Numerical simulation of photoionization of atoms and attosecond laser pulses

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<u>Abstract</u>

With appropriate filters, high harmonic generation (HHG) can produce X-ray pulses on the attosecond time scale. We formulate a procedure to minimize pulse duration in various energy regimes and predict photoionization rates for a number of noble gases. Bridging the gap between the theory of high harmonic generation and the experimental reality, we seek to support the production of tabletop X-ray sources able to probe electron dynamics in inner shells. This thesis builds on the work of other researchers in the Ultrafast AMO Theory group, including a novel analytic fitting to noble gas potentials, and predictions of the energy spectrum and phases obtained by high harmonic generation. Specifically, computational methods (including Crank-Nicolson) and the single active electron model allow for the solution of the timedependent Schrödinger equation and calculation of photoionization probabilities, while numerical Fourier transforms enable rapid analysis of filtered HHG spectra.

Keywords: attosecond, photoionization, high harmonic generation, strong-field physics, AMO physics, ultrafast

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Introduction

1 STRONG-FIELD PHYSICS

The field of strong-field ultrafast physics seeks to understand atomic and molecular behavior under the interaction with laser pulses on their intrinsic time scale. Strong-field lasers have field strengths comparable to the Coulomb interaction between the nucleus and electrons; ultrafast research studies electron dynamics on the attosecond $(10^{-18} s)$ time scale. While previously beyond reach, advances in recent decades have brought laser pulses with such short durations and high intensities into many labs of even moderate size [1]. Thus, a breadth of new opportunities has been opened for both theory and experiment to study new phenomena [2]. Under exposure to such novel laser sources, the process of atomic ionization acquires greater complexity in its own right, even as it becomes a useful tool in developing newer specialized lasers.

While the inert noble gases are ideal for many experiments, the difficulty of efficient theoretical calculations must be overcome. With well-known computational methods and a novel analytic approximation, we begin to characterize common behavior, such as photoionization for single- and many-electron systems. Opening with a general discussion, we will then introduce our methods before turning to the application of high harmonic generation (HHG).

2 PHOTOIONIZATION

It has long been known that exposure to laser light will ionize atoms, freeing the electrons. For weak lasers, perturbation theory can accurately describe many properties

of this process. However, in the strong-field regime this approach becomes invalid, requiring the use of other methods. In our research, the time-dependent Schrödinger equation (TDSE) is solved numerically, allowing for modeling photoionization under a wide selection of laser parameters. In particular, we can simulate interaction with realistic laser pulses (including frequency and phase distribution).

For the traditional single-photon process – where photon energy ($\hbar\omega$) is greater than the electron's binding energy – photoionization probability is simply proportional to total laser pulse energy. With high enough laser intensities, however, multi-photon ionization can occur, where *n* photons are absorbed and it is their total energy $n\hbar\omega$ that exceeds the binding energy. This work involves calculating ionization probabilities for both processes via the numerical solution of the TDSE. The method of imaginary time propagation allows us to construct the ground state for an electron given a potential curve – such as Hydrogen's simple Coulombic formula or more complex multi-electron potentials. Similarly, we can add any temporal form of a laser field, propagate the wavefunction in time, calculate the final state, and determine the ionization probability.

3 THE SINGLE ACTIVE ELECTRON MODEL

The potential curve for atoms with greater numbers of electrons than Hydrogen requires approximation due to the large number of dimensions needed to simulate each electron simultaneously. The single active electron (SAE) model is one such approximation for noble gases and certain other multi-electron atoms. Various methods, such as density functional theory, allow for the calculation of the ground state electron orbitals. Then, we assume that only one electron – usually the outermost – interacts with the external field while the others remain in their ground state configuration. We consider the other electrons as supplying a static potential, allowing for simpler, single-electron, calculations.

While the potential of noble gases previously required point-by-point definition, a graduate student in the Ultrafast AMO Theory group, Michelle Miller, has systematically fit analytic forms to the numerical density functional theory calculations for certain noble gases (He, Ne, Ar, and Kr). Among other applications, her work allows me to calculate ionization in much the same way as for Hydrogen. Comparison with literature data [3, 4] provides a test for the analytic forms of the potential. In turn, we prepare to model ionization from HHG pulses and provide a simple measure useful for later research.

4 HIGH HARMONIC GENERATION

High harmonic generation (HHG) is a process by which radiation with energy up to the X-ray regime can be generated from an infrared driving laser (Figure 1). Atoms are subjected to a powerful infrared laser, leading to absorption of multiple photons and subsequent ionization. When the laser field reverses direction, the free electron is directed back to the nucleus, gaining kinetic energy along the way. When the electron re-enters its original bound state, excess energy is released in the form of a photon (Figure 1) [5].

The Ultrafast AMO Theory group (particularly Dr. Carlos Hernández-García) has worked on calculations of the radiated energy spectrum from HHG and has determined that attosecond or shorter pulses can be generated through careful filtering and phasematching [6].

Recently, we have begun investigating the time domain laser pulses resulting from various Fourier transformed HHG spectra. The ideal case of constant phase across all harmonics leads to the shortest pulses, but we must also consider the more practical scenario where the phase varies across different energies. To this end, we have computed pulses using spectra and phases calculated by Dr. Hernández-García. Applying experimentally-feasible filters, we seek to refine the spectrum and reduce the resultant pulses to shorter durations. Sampling central energies across the HHG spectrum, we



vary the filter width and analyze the resulting pulse duration and shape. In pursuit of a general model, connections are made between the initial spectrum (namely its intensity profile as well as phase) and the duration of pulses drawn from different regions therein.

Once the shortest pulses have been identified, we aim to combine the different aspects of this research. Seeking to understand and characterize these novel pulses, we plan to simulate their interaction with various noble gases and calculate the resultant photoionization probabilities.

Methods

1 INTRODUCTION

We will first briefly review the general procedures for solving the one-electron Schrödinger equation beyond the limits of perturbation theory. A discussion of our general computational methods is followed by the application to our particular interest: photoionization in single- and multi-electron atoms. Turning to high harmonic generation, we provide a broader description of the process and the resultant spectrum. Beyond computational techniques for filtering the spectrum and converting it into the time domain, we include a discussion of our more general methods for extracting meaningful results. Finally, the topics are tied together by the study of photoionization of multi-electron atoms with HHG attosecond laser pulses.

