduction

In strong laser fields, an ionized photoelectron can be driven back to revisit its parent ion due to the periodic oscillation of external laser field, a process well known as rescattering [1]. Rescattering has been proposed as the mechanism behind nonsequential ionization (NSI) [2] and high harmonic generation (HHG) [3]. Rescattering has also been used to generate coherent attosecond X-ray light [4] and perform molecular tomography [5]. Based on the single atom response, the maximum kinetic energy a returning photoelectron can possess is shown to be $3.17U_p$ [6], where $U_p = e^2 E_0^2 / 4m\omega^2$ is the pondermotive energy [7] with E_0 and ω as the peak magnitude and angular frequency of laser electric field.

As the laser intensity proceeds to the ultrastrong regime ($> 3 \times 10^{16} \text{W/cm}^2$), the Lorentz force on the photoelectron due to the laser magnetic field starts to become significant to the photoelectron dynamics and deflects the electron away from the parent ion [8, 9], and results in lower rescattering. The rescattering is also dependent on the laser wavelength [10, 11], as both the pondermotive energy and the wavepacket spread in the continuum scale with wavelength.

In this chapter, we investigate the returning fluence and inelastic impact ionization in strong to ultrastrong fields. The studies here address issues related to wavelength dependence and the matching between returning fluence and cross section on rescattering energies. The ultimate cutoff in the rescattering flux is observed in the

relativistic regime. This work serves as guidance for experimentalists in searching for detectable inelastic impact ionization yield.

6.2 Methods

Linearly polarized light is used in the calculation with the form of $\vec{E} = E_0 \sin(kz - \omega t)\hat{x}$ (E_0 is the electric field, ω is the optical frequency, and k is the vector number) and the magnetic field $\vec{B} = |\vec{E}|/c\hat{y}$ is included. Tunneling ionization rate is calculated using the Ammosov-Delone-Krainov (ADK) model (see equation 2.2) for all species. The tunneling ionization rate and ion population for Ar^{6+} are shown in Fig 6.1 (a) along with the electric field shown in Fig 6.1 (b). In order to keep comparisons across species similar, the intensity for each species is chosen so that ion population reaches 10% at the end of the pulse.

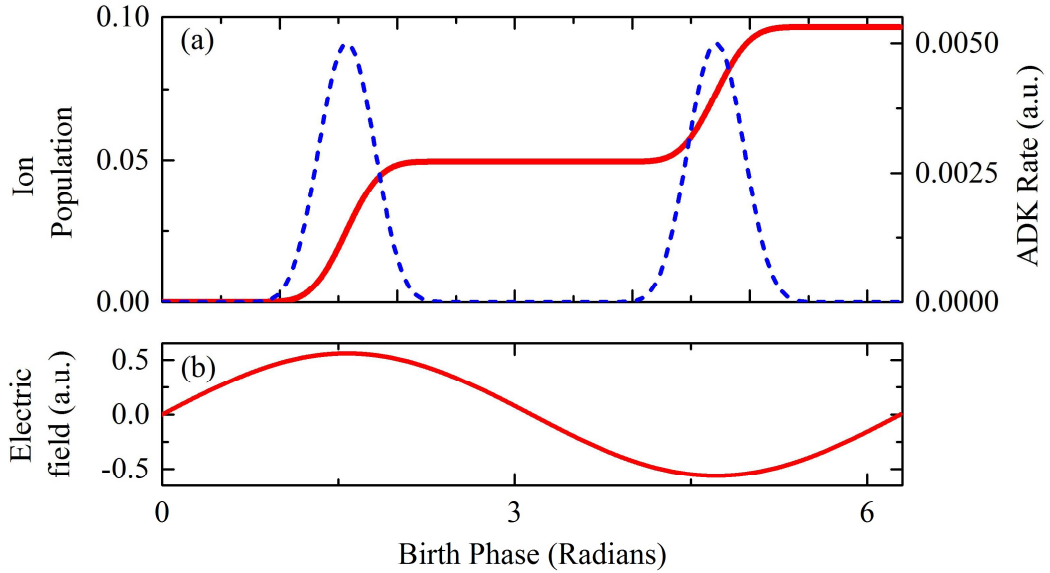


Figure 6.1 Ar^{6+} (solid) ion population (a) and tunneling ionization rate (dashed) (a) as a function of time in the laser field (b) for one optical cycle. The laser peak intensity is $1.1 \times 10^{16} \text{ W/cm}^2$ and wavelength is 800 nm.

After ionization, a Gaussian Monte Carlo ensemble with 1,000 electron trajectories is launched in the continuum. To mimic the quantum wavepacket, the ensemble is assigned with a momentum spread defined in equation 2.18. The spatial distribution can be subsequently calculated using the Heisenberg uncertainty principle. The momenta and position of each trajectory in the ensemble is generated by integrating the relativistic Hamilton's equations of motions, where the Lorentz force due to magnetic field is included and the electron interacts with a soft core potential as shown in equations (4.1) – (4.6). For most calculations, the interaction range of soft-core potential is much smaller than the electron excursion, and does not affect the electron dynamics in the continuum.

Experimental collections are results of pulse integration, and it is worthy to investigate the wavepacket dynamics at different birth phase within the pulse. It has been shown that rescattering can only happen in the second and the fourth quarter of one optical cycle ($\pi/2$ to π and $3\pi/2$ to 2π) (see figure 2.11). Within the quarter, electrons ionized at the beginning take the longest time to return to the parent ion, the so called 'long' trajectories, and electrons ionized toward the end are called 'short' trajectories. By calculating the position of each trajectory in the electron ensemble, we show the initial and returning spread of the electron wavepacket as a function of birth phase in figure 6.2. For each species, the initial spread increases as the external electric field decreases from the peak at $\pi/2$ to zero at π . This inverse dependence has been described in equation 2.18. Electrons ionized early (near the peak of the pulse) spend a longer time in the continuum before revisiting parent ion and the electron wavepacket diverges more as can be seen in Figure 6.2 (b). The returning spread does not decrease monotonically from $\pi/2$ to π and instead it bounces back toward the end,

this is due to the increase in initial spread. Comparing all four species, Ar^{9+} consistently has the largest returning spread as the deviation of trajectories due to Lorentz force in the ultrastrong field ($1.6 \times 10^{18} \text{ W/cm}^2$) is the largest.

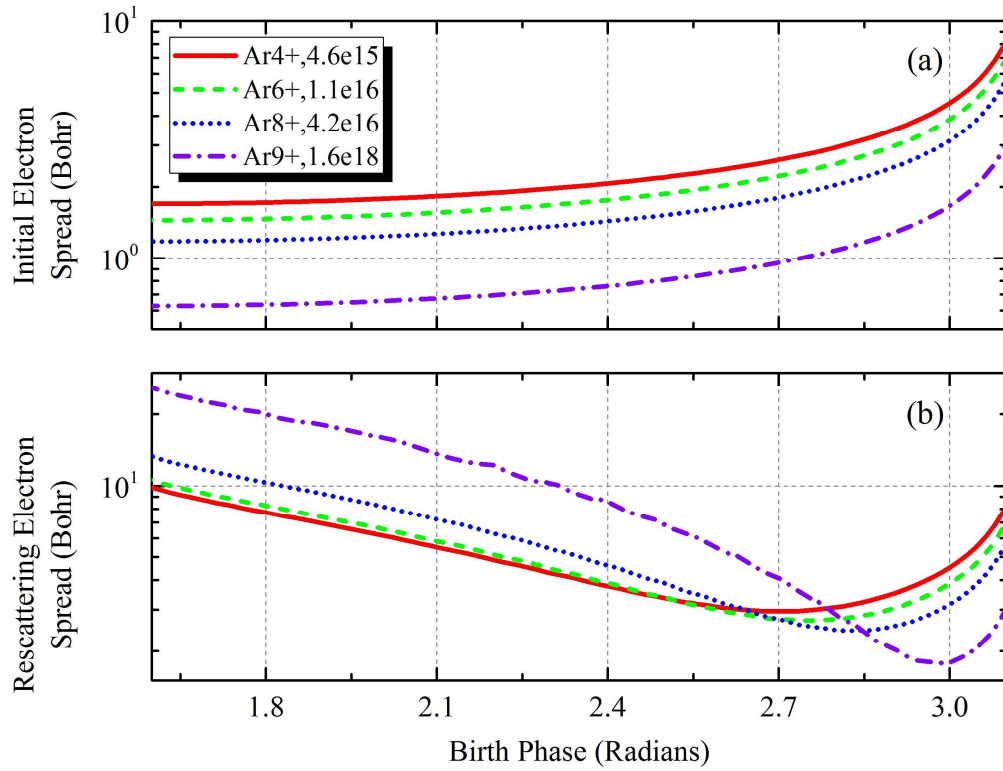


Figure 6.2 Initial spread (a) and return spread (b) of electron ensemble as a function of birth phase for Ar^{4+} at $4.6 \times 10^{15} \text{ W/cm}^2$ (solid), Ar^{6+} at $1.1 \times 10^{16} \text{ W/cm}^2$ (dashed), Ar^{8+} at $4.2 \times 10^{16} \text{ W/cm}^2$ (dotted), and Ar^{9+} at $1.6 \times 10^{18} \text{ W/cm}^2$ (dash dotted). The wavelength is 266 nm for all cases.

6.3 Results

The returning fluence is a function of wavepacket spread, deflection, and tunneling ionization rate, but mostly dominated by the tunneling ionization rate. The

difference in the tunneling ionization between $\pi/2$ and π has a typical value of 10^{20} . Since both elastic and inelastic scattering crosssection are functions of the energy of incident electrons, we decide to look at the rescattering flux dependence on returning energy. Three scenarios with different intensity and wavelength are shown in Figure 6.3. The rescattering flux contributed by ‘long’ trajectories and ‘short’ trajectories are both shown, where ‘long trajectories’ represented by solid symbols and ‘short trajectories’ shown with unfilled symbols. As intensity increases from $4.6 \times 10^{15} \text{ W/cm}^2$ (circle) to $1.6 \times 10^{18} \text{ W/cm}^2$ (inverse triangle), the Lorentz force due to magnetic field becomes significant and the Lorentz deflection of the electron ensemble largely reduces the returning fluence. On the other hand, as wavelength moves from 266 nm (square) to 800 nm (inverse triangle), the returning wavepacket spread increases and leads to lower returning fluence. The returning kinetic energy for the three cases scales with the well-known pondermotive energy $U_p = e^2 |\mathbf{E}_0|^2 / 4m_e \omega^2$. Besides the difference in magnitude of rescattering flux, the contributions from ‘long’ and ‘short’ trajectories also show differences. For the case of Ar^{4+} at $4.6 \times 10^{15} \text{ W/cm}^2$ and 800nm, the ‘long’ trajectories dominates the rescattering flux as the tunneling ionization rate peaks at the birth phase of ‘long’ trajectories. For the case of Ar^{9+} at $1.6 \times 10^{18} \text{ W/cm}^2$ and 800 nm, the contributions from ‘long’ trajectories are not observable at the level of 10^{-28} due to the huge spread width and deflection of returning wavepacket for ‘long’ trajectories. The case of Ar^{9+} at $1.6 \times 10^{18} \text{ W/cm}^2$ and 266 nm sits at the transition regime as the ‘short’ trajectories contribution starts to exceed ‘long’ trajectories contribution.

The structure of rescattering flux reflects the signature of relativistic effect and it is convenient to employ the rescattering deflection parameter $\Gamma_r = U_p^{3/2} V_{IP}^{1/2} /$

$(3c^2\omega)$ [13] to gauge the progression from strong- to ultrastrong-field rescattering flux. When $\Gamma_r < 1$ it is considered in the nonrelativistic regime, and when $\Gamma_r > 1$ it is considered in the relativistic regime. The deflection parameter of the three cases in Figure 6.3 changes from 0.015 to 3.126, and to 255.8.

