Abstract

We present theoretical studies of above threshold ionization (ATI) using sculpted laser pulses for noble gas atoms. The time-dependent Schrödinger equation is solved to calculate the ATI energy spectrum, and a qualitative understanding of the electron motion after ionization is explored using a classical model that solves Newton’s equation of motion. Results are presented for Gaussian and Airy laser pulses with identical spectral intensities, but differing spectral phases. The simulations show that the third order spectral phase of the Airy pulse, which alters the temporal envelope of the electric field, causes changes to the timing of ionization and the dynamics of the rescattering process. Specifically, the use of Airy pulses in the ATI process results in a shift of the Keldysh plateau cutoff to lower energy due to a decreased pondermotive energy of the electron in the laser field. A reduced electron yield is also observed for the low-energy direct electrons liberated by an Airy pulse. Additionally, the side lobes of the Airy laser pulse change the number and timing of rescattering events, which results in changes to the high-order ATI plateau.

I. Introduction

One of the most instructive and valuable processes in attosecond and strong-field physics is that of above threshold ionization (ATI), in which an atom absorbs more photons than are required for ionization. The excess absorbed energy is converted to the kinetic energy of the ionized electron and the corresponding ATI spectrum has proven to be a valuable tool in many applications. In general, the ATI process can be largely understood using the three-step recollision model in which an electron is ionized and ‘born’ into the continuum (step 1), after which it accelerates in the electric field. When the electric field changes sign, it causes the electron to reverse direction (step 2) and recollide with the parent ion (step 3). If the electron is elastically scattered from the ion, it can then be accelerated again by the electric field before making its way to the detector. This leads to the so-called high-order ATI (HATI) electrons, which have been shown to be of use in reconstructing electron-ion scattering cross sections that contain target structure information [1–4]. HATI electrons are also used in the laser induced electron diffraction technique, in which the recolliding electrons are used to probe the target structure with high temporal and spatial resolution [5]. Additionally, the ATI process can be used to characterize the carrier envelope phase of the laser pulse, a quantity that is vital to accurately understanding processes involving few-cycle pulses [6].

Since the ATI process was first observed [7], there have been countless experimental and theoretical studies aimed at elucidating its dynamics and developing applications based on the process. Despite these decades of study, the ATI process still has insights to share. In this work, we combine the familiar ATI process with the use of sculpted laser fields [8–11]. Unlike their more traditional sin-squared or Gaussian wave forms, sculpted wave forms can have more

* alharri@ilstu.edu
complicated envelope functions with multiple peaks, carry quantized orbital angular momentum, or exhibit self-acceleration, self-healing, and limited diffraction [12–14]. There are several studies using sculpted wave forms to examine processes such as photoionization [15–17], atomic and molecular electronic and rotational transitions [18–22], high order harmonic generation [23–26], and strong field physics with twisted electrons [27,28]. These studies have demonstrated novel physics, including alteration of selection rules [16,22], orbital angular momentum transfer [18,19], the production of twisted UV light [23–26], and the ability to control the orbital angular momentum of XUV probes [23,25,29]. However, the ATI process, and the effects of sculpted wave packets on the ATI energy spectrum, have received only limited attention for spatially sculpted wave packets [30–32]. In these studies, it was demonstrated that a vortex pulse alters the ATI energy spectrum with an increased ionization probability as the orbital angular momentum of the vortex increases. Additionally, the photoelectron is accelerated in the laser propagation direction, something that does not occur for plane wave pulses.

Here, we present a theoretical study of ATI in noble gas atoms using few-cycle temporal Airy laser pulses. We present numerical simulations of ATI for hydrogen, helium, neon, argon, xenon, and krypton using the time-dependent Schrödinger equation (TDSE) and combine this with analysis from a classical model that is aimed at elucidating the effects of the sculpted laser fields on the ionized electron dynamics. We show that the direct and rescattering plateau cutoffs in the ATI energy spectrum shift to lower energies for Airy pulses, and our classical simulations indicate that this shift in electron kinetic energy can be traced to altered trajectories in the laser field following ionization.

The remainder of the paper is organized as follows. Section II contains the basic theoretical and numerical methods used. Section III presents the results along with a discussion of their significance. Lastly, section IV provides a brief summary and outlook. Atomic units are used throughout unless otherwise noted.