2 STRONG-FIELD PHYSICS

The Time-dependent Schrödinger equation (TDSE) exactly describes the behavior of any quantum mechanical system in a state ψ :

$$\widehat{H}\psi(\vec{r},t) = i\hbar \frac{\partial \psi(\vec{r},t)}{\partial t}.$$
(1)

For Hydrogen, we fix the nucleus (a single proton) at the origin, define a spatial electron wavefunction in terms of \vec{r} and apply an electric field $\vec{E}(t)$. Thus, we have $\hat{H} =$ Kinetic Energy + Coulomb potential + E-field coupling:

$$\left[\frac{-\hbar^2}{2m_e}\nabla^2 - \frac{e^2}{4\pi\epsilon_0|\vec{r}|} + e\vec{E}(t) * \vec{r}\right]\psi(\vec{r},t) = i\hbar\frac{\partial\psi(\vec{r},t)}{\partial t}.$$
 (2)

To simplify the equation, we use Hartree atomic units, where

$$\hbar = m_e = q_e = 4\pi\varepsilon_0 = 1$$
 and $c = \frac{1}{\alpha} = 137.$ (3)

This is the natural unit system for Hydrogen, with one atomic unit (a.u.) equal to the Bohr radius a_B and twice the magnitude of the ground state energy R_y . In this thesis, we define the z-direction to be along the polarization of the electric field, leading to the simpler TDSE:

$$\left[\frac{-1}{2}\nabla^2 - \frac{1}{|\vec{r}|} + E(t)z\right]\psi(\vec{r},t) = i\frac{\partial\psi(\vec{r},t)}{\partial t}.$$
(4)

This equation cannot be solved exactly, but for weak laser fields (in comparison to the Coulomb interaction), perturbation theory can give an approximate analytical solution.

As can be seen in Figure 2, an electric field of . 1 *a*. *u*. magnitude (5.14 \times $10^{10} V/m$) dramatically alters the potential energy curve of a Hydrogen-bound electron. In this strong-field regime, perturbation theory does not apply and we must



Shown is the potential energy of the Coulombic field for an electron in Hydrogen (blue line), the ground state energy (red line) and the potential energy when a .1 a.u. electric field is applied (green line). Note that electrons with ground state energy are no longer bound with this .1 a.u. electric field. For weaker fields, the potential energy distortion can result in tunneling ionization.

3 COMPUTATIONAL QUANTUM MECHANICS AND THE CRANK-NICOLSON METHOD

Stepping away from analytic solutions entirely, we turn to computational techniques based on the Crank-Nicolson method. Using a code library written by former members of the Ultrafast AMO Theory group, we can solve the TDSE for many systems. Wavefunctions are defined at discrete spatial and temporal points on a spatial grid of finite size.

To propagate the wavefunction through a small time step Δt , we take the approximate solution:

$$\psi(t + \Delta t) \simeq e^{-i\hat{H}(t)\Delta t}\psi(t) \simeq \frac{1 - i\frac{\Delta t}{2}\hat{H}(t)}{1 + i\frac{\Delta t}{2}\hat{H}(t)}\psi(t),$$
(5)

write the Hamiltonian in a particular coordinate system (e.g., Cartesian as shown here):

$$\widehat{H}(t) = \frac{-1}{2} \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} \right) - \frac{1}{\sqrt{x^2 + y^2 + z^2}} + z E(t), \tag{6}$$

and approximate the derivatives as finite differences:

$$\frac{\partial f(x,t)}{\partial t} \approx \frac{f(x,t+\Delta t) - f(x,t)}{\Delta t} \quad \text{and} \quad (7)$$
$$\frac{\partial^2 f(x)}{\partial x^2} \approx \frac{\frac{f(x+\Delta x) - f(x)}{\Delta x} - \frac{f(x) - f(x-\Delta x)}{\Delta x}}{\Delta x} = \frac{f(x+\Delta x) - 2f(x) + f(x-\Delta x)}{\Delta x^2}. \quad (8)$$

Thus, the Hamiltonian \widehat{H} becomes an operator depending only on the values of ψ at points on the grid, leading to a system of linear equations:

$$\begin{pmatrix} \psi(x_0, y_0, z_0, t_1) \\ \psi(x_1, y_0, z_0, t_1) \\ \dots \\ \psi(x_N, y_N, z_N, t_1) \end{pmatrix} = A \begin{pmatrix} \psi(x_0, y_0, z_0, t_0) \\ \psi(x_1, y_0, z_0, t_0) \\ \dots \\ \psi(x_N, y_N, z_N, t_0) \end{pmatrix},$$
(9)

where A is a matrix and a function of the Hamiltonian. This system can be solved computationally through a variety of methods included in the code library.

Given any initial electronic state, we can determine its behavior for all times. Note that extending this procedure to other single-electron systems only requires exchanging Hydrogen's $\frac{1}{\sqrt{x^2+y^2+z^2}}$ potential term with the target's potential (such as $\frac{Z}{\sqrt{x^2+y^2+z^2}}$ for Hydrogenic ions).

4 GENERATING A GROUND STATE THROUGH IMAGINARY TIME PROPAGATION

Determining the ground state can be accomplished through a simple procedure, called imaginary time propagation (ITP). First, we make a guess for the spatial wavefunction, usually consisting of a Gaussian distribution:

$$|\psi\rangle = c e^{-\frac{|r|^2}{2\sigma}}.$$
(10)

This state is a superposition of many energy eigenstates:

$$|\psi\rangle = \sum_{E} a_{E}|E\rangle. \tag{11}$$

Left alone in the electric field-free Hamiltonian, the states will oscillate in time:

$$|\psi\rangle(t) = \sum_{E} a_{E} e^{-iEt} |E\rangle.$$
(12)

Next, we propagate backward in imaginary time:

$$|\psi\rangle(-i\Delta t) = \sum_{E} a_{E} e^{-E\Delta t} |E\rangle.$$
(13)

Recall that E < 0 for all bound states:

$$|\psi\rangle(-i\Delta t) = \sum_{E} a_{E} e^{|E|\Delta t} |E\rangle, \qquad (14)$$

and the ground state has the greatest binding energy:

$$|E_0| > |E_{excited}|. \tag{15}$$

So, the ground state term will have the fastest-growing exponential and

$$\sum_{E} a_{E} e^{|E|\Delta t} |E\rangle \approx a_{E_{0}} e^{|E_{0}|\Delta t} |E_{0}\rangle, \quad \text{for large } \Delta t.$$
(16)