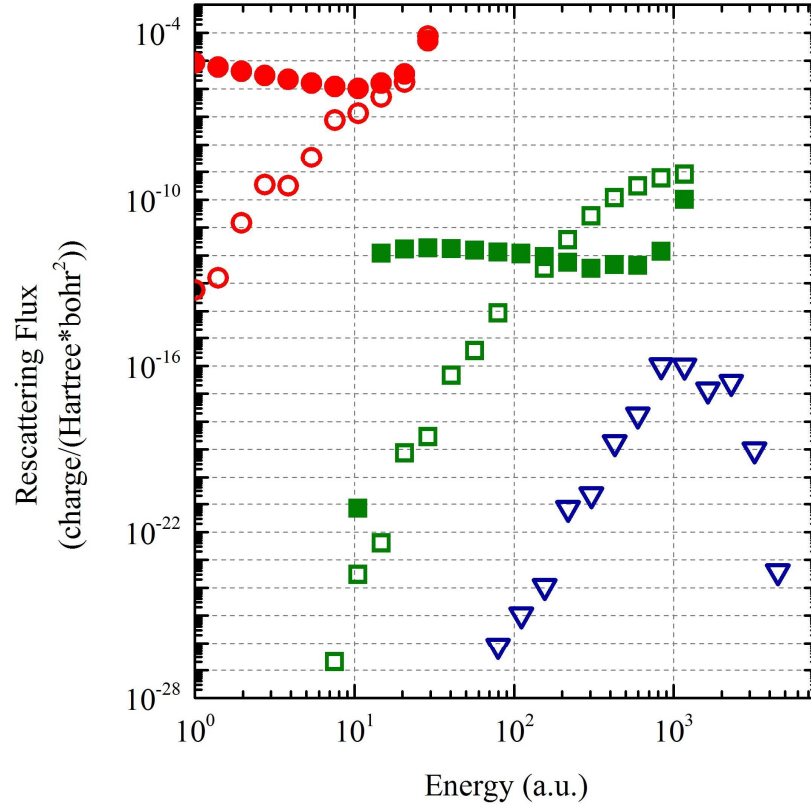


Figure 6.3 Rescattering flux as a function of returning energy for Ar^{4+} at $4.6 \times 10^{15} \text{ W/cm}^2$ and 800 nm (circle), Ar^{9+} at $1.6 \times 10^{18} \text{ W/cm}^2$ and 266 nm (square) and Ar^{9+} at $1.6 \times 10^{18} \text{ W/cm}^2$ and 800 nm (inverse triangle).

For the sake of completeness we show more scenarios with different ion species, intensity and wavelength in Figure 6.4.

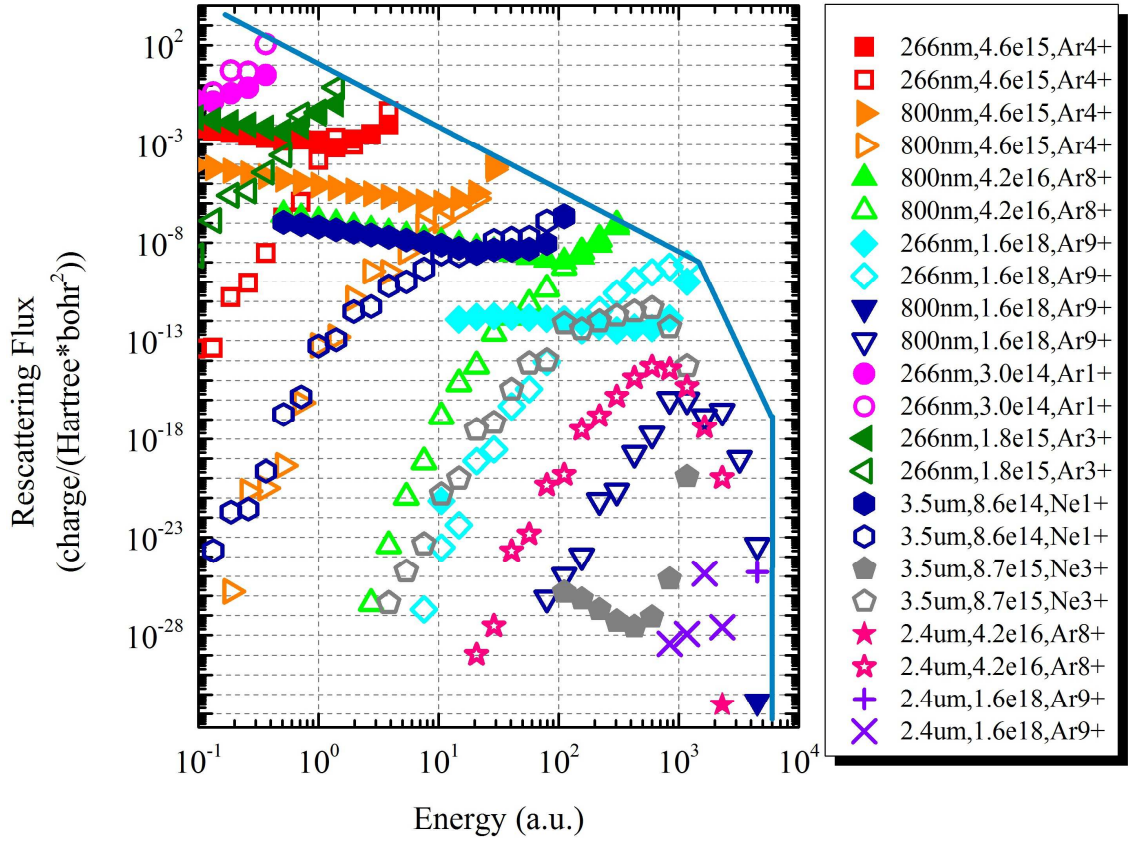


Figure 6.4 Rescattering flux as a function of returning energy for various scenarios.

The plot consists of three regions. From $\Gamma_r = 2.0 \times 10^{-6}$ a.u. (Ar^{1+} at 3.0×10^{14} W/cm² and 266 nm) to $\Gamma_r = 0.64$ a.u. (Ar^{8+} at 4.2×10^{16} W/cm² and 800 nm), the curves in this region are characterized as nonrelativistic. The curves in this region demonstrate the same structure and scale with pondermotive energy. The maximum of rescattering flux of each curve form a straight line which perfectly outlines the nonrelativistic region. Two scenarios, Ar^{9+} at 1.6×10^{18} W/cm² and 266 nm ($\Gamma_r = 3.1$ a.u.) and Ne^{3+} at 8.7×10^{15} W/cm² and 3.5 μm ($\Gamma_r = 14.6$ a.u.), are in the transition region, where the ‘short’ trajectories contribution start to dominate the

rescattering flux. The relativistic region is extended to Ar^{9+} at $1.6 \times 10^{18} \text{ W/cm}^2$ and $2.4 \mu\text{m}$ ($\Gamma_r = 20718 \text{ a.u.}$) where only ‘short’ trajectories contributions can be seen at 10^{-28} level. The relativistic region shows a distinct cutoff at $\sim 10^4 \text{ a.u.}$ where higher intensity and longer wavelength will not yield detectable rescattering flux.

After obtaining the energy spectrum of rescattering flux, we decide to calculate the inelastic impact ionization yield, which can be computed by multiplying the returning fluence with the inelastic impact ionization cross section. We use the formula defined in equation (2.23) to calculate the cross section and figure 6.5 shows the comparisons between calculated result and experimental results for the process of $\text{Ar}^{8+} + e \rightarrow \text{Ar}^{9+} + 2e$ ($2p^5$) process.

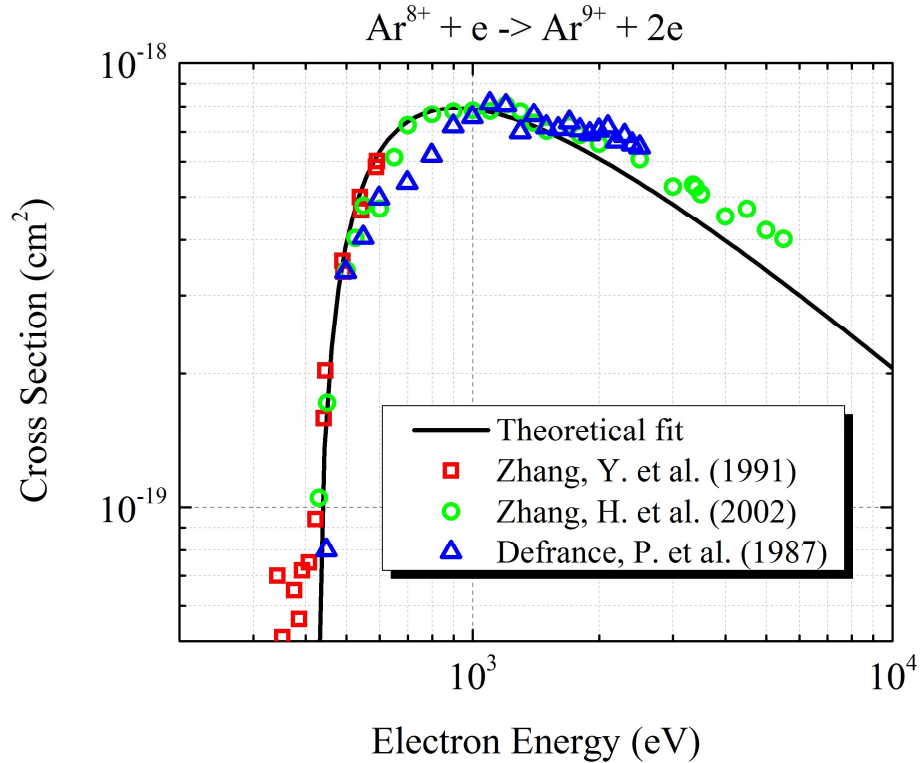


Figure 6.5 Cross section for $\text{Ar}^{8+} + e \rightarrow \text{Ar}^{9+} + 2e$ ($2p^5$) process.

Table 6.1 shows the calculated inelastic impact ionization yield for single atom single pulse response for the $(e, 2e)$ process of Ar^{8+} and Ar^{9+} . The ionization energy for $\text{Ar}^{8+} + e \rightarrow \text{Ar}^{9+} + 2e$ (1s), $\text{Ar}^{8+} + e \rightarrow \text{Ar}^{9+} + 2e$ (2s), $\text{Ar}^{9+} + e \rightarrow \text{Ar}^{10+} + 2e$ (1s), and $\text{Ar}^{9+} + e \rightarrow \text{Ar}^{10+} + 2e$ (2s) processes are 3.39×10^3 eV, 4.99×10^2 eV, 3.47×10^3 eV, and 5.53×10^2 eV, respectively. For the calculations of inelastic impact ionization cross section, the complete description of the atomic structure calculation based on Hartree Fock can be found in [12].

The 2s shell ionization generally has a higher yield than 1s shell due to its lower ionization potential. And for the same impact ionization process and same intensity, the yield decreases as wavelengths increases, which is consist with the fact that returning wavepacket spread scales with wavelength, and again demonstrates the effect of Lorentz deflection in the rescattering process. We believe table 6.1 will serve as guidance for experimentalists in searching for detectable inelastic impact ionization yield.

6.4 Conclusion

We study the rescattering flux for various ion species, intensity and wavelength, as one progress from nonrelativstic to relativist regime. The dynamics of electron wavepacket are investigated. It is found that an ultimate cutoff specifies the maximum returning kinetic energy with observable returning fluence. We expect this study facilitate the experimental research of inelastic scattering in the future.

Intensity	Wavelength	Begin State	End State	Yield
$4.2 \times 10^{16} \text{W/cm}^2$	266nm	$\text{Ar}^{8+} (2p^6)$	$\text{Ar}^{9+} (1s^1 2s^2 2p^6)$	0
$4.2 \times 10^{16} \text{W/cm}^2$	266nm	$\text{Ar}^{8+} (2p^6)$	$\text{Ar}^{9+} (2s^1 2p^6)$	9.54×10^{-6}
$4.2 \times 10^{16} \text{W/cm}^2$	800nm	$\text{Ar}^{8+} (2p^6)$	$\text{Ar}^{9+} (1s^1 2s^2 2p^6)$	6.57×10^{-10}
$4.2 \times 10^{16} \text{W/cm}^2$	800nm	$\text{Ar}^{8+} (2p^6)$	$\text{Ar}^{9+} (2s^1 2p^6)$	1.42×10^{-8}
$4.2 \times 10^{16} \text{W/cm}^2$	2400nm	$\text{Ar}^{8+} (2p^6)$	$\text{Ar}^{9+} (1s^1 2s^2 2p^6)$	8.14×10^{-17}
$4.2 \times 10^{16} \text{W/cm}^2$	2400nm	$\text{Ar}^{8+} (2p^6)$	$\text{Ar}^{9+} (2s^1 2p^6)$	1.03×10^{-15}
$1.6 \times 10^{18} \text{W/cm}^2$	266nm	$\text{Ar}^{9+} (2p^5)$	$\text{Ar}^{10+} (1s^1 2s^2 2p^5)$	1.75×10^{-11}
$1.6 \times 10^{18} \text{W/cm}^2$	266nm	$\text{Ar}^{9+} (2p^5)$	$\text{Ar}^{10+} (2s^1 2p^5)$	1.85×10^{-10}
$1.6 \times 10^{18} \text{W/cm}^2$	800nm	$\text{Ar}^{9+} (2p^5)$	$\text{Ar}^{10+} (1s^1 2s^2 2p^5)$	2.40×10^{-18}
$1.6 \times 10^{18} \text{W/cm}^2$	800nm	$\text{Ar}^{9+} (2p^5)$	$\text{Ar}^{10+} (2s^1 2p^5)$	2.37×10^{-17}
$1.6 \times 10^{18} \text{W/cm}^2$	2400nm	$\text{Ar}^{9+} (2p^5)$	$\text{Ar}^{10+} (1s^1 2s^2 2p^5)$	1.65×10^{-27}
$1.6 \times 10^{18} \text{W/cm}^2$	2400nm	$\text{Ar}^{9+} (2p^5)$	$\text{Ar}^{10+} (2s^1 2p^5)$	1.49×10^{-26}

Table 6.1 Calculated inelastic impact ionization for $\text{Ar}^{8+} + e \rightarrow \text{Ar}^{9+} + 2e$ and $\text{Ar}^{9+} + e \rightarrow \text{Ar}^{10+} + 2e$ process.