II. Theory
A. TDSE

Because the primary dynamics of ATI with linearly polarized pulses happen along the laser polarization direction, a one-dimensional approximation is typically sufficient to capture the important physics. We solve the one-dimensional TDSE for a single active electron atom in a laser field

\[
\frac{i\partial}{\partial t} \psi(x, t) = \left[ -\frac{1}{2} \frac{d^2}{dx^2} + V_a(x) + xE(t) \right] \psi(x, t). \tag{1}
\]

The atomic potential \( V_a(x) \) is approximated with the pliant core model [33]

\[
V_a(x) = -\frac{1}{(|x|^\alpha + \beta)^{1/\alpha}}, \tag{2}
\]

which has been shown to most reasonably approximate the results of a full three-dimensional TDSE calculation for HHG [33] and ATI [34]. For the pliant core model, \( \alpha = 1.5 \) and the values for \( \beta \) are given in Table 1 for the different noble gas atoms.

| Atom | \( \beta \) for pliant core potential of Eq. (2) |
|------|---------------------------------------------|
|      |                                             |


| Element | Parameter $\beta$ |
|---------|------------------|
| H      | 1.45             |
| He     | 0.49             |
| Ne     | 0.62             |
| Ar     | 1.11             |
| Kr     | 1.37             |
| Xe     | 1.78             |

Table 1 Parameter $\beta$ for the pliant core potential function for the noble gas atoms [33].

Two different temporal laser pulse fields $E(t)$ are used: a Gaussian pulse and a truncated Airy pulse. The untruncated Airy function is a solution to the free particle Schrödinger equation [14] with infinite transverse extent and infinite energy (much like the plane wave). Alternatively, the truncated Airy pulse is a finite-width wave packet with finite energy and is a more reasonable model of a physical Airy laser pulse. Additionally, the truncated Airy pulse provides a tunable parameter $\phi_3$ that can be used to adjust the shape of the envelope (see Fig. 1). The Gaussian pulse is given by

$$E_G(t) = E_0 e^{-2 \ln 2 \left( \frac{t-t_c}{\Delta \tau} \right)^2} \sin(\omega_0(t-t_c))$$

and the truncated Airy pulse is given by [35]

$$E_A(t) = E_0 \sqrt{\frac{\pi}{\Delta \tau}} \text{Ai} \left( \frac{\tau}{\Delta \tau} \frac{t-t_c}{\Delta \tau} \right) e^{\ln 2 \left( \frac{2\pi}{3} (t-t_c) \right)} \sin(\omega_0(t-t_c)),$$

where $\tau_{1/2}$ is the exponential truncation half-life of the Airy, $\omega_0$ is the carrier frequency, $\Delta \tau$ is the stretch of the Airy, $\Delta t$ is the FWHM of the temporal intensity, $\tau$ is the shift of the Airy, $t_c$ is the center of the pulse, and $\tau_0 = \left( \frac{|\phi_3|}{2} \right)^{1/3}$ is a parameter related to the third order term $\phi_3$ of the spectral phase. The third order spectral phase controls the temporal envelope of the field and is related to the truncation half-life, stretch, and shift of the Airy by

$$\tau_{1/2} = \frac{2(\ln 2)^2 \phi_3}{\Delta t^2}$$

$$\Delta \tau = \tau_0 \text{sign}(\phi_3)$$

$$\tau = \frac{\Delta t^4}{32(\ln 2)^2 \phi_3}.$$  

Figure 1 depicts the Airy and Gaussian pulses used here. As the magnitude of the third order spectral phase decreases, the Airy pulse envelope becomes more Gaussian-like and its width decreases. The number of side lobes of the Airy also decreases with decreasing magnitude of the third order spectral phase. If $\phi_3$ is negative, the orientation of the Airy pulse is reflected about its center. We refer to these pulses as inverted Airy pulses.
Figure 1 Temporal electric fields for the Airy and Gaussian laser pulses used in the calculations. The shaded areas represent the pulse envelope. Pulse parameters were $\omega_0 = 0.057$ a.u. ($\lambda = 800$ nm), $E_0 = 0.0533$ a.u. $(I = 1 \times 10^{14}$ W/cm$^2$), $\Delta t = 330$ a.u., $t_c = 3000$ a.u. The third order phase terms $\phi_3$ for the Airy and inverted Airy pulses are shown in the figure.
A unique feature of the Gaussian and Airy pulses used here is that their spectral intensities are identical, and the frequency domain electric fields differ only by a phase. The Gaussian frequency domain electric field is