Finally, we normalize the wavefunction (a simple numerical task). Consider the projection onto a given eigenstate:

$$\frac{|\langle \psi|E\rangle|^{2}}{|\langle \psi|\psi\rangle|} = \frac{a_{E}^{2}e^{2|E|\Delta t}}{\sum_{E}a_{E}^{2}e^{2|E|\Delta t}} \approx \frac{a_{E}^{2}e^{2|E|\Delta t}}{a_{E_{0}}^{2}e^{2|E_{0}|\Delta t}}$$
$$= \frac{a_{E}^{2}}{a_{E_{0}}^{2}}\frac{e^{2|E|\Delta t}}{e^{2|E_{0}|\Delta t}} = \frac{a_{E}^{2}}{a_{E_{0}}^{2}}e^{2(|E|-|E_{0}|)\Delta t}$$
$$= \frac{a_{E}^{2}}{a_{E_{0}}^{2}}e^{-2|E_{0}-E|\Delta t}.$$
(17)

The ground state projection is simply:

$$\frac{|\langle\psi|E_0\rangle|^2}{|\langle\psi|\psi\rangle|} \approx \frac{a_{E_0}^2}{a_{E_0}^2} e^{-2|E_0 - E_0|\Delta t} = 1,$$
(18)

while the excited states are suppressed:

$$\frac{|\langle \psi|E\rangle|^2}{|\langle \psi|\psi\rangle|} \approx \frac{a_E^2}{a_{E_0}^2} e^{-2|E_0 - E|\Delta t} \to 0 \qquad \text{as } \Delta t \to \infty.$$
(19)

Thus, after imaginary time propagation and normalization, we are left with only the ground state wavefunction. In practice, it is necessary to take many small time steps and normalize after each one to ensure accuracy, but the end result is the same. Note also that while $a_{E_0} = 0$ (i.e., the ground state is not at all contained in the initial guess) would lead the ITP method to fail, this would require an astoundingly unlucky guess and does not, in practice, occur. ITP can also calculate excited states, though with less accuracy than for the ground state. This requires an iterative method: to determine the first excited state, we need the ground state; to find the second, we need the ground and first excited states; and so on. When calculating an excited state, we subtract off the projection onto all states with greater binding energy at each step. So, the target state has the fastest exponential growth and thus dominates.

5 PHOTOIONIZATION

In the simplest photoionization scenario, an electron bound to a single proton (i.e., H) absorbs one photon with sufficient energy to ionize (for H: 13.6 eV). Treating the photon explicitly as a particle, if we assume the probability that one photon is absorbed is independent of the number of photons (but may depend on, e.g., wavelength), then we may use a simple law of probability: the probability of an event occurring **one or more** times within N trials is the probability of each individual event times the number of trials minus the probability of **more than one** occurrence:

$$P_{1+events} = NP_{event} - P_{2+events}.$$
 (20)

For a single-photon process, a single absorption event is sufficient; absorbing additional photons merely results in greater electron kinetic energy. If the total probability of ionization is low enough that we can ignore multiple absorption double-counting, then:

$$P_{ionization} \approx NP_{absorption}(\omega),$$
 (21)

with N equal to the number of photons which may be absorbed (i.e. the number in the pulse). Now, the number of photons is itself proportional to the total energy of the

pulse. For a given pulse envelope, the energy scales with intensity. We therefore expect ionization probability will be directly proportional to intensity:

$$P_{ion} \approx NP_{abs}(\omega) = \frac{E_{pulse}}{\hbar\omega} P_{abs}(\omega) = \frac{k_{shapel}}{\hbar\omega} P_{abs}(\omega) \propto I.$$
 (22)

We may also treat light as a wave, and consider the interaction between an atom and an electromagnetic field. For electric field strengths small in comparison to the Coulombic field (for Hydrogen: $5.14 \times 10^{11} V/m$ or 1 a. u.), time-dependent perturbation theory applies. I will not repeat the derivation, as it can be found in most introductory quantum mechanics texts. The result, however, is simple:

$$P_{ion} = k(\omega) \left| \vec{E}_{max} \right|^2 = k(\omega) * (2c\mu_0) I \propto I.$$
(23)

In either treatment, we expect the probability of single photon ionization will be proportional to laser intensity. In both cases, we assume that the intensity is "small" so that: there are few enough photons to ignore multiple absorption; and, perturbation theory is valid.

Now, let us turn to wavelengths too long (corresponding to photon energies too low) for single photon ionization. In this case, *n*-photon absorption occurs with $n\hbar\omega > E_{binding}$. Since electrons are restricted to discrete energy levels, multi-photon ionization cannot normally occur by steps – the electron must absorb all *n* photons in a "very short" period. There remains some debate as to whether the photons must be absorbed *simultaneously*, but for this thesis the distinction is not important. Higher-order perturbation theory dictates that $P_{ion} \propto I^n$, but this requires a weak field. In practice, multi-photon absorption is usually observed at $I \gtrsim 10^{13} W/cm^2$, corresponding to field strengths greater than 1% of the proton-electron attraction. Thus, perturbation theory is not valid for multiphoton ionization. I am aware of no derivation, but studies (both computational and experimental) on particular multiphoton ionization processes have consistently observed that:

$$P_{n-\nu \ ion} \propto I^k, for \ some \ k \in \mathbb{R}.$$
(24)

The relationship (if any) between k and n has not been discovered, though it is clear that k is **not** proportional to n.

In this thesis, we calculate photoionization by computational solution of the TDSE. The electric field of a laser is treated as a wave through the $\vec{E}(t) * \vec{z}$ term in the Hamiltonian. This has the advantage of treating single- and multi- photon processes comparably, without the need for field strength approximations or prior knowledge of which process should occur. Given that we can calculate a variety of initial states with ITP and propagate them in time, determining the electron-laser interaction becomes relatively straightforward. The primary issue is the finite nature of the spatial grid. In photoionization, electrons often leave the neighborhood of the source atom at high speeds. If we are not cautious, these ionized electrons will reflect in un-physical ways when they reach the boundary of our grid.

Two popular methods for resolving this boundary issue are described in [7] and briefly summarized here. The simpler solution, the masking function method, "multiplies the wavefunction by a cosine-based function that smoothly decays from 1 to 0 in the absorbing region" [7] at the end of each propagation step. This procedure absorbs most of the ionized electron wavefunction and suffices when reflection from grid boundaries is not of high concern. The second method, Exterior Complex Scaling (ECS), must be inserted directly into the propagation calculation. ECS adds a complex term to the limits of the axes, resulting in a more robust absorption [8]. The group library includes codes for both methods, so we chose to use ECS for all calculations.