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Chapter 7

SUMMARY AND FUTURE STUDY

The works presented in this dissertation study the light-matter interaction in ultrastrong laser field. Theoretical studies are carried out to understand the relativistic effect and the influence of magnetic field on the atomic ionization, electron dynamics in the continuum, and rescattering process. In this chapter, we briefly summarize the findings in each work and address their significances. We finish the whole dissertation with discussing the future studies.

7.1 Atomic Ionization and Bound State Dynamics

We study the ionization of atoms in strong to ultrastrong circular polarized (CP) fields. A relativistic, three-dimensional trajectory ensemble method is used to account for the role of the laser magnetic field. The results show trajectories in ultrastrong fields are best characterized as chaotic when analyzed by Poincaré plots. Including the laser magnetic field can be observed to slightly change the distributions within these plots. A Fourier analysis of the trajectories does not reveal any significant change in the energy of the bound states due to the laser magnetic field as the energy shifts are linear in intensity well into the ultrastrong field regime. A result of the magnetic field was a stabilization of the atomic bound state as the Lorentz force preferentially aligned trajectories along z , perpendicular to the rotating x - y plane of the field where ionization occurs. The results indicate relativistic effects in ultrastrong

field ionization may most easily be seen with CP fields in experiments with ultrahigh intensities and occur at lower intensities than expected for LP light.

7.2 Elastic Rescattering in Ultrastrong Laser Field

A three-step model is extended into the relativistic, ultrastrong field regime ($\Gamma_r > 1$ and $a_0 < 10$). Continuum dynamics are treated semiclassically with Monte Carlo trajectory ensembles to account for relativistic and B effects while ionization and rescattering is treated quantum mechanically. Studies of scattering in hydrogenlike systems show elastic rescattering generally obeys a $1/I^2$ scaling when the Lorentz deflection is small, i.e., $\Gamma_r < 1$. Elastic scattering decreases roughly as a function of $\exp(-\Gamma_r)$ until becoming undetectable. Relativistic mass effects are noted but play a smaller role, contributing for intensities beyond 10^{18} W/cm^2 . In addition to work with fundamental hydrogenlike species, we calculated how the elastic scattering would be observed for noble gas species with screened atomic potentials. The results compare favorably with experimental data. The theory approach is well suited to modeling scattering in the ultrastrong intensity regime that lies between traditional strong fields and extreme relativistic interactions.

7.3 Future Studies

More groups around the globe, from USA to Japan, from China to Germany, are building ultrastrong laser systems to conduct experiments which will provide innovations for next generation technologies including laser based plasma acceleration, attosecond X-ray radiation generation, and laser fusion. We believe the works presented in this dissertation would be helpful to their research in the future in understanding the fundamental theories in ultraintense laser-matter interactions.

With the knowledge we already learned on returning fluence (Chapter 6), we plan to study the non-sequential ionization's wavelength dependence. It is known that the pondermotive energy scales as λ^2 and the spread of wavepacket in the continuum is also a function of wavelength. The maximum non-sequential ionization can be obtained by matching the returning fluence as a function of returning kinetic energy with the scattering cross section. This will enable the experimentalists to choose the optimal wavelength of different atom species to produce the maximum non-sequential ionization yield.

In order to calculate the non-sequential ionization contribution more accurately, our model should include not only inelastic impact ionization but also excitation ionization. It requires acquiring data of excitation ionization cross sections and an appropriate approach to calculate them. And studying the excitation ionization in ultrastrong field also involves understanding the non-dipole and relativistic effects.

Our current semi-classical model should be extended to deal with molecular response in ultrastrong laser field. Experiment of photoionization of chlorinated methane has been conducted and the corresponding theoretical follow up is required. Specifically, the theoretical modeling of multi-atom system and Coulomb explosion is needed.

Appendix A

SOURCE CODE FOR PHOTOELECTRON YIELD OF NEON

```
c      This Fortran 77 code calcutes the photoelectron energy spectra
of
c      the ionization of Neon atom in a linearly polarized laser field.
c      This program is largely based on the code of Isaac.
c      Writing code started in mid-august 2011 and the first beta
version
c      finished on Oct.20.2011.
c      Thanks for the help from @Patrick and @Nagitha~

c      Sui Luo
c      Department of Physics and Astronomy.University of Delaware.

      program ne_v13

c                                  !!!Neon!!!
c                                  !!!Circular Polarization!!!

*****
c                                  Declaration
c
*****

c      *****Common*****
double precision c,pi,freq,wavelength,e0,sigma,zr,s0
common/params1/ c,pi,freq,wavelength,e0,sigma,zr,s0

c      *****Rksuit_mod.f*****
integer neq,method,lenwrk,uflag,uflag3
parameter (neq=6,method=2,lenwrk=32*neq)
double precision hnext,hstart,tol,waste,twant,tnow
double precision thres(neq),work(lenwrk),traj(neq),
&      trajp(neq),trajmax(neq)
logical errass,mesage

c      *****Parameters of spatial discretion**
integer n_space,n_atom,n_atom_vol
integer atom_dep(19)
parameter (n_space=84)
double precision zd(n_space),vol_frac(n_space),sz(n_space),
&      rad1(n_space),rad2(n_space),vol,zd_delta,
&      int_ratio_array(10),int_ratio_1,int_ratio_2
```

```

c      *****Parameters of laser pulse*****
      integer n_time
      parameter (n_time=10000)
      double precision fwhm,int,int_au,e_mag,
&      int_si,int_array(51),int_array_si_max,
&      int_array_si_min,int_array_lg_del,
&      period
      double precision t_delta,t_start,t_final

c      *****Parameters of ADK & Probability***
      integer n_charge
      parameter (n_charge=10) !we go to Neon10+
      double precision rate_adk(n_charge),sum_adk(n_charge),
&      runinteg_adk(n_charge),sum_adk_diff,sum_ion,
&      prob_ion(n_charge+1),prob_ion_pre(n_charge+1),
&      prob_e,
&      prob_e_i(n_charge),prob_ion_sum,prob_ion_sum_pre
      double precision ip(n_charge),nstar(n_charge),c2nl(n_charge),
&      flm(n_charge)

c      *****Parameters of trajectory*****
      double precision t_integ_delta,t_integ_final
      double precision sz_ut,r_ut,gamma_ut
      double precision traj_start(neq),traj_rad,traj_phi

c      *****Parameters of angular distribution
      integer n_ang_array
      parameter (n_ang_array=1000000)!Max # of atom can use
      double precision
&
polar_mom_sptr_1(n_ang_array),polar_mom_sptr_2(n_ang_array),
&      azimu_mom_sptr_1(n_ang_array),azimu_mom_sptr_2(n_ang_array)
      double precision polar_mom_double,azimu_mom_double
      double precision polar_scan_min,polar_scan_max,polar_scan_del,
&      polar_scan_array(51),polar_time

c      *****Parameters for energy spectrum and intensity depend
      double precision theta,phi,inten_depend,gammalow,gammahigh,
&      gammalow_kin(4),gammahigh_kin(4)
      integer n_gamma_array
      parameter (n_gamma_array=50000000)!Max # of atom can use
      double precision
&      gamma_sptr_1(n_gamma_array),gamma_sptr_2(n_gamma_array),
&      gamma_sptr_3(n_gamma_array),gamma_sptr_4(n_gamma_array)
      double precision ener_au
      logical insidephi,insidetheta,insidegamma

c      *****Counters*****
      integer ii,tt,ixx,izz,hh,gg,zz,aa,cc,pp,uu,zztemp,c_start,
&      gammacount,angcount,azicount,ss,ee,ll
      logical calculate,outside

```

```

C      *****Misc parameters*****
      integer file1,file2,file3,file4,file5,file6,file7,file8,
&      file9,file10,file11,file12,file13,file14,file15,file16,
&      file17,file18,file19,file20
      parameter (file1=11,file2=12,file3=13,file4=14,file5=15,
&      file6=16,file7=17,file8=18,file9=19,file10=20,file11=21,
&      file12=22,file13=23,file14=24,file15=25,file16=26,
&      file17=27,file18=28,file19=29,file20=30)
      double precision adk_thresh,prob_e_thresh
      parameter(adk_thresh=1.0d-300,prob_e_thresh=1.0d-3)

C      random number generate
      integer*4 now(3)
      double precision seed_pe,randr,randphi,randz

C      file versions
      character*6 n_atomfile
      character*4 n_version
      character*2 n_intfile,n_polfile,n_rat1file,n_rat2file
      character*1 n_kinfile

C      elapse time
      real etime
      real elapsed(2)
      real total

C      input flag
      integer flag_int,flag_kin,flag_pol,flag_rat1,flag_rat2

C      *****Subroutines*****
      external adkrate,volume,emfield,rand,derivs,exitstatus,
&      angspectrum,setup,ut,filenamepara
      intrinsic log10,dsqrt,sin,cos,atan,itime

C
C      *****
C      Set parameters
C      *****
C
C      atom deployed array
      atom_dep(1)=1000
      atom_dep(2)=1808
      atom_dep(3)=2207
      atom_dep(4)=2540
      atom_dep(5)=2834
      atom_dep(6)=3100
      atom_dep(7)=3345
      atom_dep(8)=3573
      atom_dep(9)=3787
      atom_dep(10)=3990
      atom_dep(11)=4183
      atom_dep(12)=4368

```

```

atom_dep(13)=4545
atom_dep(14)=4715
atom_dep(15)=4879
atom_dep(16)=5038
atom_dep(17)=5192
atom_dep(18)=5342
atom_dep(19)=5488

c #####
c #####input values#####

flag_int=51
flag_kin=1
flag_pol=77
flag_rat1=1!>=1
flag_rat2=5!ratio2>ratio1
n_atom=atom_dep(1)!1000
n_version='ne13'
c polar_time=-4215.0d0

c #####
c #####

c set constants
c=137.03545d0
pi=4.0d0*ATAN(1.0d0)
int_au=6.43640931d15
ener_au=27.2113962d0

c set laser pulse parameters
fwhm=1653.65d0!corresponds to 40fs
wavelength=15118.015d0!corresponds to 800nm
freq=(2.0d0*pi*c)/wavelength
period=wavelength/c
sigma=fwhm/(2.0d0*dsqrt(2.0d0*abs(log(2.0d0))))!T0=2*sigma
s0=(2.0d0*2.0d0*wavelength)/pi!fnum=3.0d0 confirmed with
Nagitha
zr=(pi*s0**2.0d0)/wavelength

c set the gamma parameters
gammalow_kin(1)=52.5d0!for 75Kev
gammahigh_kin(1)=97.5d0!for 75Kev
gammalow_kin(2)=175.0d0 !for 250Kev
gammahigh_kin(2)=325.0d0!for 250Kev
gammalow_kin(3)=350.0d0!for 500Kev
gammahigh_kin(3)=650.0d0!for 500Kev
gammalow_kin(4)=700.0d0!for 1Mev
gammahigh_kin(4)=1300.0d0!for 1Mev

gammalow=gammalow_kin(flag_kin)
gammahigh=gammahigh_kin(flag_kin)

```

```

c      set the intensity scan window(parallel codes running needed)
      int_array_si_min=1.0d17
      int_array_si_max=2.0d19
      int_array_lg_del=(log10(int_array_si_max)-
&      log10(int_array_si_min))/50
      do ii=1,51
        int_array(ii)=(10.0d0**(log10(int_array_si_min)+
&      (ii-1)*int_array_lg_del))/int_au
      end do

c      set the polar scan window(parallel codes running needed)
c      polar_scan_min=10.0d0
c      polar_scan_max=100.0d0
c      polar_scan_del=(polar_scan_max-polar_scan_min)/dble(50)
c      do ll=1,51
c        polar_scan_array(ll)=polar_scan_min+dble(ll-1)*
c      &      polar_scan_del
c      end do

c      set forward angle and azimuthal central angle
      phi=10.0d0          !checked with Nagitha
      theta=77.5d0        !checked with Nagitha
c      theta=polar_scan_array(flag_pol)

c      set the intensity ratio of each shells
      int_ratio_1=dble(flag_rat1) !>=1
      int_ratio_2=dble(flag_rat2) !larger
c      int_ratio_2=1.1d0

c      set adk parameters for Neon
      ip(1)=0.79249674d0
      ip(2)=1.5053515d0
      ip(3)=2.3317466d0
      ip(4)=3.5692252d0
      ip(5)=4.6379742d0
      ip(6)=5.8038474d0
      ip(7)=7.6172165d0
      ip(8)=8.7867081d0
      ip(9)=43.945729d0
      ip(10)=50.059803d0

      nstar(1)=0.7943031d0
      nstar(2)=1.1526462d0
      nstar(3)=1.3892026d0
      nstar(4)=1.4971248d0
      nstar(5)=1.6416888d0
      nstar(6)=1.7610767d0
      nstar(7)=1.7934319d0
      nstar(8)=1.908367d0
      nstar(9)=0.95999545d0
      nstar(10)=0.99940251d0