\[ E_G(\omega) = \frac{E_0 \Delta t \sqrt{\pi}}{2^{3/2} \sqrt{\ln 2}} e^{-\Delta t^2 (\omega_0 - \omega)^2 / (8 \ln 2)} \]  

and the Airy frequency domain electric field is [35]

\[ E_A(\omega) = E_G(\omega) e^{-i \phi_3 (\omega - \omega_0)^3} \]  

where \( \phi_3 \) is the same third order term of the spectral phase as described above.

In solving the TDSE, the initial state atomic wave function was found by imaginary time propagation, and the 1D TDSE was solved using the Crank-Nicolson method [36]. Absorbing boundary conditions [37] were used to prevent reflections from the grid boundary and the density of the wave function was checked at each time step to ensure that no probability was lost. The ATI energy spectrum was calculated using the window operator technique [38,39]. All codes are available through figshare.com [40–42] and details of the numerical calculations are provided in Appendix A.

B. Classical Model

Once the electron has been ionized from the atom, it moves in the presence of a classical electric field. Thus, it is possible to calculate its classical trajectory and kinetic energy at the detector by solving Newton’s equation of motion

\[ \ddot{x} = -E(t). \]  

We modified the ClassSTRONG program [43] to solve Eq. (10) for the ionized electron motion in either an Airy, inverted Airy, or Gaussian laser pulse. From this, the kinetic energies of the direct and rescattered electrons were found as a function of ionization time. Additionally, the times of recollision and the classical trajectories were calculated from solving Eq. (10). Results from the classical models are used to provide qualitative physical insight into the electron dynamics for the different pulse types.

III. Results and Discussion

A. Hydrogen Energy Spectra

In general, the ATI energy spectrum shows two distinctive plateaus that occur in regions of low and high energy. In the Keldysh regime [44], photoelectron kinetic energies are below approximately \( 2U_p \), where \( U_p = \frac{e^2 A_0^2}{4m} \) is the pondermotive energy [45]. In this regime, the ATI electrons are a result of direct ionization in which the electrons make their way to the detector without further interaction with the parent ion. The second plateau that is observed in the ATI spectrum is referred to as the high-order ATI (HATI) plateau, and it occurs for photoelectrons with energies between about \( 2U_p \) and \( 10U_p \). It can be largely understood through the latter two steps of the three-step recollision model. Following ionization, the electron is accelerated by the electric field, which ultimately changes sign, causing the electron to reverse direction (step 2)
and recollide with the parent ion (step 3). If the electron is elastically scattered from the ion, it can then be accelerated again by the electric field before making its way to the detector. The maximum kinetic energy of the rescattered electron occurs for the backscattering geometry, and classical momentum conservation yields a cutoff energy of approximately $10U_p$ [46]. The probability of HATI electrons is approximately the same for energies between 2 and $10U_p$, leading to the HATI plateau.

We performed calculations for an 800 nm wavelength laser pulse ($\omega_0 = 0.057$ a.u.) with intensity of $I = 1 \times 10^{14}$ W/cm$^2$ ($E_0 = 0.0533$ a.u.). The pulse had either a Gaussian or truncated Airy envelope with full-width half-max of $\Delta t = 330$ a.u. (approximately 6 cycles) temporally centered at $t_c = 3000$ a.u. Results are presented for Airy pulses with third order phase terms $\phi_3 = \pm 1 \times 10^7, \pm 3 \times 10^7$, and $\pm 5 \times 10^7$. We begin with a detailed discussion of the ATI spectra for hydrogen atoms, and in section III.C, expand to examine spectra for all noble gas atoms.