Now, we can calculate the cumulative ionization probability as the initial norm minus the final (i.e. the percentage of the wavefunction that has left the grid):

$$P(ionization) = |\langle \psi | \psi \rangle|_{off} = |\langle \psi | \psi \rangle|_{init} - |\langle \psi | \psi \rangle|_{on}.$$
 (25)

With a normalized initial state, $|\langle \psi | \psi \rangle|_{init} = 1$, and we only need to calculate the ongrid norm:

$$P(ionization) = 1 - |\langle \psi | \psi \rangle|.$$
(26)

6 THE SINGLE ACTIVE ELECTRON MODEL

Thus far, our methods have applied generally to any single-electron system with a known potential. Unfortunately, this limits us to H, H_2^+ , He^+ , Li^{2+} , etc., which are unsuited to experimentation (due to chemical reactivity, production issues, etc.). Unreactive noble gases (He, Ne, Ar, Kr, Xe) are preferable, but suffer theoretical challenges. The TDSE solution methods from above can be used with multiple electrons, but the Coulombic coupling between electrons dramatically increases the computation time required to fully model, e.g., even a two-electron system:

$$\widehat{H} = -\frac{1}{2}\nabla_1^2 - \frac{1}{2}\nabla_2^2 - \frac{Z}{|\vec{r}_1|} - \frac{Z}{|\vec{r}_2|} + \frac{1}{|\vec{r}_1 - \vec{r}_2|} + E(t)z_1 + E(t)z_2$$
(27)

The single active electron model offers an approximate solution. We assume that only one of the electrons interacts with the laser field, while all others remain in their initial states. Thus, our active electron experiences a time-independent Coulomb field with contributions from the static electrons as well as the nucleus. For our situation of interest – noble gases interacting with lasers on a femtosecond or shorter time scale – this is a good approximation for the photoionization process.

Now, the issue becomes determining the multi-electron potential. A variety of methods can be used to numerically calculate the effective electron potential [7], but these are both theoretically unilluminating and computationally difficult. For this reason, a number of researchers have developed analytical fits to the numerical potential.

Tong and Lin [10] propose:

$$V(r) = -\left(\frac{Z_c}{r} + a_1 \frac{e^{-a_2 r}}{r} + a_3 e^{-a_4 r} + a_5 \frac{e^{-a_6 r}}{r}\right).$$
 (28)

 Z_c is the total atomic charge with the active electron removed (+1 for neutral atoms), and $\frac{Z_c}{r}$ therefore fits the long-range potential. The exponential decay term ($a_3e^{-a_4r}$) and the Yukawa terms $(a_1 \frac{e^{-a_2 r}}{r} + a_5 \frac{e^{-a_6 r}}{r})$ are fitted to the short-range potential including the static electrons' contribution. This model can be fit to observed binding energies moderately well, but has little theoretical motivation. The reasoning behind the three short-range terms is particularly opaque.

The Ultrafast AMO Theory group (particularly Michelle Miller) [9] proposes a systematic, physically-motivated procedure for constructing fits of the form:

$$V(r) = -\left(\frac{Z_c}{r} + a_1 \frac{e^{-a_2 r}}{r} + a_3 e^{-a_4 r} + \cdots\right).$$
 (29)

As in Tong's model, a $\frac{Z_c}{r}$ term matches the long-range potential seen by our active electron. Exponential terms are non-divergent for small r ($a_3e^{-a_4(0)} = a_3$) and are used to model structure very close to the nucleus. Yukawa terms do diverge at small r, but have the advantage of matching the Coulombic $\frac{1}{r}$ behavior for mid-range distances (less than, e.g., a_2). The number of these short-range terms required depends on the principle quantum number, n, of the atom. Essentially, electronic orbitals add structure to V at some particular r_n and require either an exponential term (if it is closely bound to the nucleus and $r_n < a_B$) or a Yukawa term (if it is further away). One additional Yukawa term is required to match overall long-range behavior. In particular, the coefficients of all the Yukawa terms plus Z_c should sum to the total nuclear charge.

Beyond the appealing theoretical justification, this system has the advantage of enabling iterative fitting for larger atoms. Starting with the parameters for Helium, we add an appropriate term to match the effect of the n = 2 orbitals. The choice of $a_5 \frac{e^{-a_6 r}}{r}$ or $a_5 e^{-a_6 r}$ depends on the particular atom. We take this new fit plus a term for n = 3 as an initial guess for the next atom. For instance:

$$V_{Xe}^{6 \ Terms}$$

$$\sim V_{Kr}^{5} + V_{Xe,n=5}^{1}$$

$$\sim V_{Ar}^{4} + V_{Kr,4}^{1} + V_{Xe,5}^{1}$$

$$\sim V_{Ne}^{3} + V_{Ar,3}^{1} + V_{Kr,4}^{1} + V_{Xe,5}^{1}$$

$$\sim V_{He}^{2} + V_{Ne,2}^{1} + V_{Ar,3}^{1} + V_{Kr,4}^{1} + V_{Xe,5}^{1}$$
(30)

With this analytic expression for the potential, we can extend all of our previous methods to certain experimentally-viable, multi-electron atoms. The extensive literature data then allow us to perform a test of both this thesis' methods as well as the new potentials themselves.



Shown is the HHG spectrum as obtained from numerical simulations by Dr. Carlos Hernández-García. Driving Field: $\lambda = 800nm$, $I = 6.8 \times 10^{14} W/cm^2$. Target: 2mm thick He gas at 400 torr. Note intensity peaks found at integer multiples of driving photon energy (1.55 eV).

7 THE HIGH HARMONIC SPECTRUM

Using an infrared laser (e.g., 800nm, 1.55 *eV*) to drive high harmonic generation, we see a resulting non-linear spectrum, where the intensities of frequencies many times that of the source field are not exponentially suppressed. In fact, a plateau structure appears, with roughly constant intensities across a wide range of energies (Figure 3). Unfortunately, HHG imparts an intrinsic chirp (frequency-dependent phase) to the spectrum (Figure 4). As we will see, the plateau structure enables short laser pulses, but a broadening effect of the chirp results in longer durations.

The work to calculate this spectrum was completed by Dr. Carlos Hernández-García, and the methods are fully described in his thesis [5], so I only briefly outline them here. He considers a region ($\sim mm$ thickness) of Helium gas at several realistic pressures. Characterizing the HHG process involves simulating four steps: 1) multi-



photon ionization, 2) acceleration, 3) recombination and emission, 4) propagation of photons. Steps 1)-3) correspond to the later three stages shown in Figure 1, while Step 4) involves the photons emitted by different atoms propagating through the gas to the edge of the region. The resultant laser spectrum is marked by intensity peaks at odd integer multiples of the source field's energy (called harmonics).