```

```

c2nl(1)=4.2435496d0
c2nl(2)=3.6637011d0
c2nl(3)=2.9995501d0
c2nl(4)=2.6753008d0
c2nl(5)=2.2483399d0
c2nl(6)=1.9154493d0
c2nl(7)=1.8295331d0
c2nl(8)=1.5417285d0
c2nl(9)=4.0689704d0
c2nl(10)=4.0010967d0

flm(1)=3.0d0
flm(2)=3.0d0
flm(3)=3.0d0
flm(4)=3.0d0
flm(5)=3.0d0
flm(6)=3.0d0
flm(7)=1.0d0
flm(8)=1.0d0
flm(9)=1.0d0
flm(10)=1.0d0

C
*****
C
                                Open files
C
*****
C    generate file names parameters
    call filenamepara(flag_int,flag_kin,flag_rat1,flag_rat2,
&    flag_pol,n_atom,
&    n_intfile,n_kinfile,n_rat1file,n_rat2file,
&    n_polfile,n_atomfile)

    open(file3,file=n_version//'_intensity_depend_n'//n_atomfile
&    //'_int'//n_intfile//'_kin'//n_kinfile
&    //'_ratio'//n_rat1file//'_T'//n_rat2file
&    //'_polar'//n_polfile
&    //'_CP.dat')
    write(file3,1305)

    open(file10,file=n_version//'_ener_spectrum_n'//n_atomfile
&    //'_int'//n_intfile//'_kin'//n_kinfile
&    //'_ratio'//n_rat1file//'_T'//n_rat2file
&    //'_polar'//n_polfile
&    //'_CP.dat')
    write(file10,1321)

C    open(file12,file=n_version//'_polar_ang_momentum_spectrum_n'
C    &    //'_atomfile'//'_int'//n_intfile
C    &    //'_kin'//n_kinfile//'_CP.dat')
C    write(file12,1323)

```

```

c      open(file13,file=n_version//'_azimu_ang_momentum_spectrum_n'
c      &      //n_atomfile//'_int'//n_intfile
c      &      //'_kin'//n_kinfile//'_CP.dat')
c      write(file13,1325)

      open(file15,file=n_version//'_distribution_shape_n'//n_atomfile
&      //'_int'//n_intfile//'_kin'//n_kinfile
&      //'_ratio'//n_rat1file//'T'//n_rat2file
&      //'_polar'//n_polfile
&      //'_CP.dat')
      write(file15,1331)

c      open(file17,file=n_version//'_exit_status_n'//n_atomfile
c      &      //'_int'//n_intfile//'_kin'//n_kinfile
c      &      //'_ratio'//n_rat1file//'T'//n_rat2file
c      &      //'_polar'//n_polfile
c      &      //'_CP.dat')
c      write(file17,1333)

c
c*****
c                                  Main program begins
c
c*****
c      initialize random number generator
      call itime(now)
      seed_pe=dbl_e(now(1)+now(2)+now(3))
      call rand(seed_pe)

c      set the intensity
      int=int_array(flag_int)
      e0=dsqrt((8.0d0*pi*int)/c)

c      set rksuit_mod.f parameters
      tol=1.0d-6
      do tt=1,neq
          thres(tt)=1.0d-12
      end do

c      initialize intensity dependence container
      inten_depend=0.0d0

c      initialize the angular distribution containers
c      do hh=1,n_ang_array
c          polar_mom_spstr_1(hh)=0.0d0
c          polar_mom_spstr_2(hh)=0.0d0
c          azimu_mom_spstr_1(hh)=0.0d0
c          azimu_mom_spstr_2(hh)=0.0d0
c      end do
c      angcount=1
c      azicount=1

```

```

c      initialize the energy spectrum containers
do gg=1,n_gamma_array
    gamma_sptr_1(gg)=0.0d0
    gamma_sptr_2(gg)=0.0d0
    gamma_sptr_3(gg)=0.0d0
    gamma_sptr_4(gg)=0.0d0
end do
gammacount=1

c
*****
c      discretize the space into a set of discs
call volume(n_space,zd,zd_delta,sz,rad1,rad2,vol,vol_frac,
&          int_ratio_1,int_ratio_2)

c
*****
c      scan the space defined by the above subroutine.the
c      number of atoms in a certain disc is proportional to its
c      volume.for each atom in a given disc,ADK rate and ionization
c      probability are calculated to determine whether a "significant"
c      ionization is triggered.if triggered an electron is freed
c      (initially zero velocity)
c      and its trajectory is recorded by integrating the Newton's
c      dynamic equation with rksuit_mod.f.finally the energy,momentum
c      and angular spectrum for the atom ensemble can be obtained.

c      initialize parameters
calculate=.false.

c      scan of space starts
do 22 zz=1,n_space
    n_atom_vol=nint(n_atom*vol_frac(zz))
    write(file15,1332) zd(zz),sz(zz),rad1(zz),rad2(zz),
&          vol_frac(zz),dble(n_atom_vol)

c      scan of a certain disc starts
do 221 aa=1,n_atom_vol

c      initialize the position and momentum of atom
traj_start(1)=0.0d0
traj_start(2)=0.0d0
traj_start(3)=0.0d0
call rand(randr)
traj_rad=rad1(zz)+sqrt(randr)*(rad2(zz)-rad1(zz))
call rand(randphi)
traj_phi=2.0d0*pi*randphi
traj_start(4)=cos(traj_phi)*traj_rad
traj_start(5)=sin(traj_phi)*traj_rad
call rand(randz)
traj_start(6)=zd(zz)+zd_delta*
&          (-1.0d0+randz*(1.0d0-(-1.0d0)))

```

```

c      initialize the variables regarding adk rate and ion prob
      do cc=1,n_charge
        sum_adk(cc)=0.0d0
        runinteg_adk(cc)=0.0d0
      end do
      prob_ion(1)=1.0d0
      prob_ion_pre(1)=1.0d0
      do cc=2,n_charge+1
        prob_ion(cc)=0.0d0
        prob_ion_pre(cc)=0.0d0
      end do
      sum_ion=1.0
      prob_e=0.0d0

c      set time step
      t_delta=(12.0d0*sigma)/n_time
      t_integ_delta=period/50.0

c      scan of pulse starts
      do 2211 pp=1,n_time
        t_start=-6.0d0*sigma+pp*t_delta
        t_final=6.0d0*sigma
        t_integ_final=t_final+10.0d0*t_integ_delta

c      calculate adk rate and ionization probability to determine
c      whether to free electron. the following derivation follows
c      page 25~27 in David Neal Fittinghoff's Ph.D. Thesis Dec 1993

c      calculate adk rate
      do cc=1,n_charge
        c_start=cc
        call adkrate(c_start,ip(c_start),nstar(c_start),
&                c2nl(c_start),flm(c_start),traj_start,
&                t_start,rate_adk(c_start),e_mag)
      end do

c      calculate running integral
      do cc=1,n_charge
        sum_adk(cc)=sum_adk(cc)+rate_adk(cc)*t_delta
        if(sum_adk(cc).lt.adk_thresh) then
          sum_adk(cc)=0.0d0
        end if
      end do

c      calculate the population of each ion state
      prob_ion(1)=dexp(-sum_adk(1))
      do cc=2,n_charge
        if(sum_adk(cc).gt.700) then
          prob_ion(cc)=0.0d0
        else
          sum_adk_diff=sum_adk(cc)-sum_adk(cc-1)

```

```

        runinteg_adk(cc)=runinteg_adk(cc)+
&         (dexp(sum_adk_diff)*rate_adk(cc-1)*
&         t_delta)
        prob_ion(cc)=dexp(-sum_adk(cc))*
&         runinteg_adk(cc)
    end if
end do
prob_ion(n_charge+1)=prob_ion(n_charge+1)+
&     (rate_adk(n_charge)*prob_ion(n_charge)*t_delta)

c    record the total ion population
    sum_ion=0.0d0
    do cc=1,n_charge+1
        sum_ion=sum_ion+prob_ion(cc)
    end do

c    record the photonelectron probability(new method)
    do ee=1,n_charge
        prob_e_i(ee)=0.0d0
        prob_ion_sum=0.0d0
        prob_ion_sum_pre=0.0d0

        do cc=ee+1,n_charge+1
            prob_ion_sum=prob_ion_sum+prob_ion(cc)
            prob_ion_sum_pre=
&                prob_ion_sum_pre+prob_ion_pre(cc)
            if(prob_ion_sum.gt.prob_ion_sum_pre) then
&                prob_e_i(ee)=
&                prob_ion_sum-prob_ion_sum_pre
            end if
        end do
    end do

    prob_e=0.0d0
    do ee=1,n_charge
        prob_e=prob_e+prob_e_i(ee)
    end do

c    update the population of each ion state
    do cc=1,n_charge+1
        prob_ion_pre(cc)=prob_ion(cc)
    end do

c    determine whether to free electron
    if(prob_e.gt.prob_e_thresh) then
        calculate=.true.
    end if

c    electron trajectory integration starts
    do while(calculate.EQV..true.)

c    set rksuite.f parameters

```

```

        mesage=.true.
        errass=.false.
        hstart=0.0d0

c      call setup for ut rksuite.f
        call setup(neq,t_start,traj_start,t_integ_final,
&               tol,thres,method,'Usual Task',errass,hstart,
&               work,lenwrk,mesage)

c      initialize counter
        uflag3=0
        outside=.false.
        insidephi=.false.
        insidetheta=.false.
        insidegamma=.false.

        twant=t_start
c      integrate the trajectory with ut rksuite.f
        do while((twant.lt.t_final).and.
&               (outside.eqv..false.))

        twant=twant+t_integ_delta

        call ut(derivs,twant,tnow,traj,trajp,trajmax,
&               work,uflag)

        if(uflag.gt.3) go to 301

        sz_ut=s0*dsqrt(1.0d0+(traj(6)/zr)**2.0d0)
        r_ut=dsqrt(traj(4)**2.0d0+traj(5)**2.0d0)
        gamma_ut=(dsqrt(traj(1)**2.0d0+traj(2)**2.0d0+
&               traj(3)**2.0d0+c**2.0d0)*c-c**2.0d0)*
&               ener_au/1.0d3
&               !This is kinetic energy!in KeV.

c      if the electron leaves the beam region
        if (r_ut.gt.(2.0d0*sz_ut)) then
            outside=.true.

c      check the exit status
        call exitstatus(traj(1),traj(2),traj(3),
&               gammalow,gammahigh,gamma_ut,
&               theta,phi,
&               insidephi,insidetheta,insidegamma)

c      if record the intensity dependence
        if ((insidephi.eqv..true.).and.
&           (insidetheta.eqv..true.).and.
&           (insidegamma.eqv..true.)) then
c      if ((insidephi.eqv..true.).and.
c      &           (insidetheta.eqv..true.)) then
c      if (t_start.gt.polar_time) then

```

```

                                inten_depend=inten_depend+prob_e
c                                end if
                                end if

                                call angpectrum(traj(1),traj(2),traj(3),
&                                polar_mom_double,azimu_mom_double)

c    if record the energy spectra
c                                if((insidephi.eqv..true.).and.
c                                &                                (insidetheta.eqv..true.)) then
c                                if (insidetheta.eqv..true.) then
                                gamma_sptr_1(gammacount)=
&                                gamma_ut !in KeV
                                gamma_sptr_2(gammacount)=prob_e
                                gamma_sptr_3(gammacount)=polar_mom_double
                                gamma_sptr_4(gammacount)=azimu_mom_double
                                gammacount=gammacount+1
c                                end if

c    if record the polar angular distribution(CP light)
c                                if((insidephi.eqv..true.).and.
c                                &                                (insidegamma.eqv..true.)) then
c                                if(insidegamma.eqv..true.) then
                                call angpectrum(traj(1),traj(2),traj(3),
c                                &                                polar_mom_double,azimu_mom_double)
c                                &                                polar_mom_sptr_1(angcount)=
c                                &                                polar_mom_double
c                                &                                polar_mom_sptr_2(angcount)=
c                                &                                prob_e
c                                angcount=angcount+1
c                                end if

c    if record the azimuthal angular distribution(CP light)
c                                if(insidegamma.eqv..true.) then
                                call angpectrum(traj(1),traj(2),traj(3),
c                                &                                polar_mom_double,azimu_mom_double)
c                                &                                azimu_mom_sptr_1(azicount)=
c                                &                                azimu_mom_double
c                                &                                azimu_mom_sptr_2(azicount)=
c                                &                                prob_e
c                                azicount=azicount+1
c                                end if

                                goto 301

                                end if

                                end do

c    post-check*****
c    check the exit status