Figure 2 shows the ATI energy spectra for a model hydrogen atom for Gaussian, Airy, and inverted Airy pulses. The Keldysh plateau for energies less than $2U_p$ and the HATI plateau for energies between $2U_p$ and $10U_p$ are clearly visible. For the Gaussian pulse, the low energy cutoff is around $2U_p$, and this does not change for the Airy pulses with the smallest values of $|\phi_3|$. The similarity of the spectra for small $|\phi_3|$ is expected since the electric fields are quite similar (see Fig. 1). As $|\phi_3|$ increases, the low energy ATI spectrum cutoff decreases to around $1.5U_p$ for the Airy pulses and the electron yield is also diminished. This indicates that there are fewer direct electrons liberated by Airy or inverted Airy pulses and these electrons gain less kinetic energy during their time in the laser field. This shift in low-energy cutoff is a direct result of a decreased maximum electric field of the Airy and inverted Airy pulses relative to the Gaussian pulses. For example, for $|\phi_3| = 5 \times 10^7$, the maximum electric field strength of the Airy pulse is 74% of the maximum for the Gaussian pulse. The reduced magnitude of the Airy or inverted Airy electric field causes a reduction in the pondermotive energy, which shifts the low-energy cutoff to smaller kinetic energies. Larger values of $|\phi_3|$ result in smaller values of the pondermotive energy and the cutoff shifts to even lower energy. These results are confirmed by the classical calculations shown below in section III.B. The values of the pondermotive energies relative to the Gaussian pulse pondermotive energy are listed in Table 2 for the different Airy and inverted Airy pulses used here.

| Pulse type                  | $U_p$ relative to Gaussian $U_p$ |
|-----------------------------|----------------------------------|
| Gaussian                    | 1                                |
| Airy/Inverted Airy $|\phi_3| = 1\times10^7$ | 0.87                            |
| Airy/Inverted Airy $|\phi_3| = 3\times10^7$ | 0.66                            |
| Airy/Inverted Airy $|\phi_3| = 5\times10^7$ | 0.55                            |

Table 2 Pondermotive energies relative to the Gaussian value for the Airy and inverted Airy pulses used in the calculations.

In the HATI plateau, differences are again most apparent for the larger values of $|\phi_3|$. As $|\phi_3|$ increases, the cutoff value for the HATI plateau shifts to lower energies for the Airy pulse, much as it did for the Keldysh plateau. However, the cutoff for the inverted Airy pulse shows little change from the Gaussian pulse. The physical explanation of these shifts is slightly more complicated due to the rescattering dynamics involved. Certainly, a reduction in the
pondermotive energy for electrons moving in the Airy or inverted Airy fields can cause a shift in the plateau cutoff to lower energy. However, as we detail below in section III.B, the number and timing of the rescatterings changes for the different pulse types and this accounts for the different shifts observed in the Airy and inverted Airy pulses.

Figure 2 ATI photoelectron energy spectra for a hydrogen atom calculated with the 1D TDSE. The laser parameters are the same as Fig. 1. For values of $|\phi_3| < 10^7$, the Gaussian and Airy spectra are very similar and therefore not shown. The black line is the spectrum for a Gaussian pulse (identical in all panels) and the red (orange) line is for a truncated Airy (inverted Airy) pulse. Third order phase values are listed in the figure. The blue dotted vertical lines represent the $2U_p$ and $10U_p$ cutoffs for the Gaussian pulse ($U_p = 5.96$ eV).

B. Classical Simulations

By solving Newton’s law for the motion of an electron in an oscillating electric field [43], the times of recollision ($t_{rec}$) as a function of ionization time ($t_{ion}$) can be found. The ionization time is defined as the time that the electron begins to move classically in the electric field, i.e. its ‘birth’ time. If the oscillating electric field drives the electron back to the parent ion, its position will return to the origin, and the time that this occurs is defined as the recollision time. It is possible that the electron returns to the origin at multiple instances as it is forced back and forth in the electric field, in which case there are multiple recollision times for a single ionization time.

Figure 3 shows a plot of the time difference between ionization and recollision $\Delta t$ as a function of ionization time for the three pulse types. A single point on the graph indicates a ($t_{ion}, \Delta t$) pair and a higher location of the point on the vertical axis indicates that the recollision occurs later after ionization. A vertical line drawn through the plot reveals that the number of $\Delta t$ values for a given $t_{ion}$ (i.e. vertical line crossings) equals the number of recollisions for a
given ionization time. In all cases, there are more recollisions for electrons ionized earlier in the pulse than later in the pulse. This is expected given that the later an electron is ionized, the fewer cycles of the electric field it experiences, and thus the fewer recollisions.