The core theory used for this computation is the strong field approximation (SFA). First, the electron's full Hilbert space is split into two subspaces: Bound and Continuum. The basic SFA assumes that no atomic excitation occurs, so the only available bound state is the ground state (1). (To be precise, Dr. Carlos Hernández-García uses "SFA+" which loosens this assumption.) While the electron is in the continuum (2), the effect of the atom's Coulomb field is ignored. Finally, after ionization, the electron is assumed to only recombine when it passes through the location of the nucleus (3). This method results in rapid calculation of the spectrum from a single atom. Combining the electromagnetic waves generated by many atoms and propagating through the source medium (4), the final complex-valued spectrum can be determined (Figure 3, Figure 4). SFA (and SFA+) are known to poorly model the lowest harmonics, so I use spectra where approximately the first 20 (800nm) or 40 (2µm) harmonics have been removed.

8 FOURIER TRANSFORMS

I) **BASIC FEATURES**

Fourier transformation (FT) converts a signal h(t) from the time-domain into the frequency (or energy) domain:

$$H(\omega) = \mathcal{F}[h(t)] \equiv \int_{-\infty}^{\infty} h(t)e^{-i\omega t}dt.$$
(31)

Similarly, the inverse Fourier transform converts a frequency-domain signal into the time domain:

$$h(t) = \mathcal{F}^{-1}[H(\omega)] \equiv \frac{1}{2\pi} \int_{-\infty}^{\infty} H(\omega) e^{i\omega t} d\omega.$$
(32)

Unsurprisingly, \mathcal{F}^{-1} is the inverse of \mathcal{F} , with $\mathcal{F}\mathcal{F}^{-1}$ and $\mathcal{F}^{-1}\mathcal{F}$ returning the original signal. Also, a signal's energy content remains constant under the transform:

$$\int_{-\infty}^{\infty} |h(t)|^2 dt = \int_{-\infty}^{\infty} |H(\omega)|^2 d\omega.$$
(33)

For the simplest case of a constant signal $h(t) = \frac{1}{\sqrt{2\pi}}$, the transform returns a delta

function, and vice-versa:



$$|d\omega$$
. (33)

(34)

The constant and delta functions can be seen as the limits of the Gaussian spectrum, with $\sigma \rightarrow \infty$ and $\sigma \rightarrow 0$, respectively. This pattern continues, with the transform of all Gaussian pulses being of Gaussian form (Figure 5). In fact, the relation between the widths is quite simple:

$$\sigma_{\omega-domain} = 1/\sigma_{t-domain},\tag{35}$$

$$\mathcal{F}\left[e^{-\frac{t^2}{2\sigma_t^2}}\right] = e^{-\frac{\omega^2 \sigma_t^2}{2}} \qquad \qquad \mathcal{F}^{-1}\left[e^{-\frac{\omega^2}{2\sigma_\omega^2}}\right] = e^{-\frac{t^2 \sigma_\omega^2}{2}}.$$
 (36)

Similarly for general spectra, the characteristic time t_0 is inversely proportional to the characteristic frequency ω_0 . So, there exists a product $t_0\omega_0$ that depends only on the shape of the spectrum and not the energy width (e.g., for Gaussian: $\sigma_\omega\sigma_t = 1$).

II) SPECTRA WITH VARYING PHASE

When $H(\omega)$ is complex-valued, the Fourier transform becomes more complicated. A constant phase carries through:

$$\mathcal{F}^{-1}\left[e^{i\phi}H(\omega)\right] \equiv \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{i\phi}H(\omega)e^{i\omega t}d\omega$$
$$= \frac{1}{2\pi} e^{i\phi} \int_{-\infty}^{\infty} H(\omega)e^{i\omega t}d\omega = e^{i\phi}h(t).$$
(37)

A phase varying linearly with frequency leads to a time-shift:

$$\mathcal{F}^{-1}\left[e^{i\phi\omega}H(\omega)\right] \equiv \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{i\phi\omega}H(\omega)e^{i\omega t}d\omega$$
$$= \frac{1}{2\pi} \int_{-\infty}^{\infty} H(\omega)e^{i\omega(t+\phi)}d\omega = h(t+\phi). \tag{38}$$

But, a non-linear phase has no straightforward effect on the time-domain pulse:



ex:
$$\mathcal{F}^{-1}[e^{i\phi\omega^2}H(\omega)] \equiv \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{i\phi\omega^2}H(\omega)e^{i\omega t}d\omega$$

$$= \frac{1}{2\pi} \int_{-\infty}^{\infty} H(\omega)e^{i\omega(t+\omega\phi)}d\omega =??.$$
(39)

However, we can note that the $(t + \omega \phi)$ argument suggests a frequency-dependent time-delay. This has the general effect of broadening the pulse in the time domain (Figure 6) so that $\sigma_t \sigma_\omega \neq 1$. For complex spectra, the pulse shape may also be affected.

III) DISCRETE FOURIER TRANSFORM

Our research considers a frequency spectrum defined at discrete points, and thus the continuous (inverse) Fourier transform described above is not quite appropriate. Instead, we use the (inverse) discrete fast Fourier transform (DFFT). For a spectrum $X(\omega_j)$, defined at N evenly-spaced frequencies (i.e., $\omega_j = jd\omega$ and j = 0,1,2, ... N - 1),

$$x(t_k) = \mathcal{F}^{-1}[X(\omega_j)] \equiv \sum_{n=0}^{N-1} X(\omega_n) e^{i\omega_n t_k},$$
(40)

where $t_k = kdt$ with the same number of points k = 0, 1, 2, ..., N - 1, and with spacing

$$dt = \frac{2\pi}{Nd\omega}.$$

The forward DFFT is defined likewise:

$$X(\omega_k) = \mathcal{F}[x(t_j)] \equiv \sum_{n=0}^{N-1} x(t_n) e^{-it_n \omega_k}.$$
(41)

Note that $\mathcal{FF}^{-1}[X(\omega_j)] = NX(\omega_j)$, though other definitions of the DFFT exist to preserve normalization. In this work, however, we only concern ourselves with relative intensities, and can thus ignore the factor of N.

The DFFT approximately retains all of the properties of the continuous FT, depending on the frequency sample. Small $d\omega$ is required to accurately reflect the frequency spectrum, but we need large $Nd\omega$ (thus small dt) to resolve pulses in the time domain. Our $d\omega$ is limited by the spectrum provided, but N may be increased – without altering the physical data – by padding the end of the spectrum with zeros. We found dt sufficiently small for our purposes and did not use this technique. The DFFT is, of course, calculated computationally [11].