```

```

                                call exitstatus(traj(1),traj(2),traj(3),
&                                gammalow,gammahigh,gamma_ut,
&                                theta,phi,
&                                insidephi,insidetheta,insidegamma)

c    if record the intensity dependence
                                if ((insidephi.eqv..true.).and.
&                                (insidetheta.eqv..true.).and.
&                                (insidegamma.eqv..true.)) then
c                                if (t_start.gt.polar_time) then
                                    inten_depend=inten_depend+prob_e
c                                end if
                                end if

                                call angpectrum(traj(1),traj(2),traj(3),
&                                polar_mom_double,azimu_mom_double)

c    if record the energy spectra
c                                if((insidephi.eqv..true.).and.
c                                &                                (insidetheta.eqv..true.)) then
c                                if (insidetheta.eqv..true.) then
                                    gamma_sptr_1(gammacount)=
&                                gamma_ut !in KeV
                                    gamma_sptr_2(gammacount)=prob_e
                                    gamma_sptr_3(gammacount)=polar_mom_double
                                    gamma_sptr_4(gammacount)=azimu_mom_double
                                    gammacount=gammacount+1
c                                end if

301                                continue

                                calculate=.false.

c    end of one trajectory integration
                                end do

c    end of pulse scan
2211                                continue

c    end of disc volume scan

221                                continue

c    end of space scan
22                                continue

c
*****
c                                Output files
c
*****
                                close(file17)

```

```

c      output angular distribution
c      do hh=1,n_ang_array
c          if (polar_mom_sptr_1(hh).gt.0.0d0) then
c              write(file12,1324) polar_mom_sptr_1(hh),
c              &          polar_mom_sptr_2(hh)
c          end if
c      end do
c      close(file12)

c      output angular distribution
c      do hh=1,n_ang_array
c          if (azimu_mom_sptr_1(hh).gt.0.0d0) then
c              write(file13,1326) azimu_mom_sptr_1(hh),
c              &          azimu_mom_sptr_2(hh)
c          end if
c      end do
c      close(file13)

c      output energy spectrum
do gg=1,n_gamma_array
    if (gamma_sptr_1(gg).gt.0.0d0) then
        write(file10,1322) gamma_sptr_1(gg),gamma_sptr_2(gg),
&          gamma_sptr_3(gg),gamma_sptr_4(gg)
    end if
end do
close(file10)

c      Intensity Dependence Output
int_si=int*int_au
write(file3,1306) int_si,inten_depend,theta
close(file3)

C
*****
C                                  Format statements
C
*****
1305 format(' ', 'Int_SI', 3x, 'Prob_PE', 3x, 'Theta')
1306 format(' ', E16.8, 3x, E16.8, 3x, E16.8)
1321 format(' ', 'Kinetic', 3x, 'Prob', 3x, 'polar', 3x, 'azimu')
1322 format(' ', 4(E16.8, 3x))
1323 format(' ', 'Polar_angle', 3x, 'Prob')
1324 format(' ', E16.8, 3x, E16.8)
1325 format(' ', 'Azimu_angle', 3x, 'Prob')
1326 format(' ', E16.8, 3x, E16.8)
1331 format(' ', 'zd', 3x, 'sz', 3x, 'rad1', 3x, 'rad2', 3x,
&          'vol_frac', 3x, 'n_atom')
1332 format(' ', 6(E16.8, 3x))
1333 format(' ', 'Time', 3x, 'px', 3x, 'py', 3x, 'pz', 3x, 'Kin')
1334 format(' ', 5(E16.8, 3x))

```

```

C
*****

C      display elapsed time
      total=etime(elapsed)
      print 1399,'Program Ends: ',
&        ' Total Elapsed Time =',total,
&        ' User Time =',elapsed(1),
&        ' System Time =',elapsed(2)
1399 format(' ',A,/,A,F9.3,/,A,F9.3,/,A,F9.3)

C      program ends

      end

C      end of the main program
C
*****

C
////////////////////////////////////
C
////////////////////////////////////Subroutines////////////////////////////////////
C
////////////////////////////////////

C
////////////////////////////////////

C
////////////////////////////////////
This subroutine calculates the ADK tunneling rate
C
////////////////////////////////////

      subroutine adkrate(zzz,ip,nstar,c2n1,flm,traj_start,t_start,
&        rate_adk,e_mag)
      integer zzz
      double precision ip,e_mag,nstar,c2n1,flm,rate_adk
      double precision epsilon,factor,nmpower1,rate1_adk,
&        traj_start(*),t_start,e_cpn(6),eff
      double precision c,pi,freq,wavelength,e0,sigma,zr,s0
      common/params1/ c,pi,freq,wavelength,e0,sigma,zr,s0

      call emfield(traj_start(4),traj_start(5),traj_start(6),t_start,
&        e_cpn,eff)
      e_mag=abs(dsqrt(e_cpn(1)**2+e_cpn(2)**2+e_cpn(3)**2))

      epsilon=(2.0d0*ip)**1.5
      factor=epsilon/abs(e_mag)
      nmpower1=2.0d0*nstar-1.0d0!assume m=0 for all charge state

```

```

    rate_adk=c2n1*dsqrt(3.0d0/(pi*factor))*ip*
&    flm*((2.0d0*factor)**nmpower1)*
&    exp(-(2.0d0*factor)/3.0d0)

    return
end

c    end of subroutine adkrate
c
////////////////////////////////////

c
////////////////////////////////////
c    This subroutine discretize the space
c
////////////////////////////////////

    subroutine volume(n_space,zd,zd_delta,sz,rad1,rad2,vol,vol_frac,
&    int_ratio_1,int_ratio_2)

    double precision zd(*),vol1,vol2,vol,vol_frac(*),
&    int_ratio_1,int_ratio_2,
&    zd_range_1,zd_range_2,zd_delta,
&    sz(*),xi1,xi2,rad1(*),rad2(*)
    integer n_space,dd
    double precision c,pi,freq,wavelength,e0,sigma,zr,s0
    common/params1/ c,pi,freq,wavelength,e0,sigma,zr,s0

    xi1=dsqrt(int_ratio_1-1.0d0)
    xi2=dsqrt(int_ratio_2-1.0d0)
    vol1=((pi**2)*(s0**4))/wavelength)*(((2.0d0/9.0d0)*(xi1**3))+
&    ((4.0d0/3.0d0)*(xi1-atan(xi1))))
    vol2=((pi**2)*(s0**4))/wavelength)*(((2.0d0/9.0d0)*(xi2**3))+
&    ((4.0d0/3.0d0)*(xi2-atan(xi2))))
    vol=vol2-vol1
    zd_range_1=xi1*zr
    zd_range_2=xi2*zr
    zd_delta=(2.0d0*zd_range_2)/(n_space+1)

    do dd=1,n_space
        zd(dd)=-zd_range_2+dd*zd_delta
        sz(dd)=s0*dsqrt(1.0d0+((zd(dd)/zr)**2))
        if((zd(dd).le.-zd_range_1).or.(zd(dd).ge.zd_range_1)) then
            rad1(dd)=0.0
        else
            rad1(dd)=
&            sz(dd)*dsqrt(abs(dlog(s0*dsqrt(int_ratio_1)/sz(dd))))
        end if
        rad2(dd)=

```

```

&      sz(dd)*dsqrt(abs(dlog(s0*dsqrt(int_ratio_2)/sz(dd))))
      vol_frac(dd)=(pi*(rad2(dd)**2-rad1(dd)**2)*zd_delta)/vol
end do

return
end

c      end of subroutine volumn
c
c      //////////////////////////////////////
c
c      //////////////////////////////////////
c      This subroutine calculates the efield in space
c      //////////////////////////////////////

      subroutine emfield(x,y,z,t,e_cpn,eff)

      double precision x,y,z,t,e_cpn(6),e_cpn1(6),e_cpn2(6),
&      sz_t,eff,phase1,phase2,alpha,r2,eps
      double precision c,pi,freq,wavelength,e0,sigma,zr,s0
      common/params1/ c,pi,freq,wavelength,e0,sigma,zr,s0
c      //////////////////////////////////////
      sz_t=s0*dsqrt(1.0d0+(z/zr)**2.0d0)
      alpha=atan(z/zr)
      r2=(x**2.0d0)+(y**2.0d0)
      eff=e0*(s0/sz_t)*dexp(-r2/(sz_t**2.0d0))*
&      dexp(-(z/c-t)/(2*sigma))**2.0d0)
      phase1=freq*t-(freq/c)*z-(z*r2)/(zr*sz_t**2.0d0)
      phase2=freq*t-(freq/c)*z-(z*r2)/(zr*sz_t**2.0d0)+pi/2.0d0
      eps=wavelength/(2.0d0*pi*s0)

c      linear light 1*****
c      //////////////////////////////////////
c      //////////////////////////////////////
c      //////////////////////////////////////
c      efield x',y',z'
      e_cpn1(1)=eff*(cos(phase1+alpha)+eps**2*
&      (((2.0d0*x**2+r2)/(sz_t**2))*cos(phase1+3.0d0*alpha)-
&      ((r2**2)/(sz_t**3*s0))*cos(phase1+4.0d0*alpha)))
      e_cpn1(2)=eff*eps**2*((2.0d0*x*y)/(sz_t**2))*
&      cos(phase1+3.0d0*alpha)
      e_cpn1(3)=- (2.0d0*eff*eps/sz_t)*(sin(phase1+2.0d0*alpha)+
&      (eps**2*r2/(sz_t**2))*(3.0d0*sin(phase1+4.0d0*alpha)-
&      (r2/(sz_t*s0))*cos(phase1+5.0d0*alpha)))*x
c      bfield x',y',z'
      e_cpn1(4)=e_cpn1(2)/c
      e_cpn1(5)=(eff/c)*(cos(phase1+alpha)+eps**2*
&      (((2.0d0*y**2+r2)/(sz_t**2))*cos(phase1+3.0d0*alpha)-

```

```

&      ((r2**2)/(sz_t**3*s0))*cos(phase1+4.0d0*alpha)))
e_cpn1(6)=- (2.0d0*(eff/c)*eps/sz_t)*(sin(phase1+2.0d0*alpha)+
&      (eps**2*r2/(sz_t**2))*(3.0d0*sin(phase1+4.0d0*alpha)-
&      (r2/(sz_t*s0))*cos(phase1+5.0d0*alpha)))*y

c      linear light *****
c      //////////x'=y;////////
c      //////////y'=-x;////////
c      //////////z'=z;////////
c      efield x'',y'',z''
e_cpn2(1)=eff*(cos(phase2+alpha)+eps**2*
&      ((2.0d0*y**2+r2)/(sz_t**2))*cos(phase2+3.0d0*alpha)-
&      ((r2**2)/(sz_t**3*s0))*cos(phase2+4.0d0*alpha)))
e_cpn2(2)=eff*eps**2*((2.0d0*y*(-x))/(sz_t**2))*
&      cos(phase2+3.0d0*alpha)
e_cpn2(3)=- (2.0d0*eff*eps/sz_t)*(sin(phase2+2.0d0*alpha)+
&      (eps**2*r2/(sz_t**2))*(3.0d0*sin(phase2+4.0d0*alpha)-
&      (r2/(sz_t*s0))*cos(phase2+5.0d0*alpha)))*y
c      bfield x'',y'',z''
e_cpn2(4)=e_cpn2(2)/c
e_cpn2(5)=(eff/c)*(cos(phase2+alpha)+eps**2*
&      ((2.0d0*(-x)**2+r2)/(sz_t**2))*cos(phase2+3.0d0*alpha)-
&      ((r2**2)/(sz_t**3*s0))*cos(phase2+4.0d0*alpha)))
e_cpn2(6)=- (2.0d0*(eff/c)*eps/sz_t)*(sin(phase2+2.0d0*alpha)+
&      (eps**2*r2/(sz_t**2))*(3.0d0*sin(phase2+4.0d0*alpha)-
&      (r2/(sz_t*s0))*cos(phase2+5.0d0*alpha)))*(-x)

c      if linear light*****
c      e_cpn(1)=e_cpn1(1)
c      e_cpn(2)=e_cpn1(2)
c      e_cpn(3)=e_cpn1(3)
c      e_cpn(4)=e_cpn1(4)
c      e_cpn(5)=e_cpn1(5)
c      e_cpn(6)=e_cpn1(6)

c      if circular light*****
e_cpn(1)=e_cpn1(1)-e_cpn2(2)
e_cpn(2)=e_cpn1(2)+e_cpn2(1)
e_cpn(3)=e_cpn1(3)+e_cpn2(3)
e_cpn(4)=e_cpn1(4)-e_cpn2(5)
e_cpn(5)=e_cpn1(5)+e_cpn2(4)
e_cpn(6)=e_cpn1(6)+e_cpn2(6)

return
end

c      end of subroutine emfield
c
//////////