Figure 3 shows that the different pulse shapes lead to different electron dynamics after ionization. For the inverted Airy pulse, there are fewer recollisions because the leading side lobes cause an increased electric field strength at earlier times. This enhanced electric field is able to drive the electron sufficiently far away from the origin that even the strong oscillations caused by the main lobe are too weak to bring the electron back to the parent ion. As $|\phi_3|$ increases, this effect is enhanced and fewer rescattering events are observed. These dynamics are expected to result in direct electrons coming from earlier ionization events and overall fewer rescattered electrons for the inverted Airy pulse, a result that is confirmed in the analysis of direct and rescattered electron kinetic energies shown below (Fig. 4).

In contrast, electrons ionized by the Airy pulse experience additional recollisions at later times compared to the Gauss and inverted Airy pulses. These additional recollisions are caused by the trailing side lobes of the Airy pulse, which are able to ionize electrons at later times and cause electron oscillations that result in additional recollisions. Again, as $|\phi_3|$ increases, this effect is enhanced and more rescattering events are observed. In the case of the Airy pulse, the post-ionization dynamics are expected to result in fewer overall direct electrons and more rescattered electrons at later ionization times. These predictions are also confirmed in the kinetic energy spectra of Fig. 4.

![Figure 3](image)

Figure 3 Time difference $\Delta t$ between ionization and recollision as a function of ionization time $t_{ion}$ calculated with the classical model. Each data point represents a recollision event. Times are in multiples of the electric field period ($T = 110$ a.u.) and $t_c$ is the temporal center of the pulse ($t_c = 3000$ a.u.). The electric fields are shown in each panel along the horizontal line at 12T. The third order spectral phase is listed at the top of each column for the Airy and inverted Airy pulses and the Gaussian plots in (a,d,g) are identical.
Further information about the dynamics of the electrons after ionization can be found from examining the kinetic energy of the ionized electrons at the detector. As discussed above, direct electrons have a lower maximum kinetic energy than rescattered electrons and therefore, the kinetic energy can be used to separate direct and rescattered electrons. Figure 4 shows the classical prediction for the kinetic energy of the direct and rescattered electrons as a function of the ionization time. The thin horizontal lines in each panel represent the $2U_p$ and $10U_p$ energy cutoffs for the different pulses (adjusted to account for changes to $U_p$ based on pulse type). Figure 4 shows that the maximum kinetic energies of the direct and rescattered electrons are accurately predicted by the $2U_p$ and $10U_p$ values, as long as the pondermotive energy is calculated using the maximum of the electric field strength, which changes with pulse type.

The laser pulse shape has a clear effect on the shape of the kinetic energy spectrum for the direct electrons. The maximum in the direct electron kinetic energy spectrum coincides with the maximum of the pulse envelope and therefore, the direct electrons with the largest kinetic energy are produced at earlier times for the inverted Airy pulse and later times for the Airy pulse. The direct electron kinetic energy spectrum also shows the multiple peak structure of the pulse envelopes. The secondary lobe of the Airy pulse results in the production of direct electrons at later ionization times that are not present for the Gaussian or inverted Airy pulse. Likewise, the inverted Airy pulse produces earlier direct electrons that are not present with the Gaussian or Airy pulses.

The laser pulse shape also affects the kinetic energy spectrum for the rescattered electrons, although this effect is more pronounced for electrons ionized at later times. As was shown in Fig. 3, the Airy pulse produces electrons at later times that undergo rescattering, while the inverted Airy and Gaussian pulses do not. These additional rescattered electrons are due to the trailing side lobes of the Airy pulse and the rescattered electron kinetic energy spectrum confirms their presence for ionization times $t_{ion} > 6T$. Conversely, Fig. 3 showed that there were fewer rescatterings for electrons ionized early by the inverted Airy pulse compared to the Gaussian and Airy pulses. This does not, however, alter the rescattered electron kinetic energy spectrum for the inverted Airy pulse at early $t_{ion}$ values. Rather, the rescattered spectra for the Airy and inverted Airy pulses look quite similar for $t_{ion} < -6T$. This indicates that while the inverted Airy pulse resulted in fewer rescattered electrons, the kinetic energy of these electrons was the same as those produced by the Airy pulse.