9 FILTERING THE HHG SPECTRUM

I) GAUSSIAN FILTERS

To generate shorter pulses, we apply experimentally-feasible filters to the HHG spectra. In particular, we multiply the magnitudes by various Gaussian masks:

$$X_{filtered}(\omega_n) = X_0(\omega_n) * e^{\frac{1}{2}\left(\frac{\omega_n - \mu}{\sigma_f}\right)^2}.$$
(42)



This is equivalent to filtering the intensities by a filter of proportional width:

$$I_{filtered}(\omega_n) = \left| X_{filtered}(\omega_n) \right|^2 = \left| X_0(\omega_n) \right|^2 * \left(e^{\frac{1}{2} \left(\frac{\omega_n - \mu}{\sigma_f} \right)^2} \right)^2$$
$$I_{filtered}(\omega_n) = I_0(\omega_n) e^{\frac{21}{2} \left(\frac{\omega_n - \mu}{\sigma_f} \right)^2} = I_0(\omega_n) e^{\frac{1}{2} \left(\frac{\omega_n - \mu}{\sigma_f / \sqrt{2}} \right)^2}.$$
(43)

For the 800nm spectrum, we systematically range central energy μ across every harmonic between the intensity cut-offs (29.2 *eV*, 30.7 *eV*, ..., 147.4 *eV*) and vary the filter width σ_f from 1-25 harmonics (1.54, 3.07, ..., 38.38 *eV*), resulting in 1,950 separate filtered spectra (ex: Figure 7).

The 2µm spectrum is significantly broader, so we sample μ every 10 harmonics (25.1, 31.3, ..., 570.0 *eV*) and vary σ_f from 5-100 harmonics in steps of 5

(3.13, 6.26, ..., 62.64 *eV*), for a total of 1,760 filtered spectra. The precise combinations with 1-harmonic steps (88,000 in all) may be completed at some future point; however, it is unlikely new physical effects will be revealed.

II) UN-PHYSICAL MODIFICATIONS

In addition to realistic filters of the spectrum, two purely theoretical modifications help make sense of the data. Namely, we may set the intensity and/or phase to a constant value, before applying the same filters:

$$I_{const,filtered}(\omega_n) = 1 * e^{-\frac{1}{2} \left(\frac{\omega_n - \mu}{\sigma_f / \sqrt{2}}\right)^2} \qquad \phi_{const}(\omega_n) = 0.$$
(44)

This allows us to parse the effects of the spectrum's shape (intensity) and its chirp (phase) independently. Performing both modifications simultaneously serves as a test of the numerical procedure.

III) FOURIER TRANSFORM

Combining our chosen intensity and phase profile into a complex signal:

$$X(\omega_n) = \sqrt{I(\omega_n)} e^{i\phi(\omega_n)},$$
(45)

we compute the inverse discrete Fourier transform described above:

$$x(t_k) = \mathcal{F}^{-1}[X(\omega_i)] \equiv \sum_{n=0}^{N-1} X(\omega_n) e^{i\omega_n t_k}.$$
(46)

From this time-domain pulse, we calculate the full width at half maximum based on intensity:

$$I_{max} = \max_{k} |x(t_{k})|^{2}, I_{1/2} = \frac{1}{2} I_{max},$$

$$t_{max} \equiv \left\{ \max_{k}(t_{k}) : |x(t_{k})|^{2} \ge I_{\frac{1}{2}} \right\},$$
(47)

$$t_{min} \equiv \left\{ \min_{k} (t_k) : |x(t_k)|^2 \ge I_{\frac{1}{2}} \right\},\tag{48}$$

$$FWHM \equiv t_{max} - t_{min},\tag{49}$$

and we define the pulse duration as *FWHM*.

For a perfect Gaussian pulse, the FWHM duration is proportional to σ :

$$I_{max} = \max \left| e^{-\frac{x^2}{2\sigma^2}} \right|^2 = 1 \qquad I_{1/2} = \frac{1}{2}$$

$$x_{max} \equiv \left\{ \max(t) : \left| e^{-\frac{x^2}{2\sigma^2}} \right|^2 = \frac{1}{2} \right\},$$
So, $e^{-\frac{x_{max}^2}{\sigma^2}} = \frac{1}{2} \qquad \text{or} \qquad -\frac{x_{max}^2}{\sigma^2} = \ln\left(\frac{1}{2}\right)$

$$x_{max} = \sigma\sqrt{\ln 2} \qquad (\text{ and } x_{min} = -\sigma\sqrt{\ln 2})$$

$$FWHM = 2\sigma\sqrt{\ln 2} \approx 1.665\sigma. \qquad (50)$$

Now, we consider a continuous FT of this Gaussian, and we can find the width product $\sigma_{\omega}FWHM_t$:

$$\sigma_{\omega}\sigma_t = 1 \quad \rightarrow \quad \sigma_{\omega}FWHM_t \approx 1.665.$$
 (51)

This will later become useful for comparison with other cases.

Results

1 INTRODUCTION

First, we consider a simple test of our general DFFT procedure. Then, we discuss the results related to the High Harmonic spectrum generated from 800nm and 2 μ m driving fields. Turning to the study of photoionization, we again begin with a test – single-photon ionization of Hydrogen. This leads into the study of a strong-field process – multiphoton ionization. To conclude, we discuss ionization from noble gases.

2 FILTERING A TEST SPECTRUM

As a test of our methods, we first consider the test spectrum with constant intensity and phase:



$$I_{const,filtered}(\omega_n) = 1 * e^{-\frac{1}{2} \left(\frac{\omega_n - \mu}{\sigma_f / \sqrt{2}}\right)^2} \qquad \phi_{const}(\omega_n) = 0.$$
(44)

In Figure 8, we show the calculated FWHM duration for each combination of central energy μ and filter width σ_f . We first note that, as expected, duration decreases as filter width increases. Aside from some deviation at low central energies, the duration is independent of where we center our Gaussian. These deviations result when our Gaussian is cut off by the lower edge of our spectrum (I is not defined for $\omega_n < 0$).

Another effect of the discrete Fourier transform is the limited resolution. Recall that the width product $\sigma_f FWHM_t$ is 1.665 (eq. 45) for the continuous Fourier transform. But, for our DFFT, the duration remains constant across a range of σ_f . This leads to width products varying from 1.3–2.2 (Figure 9). We do not have the perfect analytic result, but come close enough for our descriptive purposes.

3 HHG FILTERING – 800NM SOURCE Assured that our methods are sound, we begin analysis of the true HHG spectrum. First, we consider the intensity spectrum alone by setting the phase to a constant zero. Next, we investigate just the phase using a constant-windowed intensity. Finally, we take the full, true



Figure 9: Width Product vs Filter Width Shown is the width product $\sigma_f FWHM_t$ as a function of the filter width. In contrast to a continuous Fourier transform (black line), the width product is not constant for a Discrete Fourier transform (blue, red points)

spectrum and phase. The interesting duration features can be resolved by a common color scale, ranging from $50 - 300 \ as$ (note this is different than for the test spectrum). For the 800nm spectrum, we have N = 4096 data points in the frequency and time domains, with resolution of $dE = .0808 \ eV$, and $dt = \frac{2\pi}{Nd\omega} \approx 12.50 \ as$, respectively.