```

```

c
//////////
c      This subroutine defines the first order derivative equation
used
c      by ut rksuite.f
c
//////////

      subroutine derivs(tgot,ygot,ypgot)

      double precision tgot,ygot(*),ypgot(*),e_cpn(6),gamma_c,
&      ex,ey,ez,bx,by,bz,eff
      double precision c,pi,freq,wavelength,e0,sigma,zr,s0
      common/params1/ c,pi,freq,wavelength,e0,sigma,zr,s0

      call emfield(ygot(4),ygot(5),ygot(6),tgot,e_cpn,eff)
      ex=e_cpn(1)
      ey=e_cpn(2)
      ez=e_cpn(3)
      bx=e_cpn(4)
      by=e_cpn(5)
      bz=e_cpn(6)
      gamma_c=dsqrt((ygot(1))**2.0d0+(ygot(2))**2.0d0+
&      (ygot(3))**2.0d0+c**2.0d0)
c      mention:gamma_c is different from the gamma in the main code,
c      gamma=gamma_c/c
      ypgot(4)=ygot(1)*c/gamma_c
      ypgot(5)=ygot(2)*c/gamma_c
      ypgot(6)=ygot(3)*c/gamma_c
      ypgot(1)=-ex-(ygot(2)*bz-ygot(3)*by)*c/gamma_c
      ypgot(2)=-ey-(ygot(3)*bx-ygot(1)*bz)*c/gamma_c
      ypgot(3)=-ez-(ygot(1)*by-ygot(2)*bx)*c/gamma_c

      return
      end

c      end of subroutine derivs
c
//////////

c
//////////
c      This subroutine calculates the exit status of an electron
c
//////////

      subroutine exitstatus(px,py,pz,
&      gammalow,gammahigh,gamma,theta,phi,
&      insidetheta,insidegamma)

```



```

double precision xda,yda,zda,polar_temp,azimu_temp
double precision c,pi,freq,wavelength,e0,sigma,zr,s0
double precision polar_mom_double,azimu_mom_double
double precision prad
common/params1/ c,pi,freq,wavelength,e0,sigma,zr,s0

c      when the electron leaves the beam, xd,yd,zd cannot equal to
zero

prad=sqrt(xda**2.0d0+yda**2.0d0)

if(zda.gt.0.0d0.and.xda.gt.0.0d0) then
  polar_temp=atan(prad/abs(zda))*(360.0d0/(2.0d0*pi))
else if(zda.lt.0.0d0.and.xda.gt.0.0d0) then
  polar_temp=180.0d0-
&      atan(prad/abs(zda))*(360.0d0/(2.0d0*pi))
else if(zda.lt.0.0d0.and.xda.lt.0.0d0) then
  polar_temp=180.0d0+
&      atan(prad/abs(zda))*(360.0d0/(2.0d0*pi))
else if(zda.gt.0.0d0.and.xda.lt.0.0d0) then
  polar_temp=360.0d0-
&      atan(prad/abs(zda))*(360.0d0/(2.0d0*pi))
end if

polar_mom_double=polar_temp

if(xda.gt.0.0d0.and.yda.gt.0.0d0) then
  azimu_temp=atan(abs(yda)/abs(xda))*(360.0d0/(2.0d0*pi))
else if(xda.lt.0.0d0.and.yda.gt.0.0d0) then
  azimu_temp=180.0d0-
&      atan(abs(yda)/abs(xda))*(360.0d0/(2.0d0*pi))
else if(xda.lt.0.0d0.and.yda.lt.0.0d0) then
  azimu_temp=180.0d0+
&      atan(abs(yda)/abs(xda))*(360.0d0/(2.0d0*pi))
else if(xda.gt.0.0d0.and.yda.lt.0.0d0) then
  azimu_temp=360.0d0-
&      atan(abs(yda)/abs(xda))*(360.0d0/(2.0d0*pi))
end if

azimu_mom_double=azimu_temp

return
end

c      end of subroutine angpectrum
c
c      //////////////////////////////////////

c
c      //////////////////////////////////////
c      This subroutine generates file name parameters

```

```

C
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%

      subroutine filenamepara(flag_int,flag_kin,flag_rat1,flag_rat2,
&      flag_pol,n_atom,
&      n_intfile,n_kinfile,n_rat1file,n_rat2file,
&      n_polfile,n_atomfile)

      integer flag_int,flag_kin,flag_rat1,flag_rat2,flag_pol,n_atom
      character*1 atomfile1,tempzero1,intfile1,ratfile1,polfile1,
&      n_kinfile
      character*2 atomfile2,tempzero2,n_intfile,n_rat1file,n_rat2file,
&      n_polfile
      character*3 atomfile3,tempzero3
      character*4 atomfile4,tempzero4
      character*5 atomfile5,tempzero5
      character*6 n_atomfile

      tempzero1='0'
      tempzero2='00'
      tempzero3='000'
      tempzero4='0000'
      tempzero5='00000'

      write(n_kinfile,'(I1)') flag_kin

      if(flag_int.lt.10) then
         write(intfile1,'(I1)') flag_int
         n_intfile=tempzero1//intfile1
      else
         write(n_intfile,'(I2)') flag_int
      end if

      if(flag_pol.lt.10) then
         write(polfile1,'(I1)') flag_pol
         n_polfile=tempzero1//polfile1
      else
         write(n_polfile,'(I2)') flag_pol
      end if

      if(flag_rat1.lt.10) then
         write(ratfile1,'(I1)') flag_rat1
         n_rat1file=tempzero1//ratfile1
      else
         write(n_rat1file,'(I2)') flag_rat1
      end if

      if(flag_rat2.lt.10) then
         write(ratfile1,'(I1)') flag_rat2
         n_rat2file=tempzero1//ratfile1
      else
         write(n_rat2file,'(I2)') flag_rat2
      end if

```

```

end if

if(n_atom.lt.10) then
  write(atomfile1,'(I1)') n_atom
  n_atomfile=tempzero5//atomfile1
else if(n_atom.ge.10.and.n_atom.lt.100) then
  write(atomfile2,'(I2)') n_atom
  n_atomfile=tempzero4//atomfile2
else if(n_atom.ge.100.and.n_atom.lt.1000) then
  write(atomfile3,'(I3)') n_atom
  n_atomfile=tempzero3//atomfile3
else if(n_atom.ge.1000.and.n_atom.lt.10000) then
  write(atomfile4,'(I4)') n_atom
  n_atomfile=tempzero2//atomfile4
else if(n_atom.ge.10000.and.n_atom.lt.100000) then
  write(atomfile5,'(I5)') n_atom
  n_atomfile=tempzero1//atomfile5
else if(n_atom.ge.100000.and.n_atom.lt.1000000) then
  write(n_atomfile,'(I6)') n_atom
end if

return
end

c      end of subroutine filenamepara
c
////////////////////////////////////
c
////////////////////////////////////
c      THIS SUBROUTINE GENERATES A RANDUM NUMBER IN THE RANGE [0,1].
c      FROM FORTRAN 77 FOR ENGINEERS AND SCIENTISTS 4TH ED., BY NYHOFF
c      AND LEESTMA PAGE 420
c
////////////////////////////////////

SUBROUTINE RAND(randnum)

INTEGER M,CONST1
DOUBLE PRECISION randnum,CONST2
PARAMETER (CONST1=2147483647,CONST2=0.4656613D-9)
SAVE
DATA M /0/

IF(M.EQ.0) M=INT(randnum)
M=M*65539
IF(M.LT.0) M=(M+1)+CONST1
randnum=M*CONST2

RETURN
END

```

```
C      END OF SUBROUTINE RAND
```

```
C
```

```
////////////////////////////////////
```

Appendix B

SOURCE CODE FOR ELASTIC SCATTERING

```
c      This program is for elastic-rescattering project.
c      Code based on resc_v1.cpp and ne_v25.f.
c      Incorporate elsepa project for cross section calculation.
c      Sui Luo
c      Department of Physics and Astronomy.University of Delaware.
c      Last updated: 17:15PM 01/13/2015

      INCLUDE 'elsepa.f'
      INCLUDE 'rksuit_mod.f'
      program resc_v3

c
c      *****
c
c      Declaration
c
c      *****

c      *****ELSEPA*****
c      IMPLICIT DOUBLE PRECISION (A-B,D-H,O-Z), COMPLEX*16 (C),
1      INTEGER*4 (I-N)
c      **** Constants.
c      PARAMETER (A0B=5.291772083D-9) ! Bohr radius (cm)
c      PARAMETER (A0B2=A0B*A0B)
c      PARAMETER (PI=3.1415926535897932D0)
c      **** Results from the partial wave calculation.
c      PARAMETER (NGT=650)
c      COMMON/DCSTAB/ECS,TCS1,TCS2,TH(NGT),XT(NGT),DCST(NGT),SPOL(NGT),
1      ERROR(NGT),NTAB
c      COMMON/CTOTCS/TOTCS,ABCS
c      **** Phase shifts.
c      PARAMETER (NDM=25000)
c      COMMON/PHASES/DP(NDM),DM(NDM),NPH,ISUMP
c      COMMON/PHASEI/DPJ(NDM),DMJ(NDM)

      DOUBLE PRECISION CROSAMP

c      *****Common*****
c      double precision c,pi,freq,wavelength,e0,sigma,zr,s0
c      common/params1/ c,pi,freq,wavelength,e0,sigma,zr,s0

      double precision iniphase
```

```

common/params2/ iniphase

C *****Rksuit_mod.f*****
integer neq,method,lenwrk,uflag,uflag3
parameter (neq=6,method=2,lenwrk=32*neq)
double precision hnext,hstart,tol,waste,twant,tnow
double precision thres(neq),work(lenwrk),traj(neq),
&      trajp(neq),trajmax(neq)
logical errass,mesage

C *****Parameters of laser pulse*****
integer n_time
parameter (n_time=30000)
double precision fwhm,fnum,int,e_mag,
&      int_si,period
double precision t_delta,t_start,t_final
double precision e_cpn(6),eff

C *****Parameters of ADK & Probability*****
integer n_charge
parameter (n_charge=18) !we go to Argon18+
double precision rate_adk(n_charge),sum_adk(n_charge),
&      runinteg_adk(n_charge),sum_adk_diff,sum_ion,
&      prob_ion(n_charge+1),prob_ion_pre(n_charge+1),
&      prob_e,
&      prob_e_i(n_charge),prob_ion_sum,prob_ion_sum_pre,
&      prob_ion_max
double precision ip(n_charge),nstar(n_charge),c2nl(n_charge),
&      flm(n_charge)
integer target_ion

C *****Parameters of trajectory*****
double precision t_integ_delta,t_integ_final
double precision gamma_ut
double precision traj_start(neq)

C *****Parameters of rescatter*****
integer n_rescatter_angle
parameter(n_rescatter_angle=180)
double precision x_pre,x_now,traj_exit,ini_spread,
&      rescatter_traj_start(neq),
&      rescatter_t_start
double precision rescatter_angle,rescatter_time,
&      rescatter_kin,rescatter_flux,rescatter_prob_e
logical checkRescatter,happenRescatter
double precision rescatter_tag

C *****Parameters for constants*****
double precision ener_au,int_au

C *****Counters*****
integer ii,aa,cc,pp,ee,tt,c_start,nn

```

```

logical calculate,outside

C *****Misc parameters*****
integer file1,file2,file3,file4,file5,file6,file7,file8
parameter (file1=11,file2=12,file3=13,file4=14,file5=15,
& file6=16,file7=17,file8=18)
double precision adk_thresh,prob_e_thresh
parameter(adk_thresh=1.0d-300,prob_e_thresh=1.0d-6)

C *****random number generate*****
integer*4 now(3)
double precision rseed,randr,randphi,randz

C *****file name flag*****
integer n_job
character*6 n_version
character*3 ch_n_job

C *****elapsed time*****
real etime
real elapsed(2)
real total

C *****Subroutines & functions*****
external adkrate,emfield,rand,derivs,
& setup,ut,
& filenamepara,
& subexitpoint,subspread,subrespread,subflux,
& getScatterTraj
intrinsic log10,dsqrt,sin,cos,atan,itime

C *****
C                               Set parameters
C *****

C target ion
target_ion = 8

C ELSEPA parameters
IELEC = -1                ! electron
IZ   = n_charge           ! no default
NELEC = n_charge-target_ion ! =Z (the present value is a flag)
MNUCL = 1                 ! Fermi nuclear charge distribution
MELEC = 1                 ! DF electron density
MUFFIN= 0                 ! free atom
RMUF  = 200.0D-8          ! free atom
MEXCH = 0                 ! FM exchange potential
MCPOL = 0                 ! no correlation-polarization
VPOLA = 0.0D0             ! atomic polarizability
VPOLB = 0.0D0             ! polariz. cutoff parameter

```

```

MABS = 0 ! no absorption
VABSA = 0.0D0 ! absorption potential strength
VABSD = 1.0D0 ! energy gap
IHEF = 0 ! high-energy factorization on

ECUT=MIN(20.0D3*IZ,2.0D6)

MCPOLC=0
VPOLBC=0.0D0
MABSC=0

c #####
c #####input values#####
n_job=1
n_version='rescv3'

c #####
c #####
c set constants
c=137.03545d0
pi=4.0d0*ATAN(1.0d0)
int_au=6.43640931d15
ener_au=27.2113962d0

c set laser pulse parameters
fwhm=1653.65d0 !40fs
wavelength=15118.015d0 !800nm
freq=(2.0d0*pi*c)/wavelength
period=wavelength/c
sigma=fwhm/(2.0d0*dsqrt(2.0d0*abs(log(2.0d0)))) !T0=2*sigma
fnum=2.0d0
s0=(2.0d0*fnum*wavelength)/pi
zr=(pi*s0**2.0d0)/wavelength

c set ADK rate parameters
c Sui's calcualted input <<< for Argon
c set adk parameters for Argon
ip(1)=0.57916593d0
ip(2)=1.0153496d0
ip(3)=1.4972388d0
ip(4)=2.1980577d0
ip(5)=2.7568085d0
ip(6)=3.3445061d0
ip(7)=4.568654d0
ip(8)=5.2721391d0
ip(9)=15.524663d0
ip(10)=17.591318d0
ip(11)=19.806515d0
ip(12)=22.720627d0
ip(13)=25.213485d0
ip(14)=27.498765d0
ip(15)=31.411924d0