Overall, the Gaussian pulse leads to the highest energy direct and rescattered electrons due to its larger pondermotive energy. The classical model’s prediction of fewer recollisions and lower energies of direct and rescattered electrons is consistent with the results of the TDSE calculations. The lower energy cutoffs observed in the TDSE ATI spectrum in Fig. 2 are explained by the lower energy direct and rescattered electrons predicted by the classical model with reduced pondermotive energy. Lower energy direct electrons produced by Airy and inverted Airy pulses cause the Keldysh plateau cutoff to shift to lower energy compared to that of the Gaussian pulse. Likewise, fewer recollisions and lower maximum kinetic energies of electrons generated by the Airy pulse results in a shift to lower energy of the rescattering plateau relative to the Gaussian pulse.

While the three pulse types used here have identical spectral intensities, their temporal shapes are quite different due to their third order spectral phases. As a result of the changes to the temporal envelope, the dynamics of the ATI process are altered. Enhanced or diminished rescattering is a result of the multiple peak structure of the Airy and inverted Airy pulses and reduced photoelectron kinetic energies result from decreased field strength of the sculpted
pulses. As the third order spectral phase increases, the pulse duration is extended and ionization and rescattering are observed over a longer period of time.

Recent studies have shown that sculpted photoelectrons can be produced through the interaction of sculpted laser pulses with atoms [32,48,49] and investigations of the effect of these sculpted photoelectron wave packets on the HHG process have shown that they can alter the polarization and propagation directions of the emitted pulses [27,28]. It should be expected that other sculpted laser pulses, such as the Airy pulse, will affect the properties of the photoelectrons, and in turn, processes involving these photoelectrons. Our results provide early evidence that the dynamics of ATI photoelectrons are altered by the use of Airy wave packets. We plan further investigation into carrier envelope phase dependence and stereo ATI simulations in order to better understand how the ATI photoelectrons are altered by sculpted pulses.

The ATI process is also used to provide information about the target atom and parent ion during the recollision process [5], and comparison of ATI spectra for varying sculpted laser pulses may be useful in providing more detailed information about the target. Sculpted electron wave packets have been shown to be useful probes of oriented targets, angular momentum-dependent effects, or multi-electron effects [50–57], and this provides an incentive to determine if similar properties can be leveraged for sculpted laser pulses and their subsequent photoelectrons.

Figure 4 Kinetic energy spectra for direct (a-c, g-i,m-o) and rescattered (d-f,j-l,p-r) ATI electrons as a function of ionization time. Results were calculated with the classical model and $t_{\text{ion}}$ is given in units of the electric field period ($T = 110$ a.u.). The kinetic energies are shown in units of the Gaussian pulse pondermotive energy ($U_p = 5.96$ eV). The thin horizontal lines in each panel represent the $2U_p$ and $10U_p$ cutoffs adjusted for the different pulse types. Relative values of $U_p$ for the pulses are listed in Table 2.
C. Noble Gas Atoms

To test our model predictions beyond the hydrogen atom, we have used the TDSE to compute the ATI spectra for all noble gas atoms using Gaussian, Airy, and inverted Airy pulses. The same laser parameters as in Figs. 1-4 were used, and the model potentials were given by Eq. (2). Results are shown in Figs. 5-9. Because the classical equations of motion depend only on the electron in the presence of the laser field, they are independent of target atom and the same as those discussed in III.B. Thus, Figs. 3-4 apply to all target atoms.

In general, many of the conclusions of sections III.A do not change for different target atoms. For argon, krypton, and xenon, the Keldysh plateau and HATI plateau cutoffs shift to lower energies as the magnitude of the third order spectral phase of the Airy and inverted Airy pulses increases. Because this feature was traced to the change in the pondermotive energy, it is a feature of the laser field and should be independent of target atom.

One noticeable difference, however, is that for helium and neon, the ATI spectra for the Airy, inverted Airy, and Gaussian pulses are very similar. For these atoms, no noticeable plateaus appear in the spectrum, but rather a mostly monotonic decay is observed with increasing energy. This feature has been observed previously in ATI spectra [1,47] and is attributable to the dependence of the photoelectron yield on the differential scattering cross section [1,2]. The energy dependence of the differential cross sections varies with target atom and this variation results in the observed changes in the HATI plateau slope.