I) CONSTANT PHASE

Flattening the phase allows us to probe the effects of the intensity distribution without worrying about effects due to phase. In Figure 10, we see that the limits of the spectrum ($\sim 29 - 164 \ eV$) become important, similar to the zero cut-off seen in the test spectrum. Unlike the test spectrum, the duration depends on where exactly we center the filter, even in the plateau region. This effect remains relatively small ($\pm \sim 25 \ as$), but



does serve to differentiate sections of the spectrum, especially at mid-range filter widths.

II) CONSTANT INTENSITY

The analysis of the HHG phase is both more complex and, as will be seen, more significant. Setting the intensity to a constant 1 within the limits of the original spectrum reveals an interesting map of the phase effects (Figure 11).

First, let us focus on central energies of $\sim 80 - 120 \ eV$. For tight filters (low σ_f), the pulses are too long to be of particular interest. As the filter broadens in frequency, we see the expected narrowing of our pulse in time. However, the shortest pulses appear for $\sigma_f \sim 12 \ eV$, with filters wider than this resulting in *longer* pulses. This



contrasts with the behavior for spectra of constant phase. As we take broader frequency samples, we also encounter a broader range of phase. This leads to narrowing and broadening, respectively, of our time pulse. It is the competition between these effects that leads to the behavior in Figure 11.

This behavior can also be understood through the width product $\sigma_f FWHM_t$. For a constant phase Gaussian, this product is roughly 1.67. When the phase varies with frequency, we diverge from this Fourier limit. Consider the filters centered at 100 eV (Figure 12): for filter widths above $\sim 10 \ eV$, $\sigma_f FWHM_t$ increases quadratically with σ_f in the case of frequency-dependent phase. We can think of the Fourier transform as becoming less efficient when phase variation is present. For constant phase, a constant factor relates width in the time and frequency domains. In this case, we may narrow our time pulse without limit. With a frequency-dependent phase, widening the frequency spectrum leads to a dramatically less efficient transform, setting a lower limit on the

breadth of our time domain pulse. Note that this is not a computational relic, but rather an essential feature of the mathematics (and similarly, the physics).

Considering the high central energy region ($\mu \gtrsim 120 \ eV$), we see similar behavior, though the intensity





cases. First, while the cut-off is essential to the actual physics, it somewhat obscures useful phase information for higher central energies. Thus, in Figure 14, we do not set the intensity to zero at the edges of the original spectrum. Note that in the low- μ regions where we do not have accurate phase information, this un-cut phase dramatically (and un-physically) alters the duration profile. However, in the higher central energy region of current interest, we see a smooth continuation of the midrange μ behavior. Namely, the duration first decreases as the filter broadens, then rises again as we reach higher filter widths. The only difference is the values of σ_f . For higher

energies, the band of short pulses is thinner and the long (> 300 *as*) pulses appear for much tighter filters.



For the next informative case, we flatten the phase of harmonics below $\sim 67 \ eV$ (Figure 13). This is the region where phase differences are greatest (Figure 4), and there is an interest in analyzing the effect of these rapid changes. Unsurprisingly, for low central energies, the effect is dramatic, returning us almost to the test case. Of greater import, however, are the effects on high central energy pulses. Comparing Figure 11 and Figure 13 for $\mu \gtrsim 90 \ eV$, we see almost no difference in pulse durations. The presence or lack of phase difference in the lower harmonics is irrelevant when the associated intensities are negligible. But, for wide filters centered around $100 \ eV$, we observe an interesting effect – the pulses have shorter durations when the low- ω phase is included. This counterintuitive effect likely results when the frequency-dependent delays arrange to cancel each other out, thus compressing the pulse.

Turning our attention to the lower central energies, we make similar observations. Comparison between the true phase (Figure 11) and that with the higher harmonics set to constant phase (Figure 15) leads to interesting insights. In both cases,

the large phase difference found in the low harmonics results in long pulses. However, this effect is alleviated when we



broaden the filter to include contributions of less varying phase. But, we see that **no** variance is not, in fact, the ideal case. Shorter pulses are found when the phase is allowed to vary somewhat at the edges of the filter. As our central energy approaches the region of relatively constant phase, the filter width required to constructively incorporate this region decreases (thus the angled band structure). Still, we see an eventual turnaround, where broadening the filter results in longer pulses – we include too wide a spectrum and the phase works against us. At these large filter widths, constant phase is preferable (Figure 15) – even if only in a portion of the target region.

III) TRUE SPECTRUM

Taking the full real spectrum, the durations (Figure 16) show a behavior surprisingly similar to that of the constant intensity case (Figure 11). While the μ , σ_f



dependence is generally smoother, the essential features remain. We have long pulses in three regions: {very low σ_f }, {low μ , low σ_f }, and {high μ , high σ_f }. Short pulses can be found at {midrange μ , low-mid σ_f } as well as in a band from {very low μ , high σ_f } to {low μ , low-mid σ_f }. The most noteworthy difference is that the short-duration band for the full spectrum covers a wider range of σ_f and includes consistently shorter pulses than those of the constant intensity spectrum. In most cases, a perfect Gaussian suffers more from phase difference than a spectrum with more complicated structure. For the Gaussian, any change must broaden the time pulse, whereas other spectra may benefit from a frequency-dependent time-shift.

4 HHG FILTERING – AT MID-INFRARED (2UM) WAVELENGTH

Now, we consider the high harmonic spectrum generated from a $2\mu m$ source.



Shown is the HHG spectrum as obtained from numerical simulations by Dr. Carlos Hernández-García. Driving Field: $\lambda = 2\mu m$, $I = 4.5 \times 10^{14} W/cm^2$. Target: 2mm thick He gas at 200 torr. Note intensity peaks found at integer multiples of driving photon energy (. 62 *eV*). For the 2µm spectrum, we have N = 32768 data points in the frequency and time domains, with resolution of $dE \approx 0.030 \ eV$, $dt = \frac{2\pi}{Nd\omega} \approx 4.23 \ as$, respectively.

i) Constant Phase

Due to the much greater total energy spread in the 2µm spectrum, we have significantly shorter pulses when setting a constant phase. Thus, the duration scale of Figure 18 ranges from 21 - 63 as, so that important features may be seen. Essentially the same features as for the 800nm driving field occur; we see a consistent pattern of increasing σ_f resulting in decreasing duration. A few deviations appear due to the shape of the spectrum, but they are, again, relatively small.