```

```

ip(16)=33.737958d0
ip(17)=151.43936d0
ip(18)=162.66046d0

nstar(1)=0.92914501d0
nstar(2)=1.4034832d0
nstar(3)=1.7336472d0
nstar(4)=1.9077675d0
nstar(5)=2.1293728d0
nstar(6)=2.3199053d0
nstar(7)=2.3157353d0
nstar(8)=2.4636645d0
nstar(9)=1.6151632d0
nstar(10)=1.6859156d0
nstar(11)=1.7477272d0
nstar(12)=1.7801479d0
nstar(13)=1.8306778d0
nstar(14)=1.887802d0
nstar(15)=1.8924699d0
nstar(16)=1.9478055d0
nstar(17)=0.97682d0
nstar(18)=0.99796775d0

c2nl(1)=4.1156451d0
c2nl(2)=2.9568218d0
c2nl(3)=1.9898244d0
c2nl(4)=1.543155d0
c2nl(5)=1.0725701d0
c2nl(6)=0.75898481d0
c2nl(7)=0.76498791d0
c2nl(8)=0.57379913d0
c2nl(9)=2.3251668d0
c2nl(10)=2.1223649d0
c2nl(11)=1.9514752d0
c2nl(12)=1.8645628d0
c2nl(13)=1.7332032d0
c2nl(14)=1.5911276d0
c2nl(15)=1.5798321d0
c2nl(16)=1.4496643d0
c2nl(17)=4.0410855d0
c2nl(18)=4.0037221d0

flm(1)=3.0d0
flm(2)=3.0d0
flm(3)=3.0d0
flm(4)=3.0d0
flm(5)=3.0d0
flm(6)=3.0d0
flm(7)=1.0d0
flm(8)=1.0d0
flm(9)=3.0d0
flm(10)=3.0d0

```

```

flm(11)=3.0d0
flm(12)=3.0d0
flm(13)=3.0d0
flm(14)=3.0d0
flm(15)=1.0d0
flm(16)=1.0d0
flm(17)=1.0d0
flm(18)=1.0d0

C
*****
C                                     Open files
C
*****
C      generate file names parameters
C      call filenamepara(n_job,ch_n_job)

      open(file1,file=n_version//'_data_'//ch_n_job//'.dat')
      write(file1,1325)

      open(file3,file=n_version//'_log_'//ch_n_job//'.txt')

      open(file4,file=n_version//'_popu_'//ch_n_job//'.dat')
      write(file4,1327)

      open(file5,file=n_version//'_res_'//ch_n_job//'.dat')
      write(file5,1329)

C
*****
C                                     Main program begins
C
*****

C      generate initial phase
C      iniphase=(1.0/100.0d0)*pi !random initial phase

C      set the intensity
C      int_si=5.2d16
C      int=int_si/int_au
C      e0=dsqrt((8.0d0*pi*int)/c)

C      write log
C      write(file3,*) '>>> ip(1) = ',ip(1)
C      write(file3,*) '>>> period = ',period
C      write(file3,*) '>>> freq = ',freq
C      write(file3,*) '>>> n_job = ',n_job
C      write(file3,*) '>>> int(si) = ',int_si
C      write(file3,*) '>>> int(au)= ',int
C      write(file3,*) '>>> e0(au) = ',e0
C      write(file3,*) '>>> fwhm(au) = ',fwhm
C      set time step

```

```

t_delta=(10.0d0*sigma)/dble(n_time)
t_integ_delta=period/100.0

c    write log
write(file3,*) '>>> n_time = ',n_time
write(file3,*) '>>> t_delta = ',t_delta
write(file3,*) '>>> t_integ_delta = period/100 =
',t_integ_delta
write(file3,*) '>>> 12sigma/period = ',12.0*sigma/period

c    current rescatter angle
aa = n_job
rescatter_angle=dble(aa)

c
*****
c    scan the space defined by the above subroutine.the
c    number of atoms in a certain disc is proportional to its
c    volume.for each atom in a given disc,ADK rate and ionization
c    probability are calculated to determine whether a "significant"
c    ionization is triggered.if triggered an electron is freed
c    (initially zero velocity)
c    and its trajectory is recorded by integrating the Newton's
c    dynamic equation with rksuit_mod.f.finally the energy,momentum
c    and angular spectrum for the atom ensemble can be obtained.

c    initialize lauch trajectory parameters
calculate=.false.

c    initialize the variables regarding adk rate and ion prob
do cc=1,n_charge
    sum_adk(cc)=0.0d0
    runinteg_adk(cc)=0.0d0
end do
prob_ion(1)=1.0d0
prob_ion_pre(1)=1.0d0
do cc=2,n_charge+1
    prob_ion(cc)=0.0d0
    prob_ion_pre(cc)=0.0d0
end do
sum_ion=1.0
prob_e=0.0d0

do nn=1,neq
    traj_start(nn)=0.0d0
end do

c    scan of pulse starts
do 22 pp=1,n_time

c    declare time domain variable
t_start=-5.0d0*sigma+dble(pp)*t_delta

```

```

t_final= 5.0d0*sigma
t_integ_final=t_final+0.1d0*sigma

c  calculate adk rate and ionization probability to determine
c  whether to free electron. the following derivation follows
c  page 25-27 in David Neal Fittinghoff's Ph.D. Thesis Dec 1993

c  calculate adk rate
      do cc=1,n_charge
        c_start=cc
        call adkrate(c_start,ip(c_start),nstar(c_start),
&                c2nl(c_start),flm(c_start),traj_start,
&                t_start,rate_adk(c_start),e_mag)
      end do

c  calculate running integral
      do cc=1,n_charge
        sum_adk(cc)=sum_adk(cc)+rate_adk(cc)*t_delta
        if(sum_adk(cc).lt.adk_thresh) then
          sum_adk(cc)=0.0d0
        end if
      end do

c  calculate the population of each ion state
      prob_ion(1)=dexp(-sum_adk(1))
      do cc=2,n_charge
        if(sum_adk(cc).gt.700) then
          prob_ion(cc)=0.0d0
        else
          sum_adk_diff=sum_adk(cc)-sum_adk(cc-1)
          runinteg_adk(cc)=runinteg_adk(cc)+
&                (dexp(sum_adk_diff)*rate_adk(cc-1)*
&                t_delta)
          prob_ion(cc)=dexp(-sum_adk(cc))*
&                runinteg_adk(cc)
        end if
      end do
      prob_ion(n_charge+1)=prob_ion(n_charge+1)+
&                (rate_adk(n_charge)*prob_ion(n_charge)*t_delta)

c  record the total ion population
      sum_ion=0.0d0
      do cc=1,n_charge+1
        sum_ion=sum_ion+prob_ion(cc)
      end do

c  record the photonelectron probability(new method)
      do ee=1,n_charge
        prob_e_i(ee)=0.0d0
        prob_ion_sum=0.0d0
        prob_ion_sum_pre=0.0d0

```

```

        do cc=ee+1,n_charge+1
            prob_ion_sum=prob_ion_sum+prob_ion(cc)
            prob_ion_sum_pre=
&            prob_ion_sum_pre+prob_ion_pre(cc)
            if(prob_ion_sum.gt.prob_ion_sum_pre) then
                prob_e_i(ee)=
&                prob_ion_sum-prob_ion_sum_pre
            end if
        end do
    end do

    prob_e=0.0d0
c    do ee=1,n_charge
c    prob_e=prob_e+prob_e_i(target_ion)
c    end do

    write(file4,1328) t_start,prob_ion(target_ion+1),prob_e

c    update the population of each ion state
    do cc=1,n_charge+1
        prob_ion_pre(cc)=prob_ion(cc)
    end do

c    determine whether to free electron
    if(prob_e.gt.prob_e_thresh) then
        calculate=.true.
    end if

c    electron trajectory integration starts
c    do while(1.eq.0)
    do while(calculate.EQV..true.)

c    initialize the position and momentum of atom
        do nn=1,neq
            traj_start(nn)=0.0d0
        end do

c    calculate the exit point
        traj_exit=subexitpoint(
&            traj_start(4),
&            traj_start(5),
&            traj_start(6),
&            t_start,
&            ip(target_ion),
&            sqrt(2.0*ip(target_ion)))
        traj_start(4)=traj_start(4)+
&            traj_exit

c    calculate the initial spread
        ini_spread=subspread(
&            traj_start(4),
&            traj_start(5),

```

```

&          traj_start(6),
&          t_start,
&          ip(target_ion))

c      reset rescatter paramters
      checkRescatter=.true.
      happenRescatter=.false.

      rescatter_time=0.0d0
      rescatter_flux=0.0d0
      rescatter_kin=0.0d0

      rescatter_t_start=0.0d0
      rescatter_prob_e=0.0d0

      rescatter_tag=0.0d0

      do nn=1,neq
        rescatter_traj_start(nn)=0.0d0
      end do

c      continue integrate after reverting momentum
430      continue

c      set rksuit_mod.f parameters
      tol=1.0d-6
      do tt=1,neq
        thres(tt)=1.0d-12
      end do
      mesage=.true.
      errass=.false.
      hstart=0.0d0

c      call setup for ut rksuite.f
      call setup(neq,t_start,traj_start,t_integ_final,
&          tol,thres,method,'Usual Task',errass,hstart,
&          work,lenwrk,mesage)

c      initialize counter
      uflag3=0

c      initialize integration time
      twant=t_start

c      initialize pre x position
      x_pre=traj_start(4)

c      integrate the trajectory with ut rksuite.f
      do while(twant.le.t_final)

        twant=twant+t_integ_delta

```

```

        call ut(derivs,twant,tnow,traj,trajp,trajmax,
&             work,uflag)

        if(uflag.gt.3) go to 301

c      record pre x position
        x_now=traj(4)

c      check if rescatter happens
        if(checkRescatter.eqv..true.) then
            if((x_now*x_pre).lt.0.0d0) then

c      limit to within one period
                if(rescatter_time.le.period) then

c      calculate the rescatter time elapsed
                    rescatter_time=tnow-t_start

c      calculate the returning kinetic energy
                    rescatter_kin=dsqrt(
&                        traj(1)**2.0d0+
&                        traj(2)**2.0d0+
&                        traj(3)**2.0d0+
&                        c**2.0d0)*c-c**2.0d0
c      rescatter_kin=0.5*(
c      &                        traj(1)**2.0d0+
c      &                        traj(2)**2.0d0+
c      &                        traj(3)**2.0d0)

c      exclude <10ev
                                if(rescatter_kin*ener_au.gt.10.5) then

c      calculate the flux
                                    rescatter_flux=subflux(
&                                        traj(5),
&                                        traj(6),
&                                        rescatter_time,
&                                        ini_spread)

c      calculate the cross section
                                        EV = rescatter_kin*ener_au
                                        OPEN(8,FILE='RESULT_TEMP.dat')
                                        CALL ELSEPA(IELEC,EV,IZ,NELEC,MNUCL,
1                                          MELEC,MUFFIN,RMUF,
1                                          MEXCH,MCPOLC,VPOLA,
1                                          VPOLBC,MABSC,VABSA,VABSD,IHEF,8)
                                        CLOSE(8)
                                        DO III=1,NTAB
                                            IF(ABS(RESCATTER_ANGLE-TH(III))
&                                                .LE.1.0E-4) THEN
                                                CROSAMP=DCST(III)/(A0B2)
                                            END IF

```

```

                                END DO

c      update the prob weighted with flux and crossSection
                                rescatter_prob_e=prob_e*
                                &      rescatter_flux*(
                                &      CROSAMP*
                                &      2*pi*sin(rescatter_angle/180.0*pi)*
                                &      (1.0d0/180.0d0*pi))*
                                &      180.0d0 !normalize

                                write(file5,1330) t_start,
                                &      rescatter_time,
                                &      ini_spread,
                                &      subrespread(traj(5),traj(6),
                                &      rescatter_time,ini_spread),
                                &      traj_exit,
                                &      rescatter_flux,
                                &      rescatter_kin,
                                &      crosamp,
                                &      rescatter_prob_e
c      GO TO 931
c      the probability which not elastic scatter
                                prob_e=prob_e

c      momentum after elastic scattering
                                call getScatterTraj(traj,
                                &      rescatter_angle,
                                &      rescatter_traj_start)

c      reset the rksuite start time
                                rescatter_t_start=tnow

c      reset the rescatter flag
                                checkRescatter=.false.
                                happenRescatter=.true.

                                end if
                                end if

                                end if
                                end if

c      record post x position
                                x_pre=x_now

c      calculate relativstic kinetic energy in a.u.
                                gamma_ut=dsqrt(traj(1)**2.0d0+
                                &      traj(2)**2.0d0+
                                &      traj(3)**2.0d0+
                                &      c**2.0d0)*c-c**2.0d0
c      gamma_ut=0.5*(
c      &      traj(1)**2.0d0+