II) CONSTANT INTENSITY

Based on our previous results at 800nm, those for $2\mu m$ at constant intensity are



straightforward to interpret. Centering the filter toward the center of the spectrum

results in the same dependence of $FWHM_t$ on σ_f : first we see a decrease, then an increase as we raise σ_f . The phase in the 2µm spectrum remains relatively constant for a greater portion of the spectrum (Figure 20), and thus this explains the features over the majority of the central energies. We see a hint of the short duration band at low μ , but it is less significant than for 800nm HHG (Figure 11).

III) TRUE SPECTRUM

The true spectrum (as obtained from simulations) lowers durations slightly, but is otherwise very much like the constant intensity case. The patterns remain intact and the shorter pulses still lie on the upper edge of our target durations – the actual intensity leaves the shortest pulses unchanged.



The advantage of the 2µm spectrum over the quite similar 800nm one is the

wide array of filters that result in pulses of < 125 as duration. We have the choice of central energies from 130 - 500 eV and filter widths as low as 12 eV or as high as 25 eV. With relative freedom and precision, we can choose central energies sufficient for inner-shell ionization.

5 PHOTOIONIZATION

To begin our study of photoionization, we chose simple pulses with either a sin² electric field envelope (Figure 23) or a pulse train of three sin² pulses with the center pulse having twice the amplitude (Figure 22). When comparing the two laser pulses, we: either match the maximum intensity or match the total energy (Figure 24). These choices are separated by a factor of $\sqrt{2}$, but it is useful to consider both, as certain processes depend exclusively on total energy, while others are also affected by maximum intensity. We first consider Hydrogen, with lasers of sufficiently low wavelength (thus, high energy) that traditional single-photon ionization occurs. We then discuss multi-photon ionization – where higher wavelength light can ionize atoms if the laser intensity is sufficient that multiple photons are simultaneously absorbed. Finally,









I) SINGLE PHOTON IONIZATION

For $15.5 \, eV$ UV light, our photon energy is larger than the Hydrogen binding

energy (13.6 eV). Thus we expect single photon ionization to dominate and predict

 $P_{ionization} \propto Intensity$. We do indeed find this to be the case across a wide range of

intensities (Figure 25). Above $I = 3 \times 10^{14} W/cm^2$ (P(Ion) = 45%), we begin to





Same as Figure 25, but including the three pulse sequence lasers with matched energy or peak intensity.

'saturate' the atom and the probability of ionization increases more slowly. We note that ionization probability does not drop below $\sim 4 \times 10^{-10}$, regardless of the pulse applied. Due to the computational limits, this is the effective zero for Hydrogen in our simulations.

For the other envelopes, we see a similar proportionality (Figure 26). Matching the total pulse energy leads to very similar ionization probabilities, while intensity peak matching results in proportionally lower ionization.

II) MULTI-PHOTON IONIZATION

Turning to photon energies much less than the H binding energy, we investigate multi-photon ionization. In particular, we choose a 1.55 eV IR pulse, requiring absorption of at least nine photons to ionize. Here, we observe no ionization until

intensities (Figure 27) many orders of magnitude larger than that required for the single-photon process above. Then, for a relatively brief intensity range, ionization probability is proportional to intensity raised to a certain power – here $P(Ion) \propto I^{6.1}$. We are not aware of a theoretical justification for this particular power, though similar values have been reported elsewhere. Above this 'power law regime,' the ionization probability saturates, and we see a slower increase to 100% ionization probability.

Looking to the other pulse envelopes, we see the single sin² pulse more closely corresponds to the peak intensity-matching triple pulse sequence than the energymatching one (Figure 28). For a multi-photon process, the peak intensity is the deciding factor in contrast to the single-photon ionization case. When we match the peak intensity, the ionization is naturally more similar, though different by a small factor due to the *width* of the peak. Matching the total pulse energy, we considerably increase the peak intensity. Resulting in both larger ionization probability and a somewhat different





iii) Noble Gas Ionization

While showing the same qualitative features of single-photon Hydrogen ionization, there are a number of interesting factors to be noted for the Neon ionization data (Figure 29). First, the calculation takes significantly longer due to the need of a



finer spatial grid (to capture the more complex potential structure). Even with the fine

spacing, the "zero" ionization level, 1.7×10^{-6} , is much higher than desired. The data fits a power law quite well for all points with probability above the zero-level. However, this data does not fit the $P(Ion) \propto I$ relationship expected given Neon's outermost electron binding energy of $21.5 \ eV$. Instead, we see $P(Ion) \propto I^{1.6}$, suggesting a multiphoton process. In fact, the electron energy we calculate for the chosen grid parameters is $-28 \ eV$, making this indeed a two-photon process. Tests with a finer grid show much better agreement ($-23 \ eV$ if we halve the spacing), suggesting the issue is not with the special form of the potential. Unfortunately, these finer grids result in propagation computation times that we deemed unreasonable for this project.

Discussion and Further Research

We have seen that simple Gaussian filters can be used to refine high harmonic spectra, resulting in pulses on the 100 attosecond scale. These low-duration pulses are ideal for probing electron dynamics within atoms and molecules. The HHG spectra for an 800nm driving field provides short pulses with energies suitable for valence-shell ionization, while the 2µm HHG can supply short pulses with a wide range of higher energies suited for inner-shell ionization. In both cases, duration is dominated by the variation of the phase in the HHG spectrum, with relatively little dependence on the intensity spectrum. Our solution of the time-dependent Schrödinger equation allows simulation of photoionization for a variety of atomic and laser parameters. With Hydrogen, we observe single- and multi- photon processes without resorting to perturbation theory. In both cases, a simple probability-intensity power law is observed up to intensities of at least $10^{14} W/cm^2$ (after which expected saturation occurs). Results for different pulse envelopes are likewise understood theoretically. Multi-electron systems can be approximated through the use of the SAE model and analytic fits, but require long computation times to ensure accuracy.

In the near-term, we hope to continue to understand the development of the phase-pulse duration relationship for HHG spectra. We pursue a more general model for other driving laser and target parameters. In addition, attention will be given to the shape of time domain pulses, considering possible uses for the non-Gaussian structure. Then, we may collaborate with experimental researchers, suggesting procedures and comparing theory to their observation. Considering photoionization, we plan to optimize our computational methods so that noble gases can be efficiently simulated. Likewise, we will analyze ionization due to filtered HHG pulses and constant-phase HHG pulses. Hopefully, comparison with more traditional envelopes (sin², Gaussian) on similar time scales will reveal experimentally-visible features particular to HHG pulses.

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