```

```

c      &          traj(2)**2.0d0+
c      &          traj(3)**2.0d0)

          end do

c      post-check*****

c      check the exit status
          write(file1,1326) dble(n_job),
&          rescatter_angle,
&          rescatter_tag,
&          gamma_ut,
&          prob_e

301          continue

c      if elastic scatter happened
          if (happenRescatter.eqv..true.) then

c      reset initial conditions
          t_start=rescatter_t_start
          do nn=1,neq
              traj_start(nn)=rescatter_traj_start(nn)
          end do
          prob_e=rescatter_prob_e

c      reset flag
          happenRescatter=.false.

c      reset tag
          rescatter_tag=1.0d0

          goto 430

          end if
c 931      CONTINUE
c      set flag - finish trajectory integration
          calculate=.false.

c      end of one trajectory integration
          end do

c      end of pulse scan
22      continue

c
*****
c      Display elapsed time
c
*****
          total=etime(elapsed)
          write(file3,1399) '>>> Program Ends: ',

```

```

&      '      Total Elapsed Time =',total,
&      '      User Time =',elapsed(1),
&      '      System Time =',elapsed(2)

C
*****
C                                     Output files
C
*****
      close(file1)
      close(file3)
      close(file4)
      close(file5)

C
*****
C                                     Format statements
C
*****
1325 format(' ', 'n_job',3x, 'res_angle',3x, 'res_tag',3x,
&          'finalKin',3x, 'prob')
1326 format(' ',5(E16.8,3x))
1327 format(' ', 't_start',3x, 'ion',3x, 'prob_e')
1328 format(' ',3(E16.8,3x))
1329 format(' ', 't_start',3x, 'res_time',3x, 'ini_spread',3x,
&          'res_spread',3x,
&          'traj_exit',3x, 'res_flux',3x, 'res_kin',3x,
&          'crosamp',3x, 'res_prob_e')
1330 format(' ',9(E16.8,3x))
1399 format(' ',A,/,A,F9.3,/,A,F9.3,/,A,F9.3)

C
*****

C      program ends

      end

C      end of the main program
C
*****

C
////////////////////////////////////
C
////////////////////////////////////Subroutines////////////////////////////////////
C
////////////////////////////////////

```

```

C
////////////////////////////////////
C      This subroutine calculates the ADK tunneling rate
C
////////////////////////////////////

      subroutine adkrate(zzz,ip,nstar,c2n1,flm,traj_start,t_start,
&      rate_adk,e_mag)
      integer zzz
      double precision ip,e_mag,nstar,c2n1,flm,rate_adk
      double precision epsilon,factor,nmpower1,rate1_adk,
&      traj_start(*),t_start,e_cpn(6),eff
      double precision c,pi,freq,wavelength,e0,sigma,zr,s0
      common/params1/ c,pi,freq,wavelength,e0,sigma,zr,s0
      double precision iniphase
      common/params2/ iniphase

      call emfield(traj_start(4),traj_start(5),traj_start(6),t_start,
&      e_cpn,eff)
      e_mag=abs(dsqrt(e_cpn(1)**2+e_cpn(2)**2+e_cpn(3)**2))

      if(e_mag.eq.0.0d0) then
         rate_adk=0.0d0
      else
         epsilon=(2.0d0*ip)**1.5
         factor=epsilon/abs(e_mag)
         nmpower1=2.0d0*nstar-1.0d0 !assume m=0 for all charge state

         rate_adk=c2n1*dsqrt(3.0d0/(pi*factor))*ip*
&         flm*((2.0d0*factor)**nmpower1)*
&         exp(-(2.0d0*factor)/3.0d0)
      end if

      return
      end

C      end of subroutine adkrate
C
////////////////////////////////////

C
////////////////////////////////////
C      This subroutine calculates the efield in space
C
////////////////////////////////////

      subroutine emfield(x,y,z,t,e_cpn,eff)

      double precision x,y,z,t,e_cpn(6),e_cpn1(6),e_cpn2(6),

```

```

&      sz_t,eff,phase1,phase2,alpha,r2,eps
double precision c,pi,freq,wavelength,e0,sigma,zr,s0
common/params1/ c,pi,freq,wavelength,e0,sigma,zr,s0
double precision iniphase,wn
common/params2/ iniphase

c      in this project we use homogeneous field, so E=E(t)
c      calculate EM field amplitude
      wn=2.0*pi/wavelength
      eff=e0*sin(freq*t+iniphase-wn*z)*dexp(-((t-
z/c)/(2*sigma))**2.0d0)

c      calculate EM components
      e_cpn(1)=eff
      e_cpn(2)=0.0d0
      e_cpn(3)=0.0d0
      e_cpn(4)=0.0d0
c      e_cpn(5)=0.0d0
      e_cpn(5)=e_cpn(1)/c
      e_cpn(6)=0.0d0

      return
      end

c      end of subroutine emfield
c
////////////////////////////////////

c
////////////////////////////////////
c      This subroutine defines the first order derivative equation
used
c      by ut rk suite.f
c
////////////////////////////////////

      subroutine derivs(tgot,ygot,ypgot)

      double precision tgot,ygot(*),ypgot(*),e_cpn(6),gamma_c,
&      ex,ey,ez,bx,by,bz,eff
      double precision c,pi,freq,wavelength,e0,sigma,zr,s0
common/params1/ c,pi,freq,wavelength,e0,sigma,zr,s0
double precision iniphase
common/params2/ iniphase

      call emfield(ygot(4),ygot(5),ygot(6),tgot,e_cpn,eff)
      ex=e_cpn(1)
      ey=e_cpn(2)
      ez=e_cpn(3)
      bx=e_cpn(4)

```

```

by=e_cpn(5)
bz=e_cpn(6)
gamma_c=dsqrt((ygot(1))**2.0d0+(ygot(2))**2.0d0+
& (ygot(3))**2.0d0+c**2.0d0)
c mention:gamma_c is different from the gamma in the main code,
c gamma=gamma_c/c
ypgot(4)=ygot(1)*c/gamma_c
ypgot(5)=ygot(2)*c/gamma_c
ypgot(6)=ygot(3)*c/gamma_c
ypgot(1)=-ex-(ygot(2)*bz-ygot(3)*by)*c/gamma_c
ypgot(2)=-ey-(ygot(3)*bx-ygot(1)*bz)*c/gamma_c
ypgot(3)=-ez-(ygot(1)*by-ygot(2)*bx)*c/gamma_c

c classical
c ypgot(4)=ygot(1)
c ypgot(5)=ygot(2)
c ypgot(6)=ygot(3)
c ypgot(1)=-ex-(ygot(2)*bz-ygot(3)*by)
c ypgot(2)=-ey-(ygot(3)*bx-ygot(1)*bz)
c ypgot(3)=-ez-(ygot(1)*by-ygot(2)*bx)

return
end

c end of subroutine derivs
c
////////////////////////////////////

c
////////////////////////////////////
c This subroutine calculates the exit point
c
////////////////////////////////////

function subexitpoint(x,y,z,t,ip,charge)

double precision charge
double precision x,y,z,t,e_cpn(6),eff,ip,exitX
double precision deterTerm
double precision c,pi,freq,wavelength,e0,sigma,zr,s0
common/params1/ c,pi,freq,wavelength,e0,sigma,zr,s0
double precision iniphase
common/params2/ iniphase

c calculate the field
call emfield(x,y,z,t,e_cpn,eff)

c the electron ionized at opposite direction to the field
deterTerm=ip**2.0-4.0d0*abs(e_cpn(1))*charge

```

```

c      if the field happens to be zero
      if(e_cpn(1).eq.0.0d0) then
        exitX=0.0d0
      else
c      non-zero field if ATI regime
        if(deterTerm.le.0.0d0) then
c      determine field direction
          if(e_cpn(1).gt.0.0) then
            exitX=-ip/(2.0d0*abs(e_cpn(1)))
          else
            exitX= ip/(2.0d0*abs(e_cpn(1)))
          end if
c      non-zero field if tunnel regime
        else
c      determine field direction
          if(e_cpn(1).gt.0.0) then
            exitX=-(ip+sqrt(deterTerm))/(2.0d0*abs(e_cpn(1)))
          else
            exitX= (ip+sqrt(deterTerm))/(2.0d0*abs(e_cpn(1)))
          end if
        end if
      end if

      subexitpoint=exitX
c      subexitpoint=0.0d0
      return
      end

c      end of subroutine exitpoint
c
c      //////////////////////////////////////

c
c      //////////////////////////////////////
c      This subroutine calculates the initial spread
c
c      //////////////////////////////////////

      function subspread(x,y,z,t,ip)

      double precision x,y,z,t,e_cpn(6),eff,ip,spread
      double precision c,pi,freq,wavelength,e0,sigma,zr,s0
      common/params1/ c,pi,freq,wavelength,e0,sigma,zr,s0
      double precision iniphase
      common/params2/ iniphase

c      calculate the field
      call emfield(x,y,z,t,e_cpn,eff)

c      calculate the initial spread

```

```

        subspread=(2.0*ip)**0.25/dsqrt(2.0*abs(e_cpn(1)))

        return
    end

c      end of subroutine initialsread
c
c      ///////////////////////////////////////////////////

c
c      ///////////////////////////////////////////////////
c      This subroutine calculates the return spread
c
c      ///////////////////////////////////////////////////

        function subrespread(y,z,elapse,inispread)

        double precision y,z,r,elapse,inispread
        double precision c,pi,freq,wavelength,e0,sigma,zr,s0
        common/params1/ c,pi,freq,wavelength,e0,sigma,zr,s0
        double precision iniphase
        common/params2/ iniphase

c      calculate the final spread
        subrespread=inispread*dsqrt(1.0+
&      elapse**2.0/(4.0*inispread**4.0))

        return
    end

c      end of subroutine return spread
c
c      ///////////////////////////////////////////////////

c
c      ///////////////////////////////////////////////////
c      This subroutine calculates the flux
c
c      ///////////////////////////////////////////////////

        function subflux(y,z,elapse,inispread)

        double precision y,z,r,elapse,inispread,respread
        double precision c,pi,freq,wavelength,e0,sigma,zr,s0
        common/params1/ c,pi,freq,wavelength,e0,sigma,zr,s0
        double precision iniphase
        common/params2/ iniphase

```

```

c      calculate the final spread
      respread=inispread*dsqrt(1.0+
&      elapse**2.0/(4.0*inispread**4.0))

c      calculate the flux
      subflux=1/(2.0*pi*respread**2.0)*
&      dexp(-(y**2.0+z**2.0)/(2.0*respread**2.0))

      return
      end

c      end of subroutine flux
c
////////////////////////////////////

c
////////////////////////////////////
c      This function returns the elastic scattering cross section
c
////////////////////////////////////
      subroutine getScatterTraj(traj,angle,traj_start)

      double precision angle,traj(6),traj_start(6),momentum,
&      directionX,directionY,directionZ,compoX,compoY
      double precision c,pi,freq,wavelength,e0,sigma,zr,s0
      common/params1/ c,pi,freq,wavelength,e0,sigma,zr,s0
      double precision iniphase
      common/params2/ iniphase
      intrinsic sqrt,cos,sin,abs

c      calculate momentum magnitude
      momentum=sqrt(traj(1)**2.0d0+traj(2)**2.0d0+traj(3)**2.0d0)

c      calculate direction
      if (traj(1).ge.0.0d0) then
        directionX= 1.0d0
      else
        directionX=-1.0d0
      end if
      if (traj(2).ge.0.0d0) then
        directionY= 1.0d0
      else
        directionY=-1.0d0
      end if
      if (traj(3).ge.0.0d0) then
        directionZ= 1.0d0
      else
        directionZ=-1.0d0
      end if

```

```

c      calculate the scattered momentum
traj_start(1)=cos(angle/180.0d0*pi)*momentum*directionX
traj_start(2)=sin(angle/180.0d0*pi)*momentum*directionY
traj_start(3)=0.0d0
traj_start(4)=traj(4)
traj_start(5)=traj(5)
traj_start(6)=traj(6)

      return

      end

c      end of function getScatterTraj
c
////////////////////////////////////

c
////////////////////////////////////
c      This subroutine generates file name parameters
c
////////////////////////////////////

      subroutine filenamepara(n,ch_n)

      integer n
      character*1 ch1
      character*2 ch2
      character*3 ch_n

c      n_job
      if(n.lt.10) then
         write(ch1,'(I1)') n
         ch_n='00'//ch1
      else if((n.ge.10).and.(n.lt.100)) then
         write(ch2,'(I2)') n
         ch_n='0'//ch2
      else if((n.ge.100).and.(n.lt.1000)) then
         write(ch_n,'(I3)') n
      end if

      return
      end

c      end of subroutine filenamepara
c
////////////////////////////////////

```


Appendix C

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