Figure 5 Same as Fig. 2 but for helium.
Figure 6 Same as Fig. 2 but for neon.

Figure 7 Same as Fig. 2 but for argon.
Figure 8 Same as Fig. 2 but for krypton.

Figure 9 Same as Fig. 2 but for xenon.
III. Summary

By solving the TDSE, we have calculated above threshold ionization spectra for noble gas atoms in the presence of sculpted laser fields. Results were compared for electrons ionized by Gaussian, Airy, and inverted Airy pulses with identical spectral intensities and an in-depth analysis of the results was presented for a hydrogen target. These calculations showed that the Keldysh plateau cutoff and the high-order ATI plateau cutoff were shifted to lower energy when either an Airy or inverted Airy pulse was used. For the Keldysh plateau, the shift of the cutoff energy was due to the reduced electric field strength, which reduces the pondermotive energy. Additionally, the direct, low-energy electron yield was reduced for the Airy and inverted Airy pulses relative to the Gaussian pulse. For the HATI plateau, a smaller, but still observable, shift in the cutoff energy was observed for the Airy and inverted Airy pulses. This shift was due in part to the reduced pondermotive energy of the electron in the sculpted electric fields, but was also influenced by changes in rescattering dynamics.

Using a classical model, we showed that the temporal pulse shape alters the post-ionization recollision dynamics. For the Airy pulse, there were more recollisions at later times due to the trailing side peaks of the pulse. In contrast, for the inverted Airy pulse, there were overall fewer recollisions due to the leading side lobes of the pulse causing ionization that drove the electron far from the origin. The classical model was also able to correctly predict the maximum kinetic energy of the direct and rescattered electrons when the change in pondermotive energy due to pulse shape was considered.

The ATI spectra for targets other than hydrogen showed similar trends, with shifts in the energy plateaus observed for argon, krypton, and xenon. However, no significant changes were observed in the ATI spectra for different pulse types for helium and neon targets. The dependence of the ATI spectrum on target atom identity is well-documented and our results demonstrate that this does not change with the introduction of sculpted pulses.

Given that the ATI process is ubiquitous in ultrafast physics, the results presented here provide qualitative and quantitative insight into the effect of sculpted laser pulses on the dynamics of the ATI process. These results lay the groundwork for applications of sculpted laser pulses in processes such as high order harmonic generation, non-sequential double ionization, streaking, RABBITT, and the attoclock. The presence of the third order spectral phase term in the complex electric field spectrum of the Airy pulse provides an additional control parameter for tuning the temporal field, while maintaining the spectral intensity.

Appendix A

A. TDSE

The 1D TDSE was solved using the Crank-Nicolson method [36]. The initial state wave function was evolved from $t = 0$ to $t = 6000$ a.u. with a time step of 0.01 a.u. The spatial grid spanned from $-3000$ a.u. to $3000$ a.u. with a step size of 0.033 a.u. Absorbing boundary conditions were employed [37] in the form of a mask function that prevented unphysical reflections at the boundaries

$$g(x) = \cos^{1/8} \left( \frac{\pi (|x| - x_{\text{max}})}{2(50 - x_{\text{max}})} \right),$$

(A1)

where $x_{\text{max}}$ is the boundary edge (i.e. 3000 a.u.). At each time step, the wave function outside the boundary (i.e. $x < -3000$ a.u. or $x > 3000$ a.u.) was multiplied by $g(x)$. 
B. Initial state wave function

The initial state atomic wave function was found by imaginary time propagation using an algorithm similar to that of the Crank-Nicolson method. The time step was 0.01 a.u. The spatial grid spanned from – 500 a.u. to 500 a.u. with a step size of 0.1 a.u. The initial guess of the wave function was [58]

\[ \psi(x, 0) = (1 + \sqrt{x^2 + 1})e^{-\sqrt{x^2+1}}. \]  

(A2)

C. Window Method

The ATI energy spectrum was calculated using the window operator technique [38,39]. The energy bin width was \( 2\gamma = 0.02 \) and the integer power for the window operator was \( n = 2 \).

D. Classical Calculations

The classical calculations for finding the recollision times and kinetic energies of the direct and rescattered electron were calculated using a modified version of the ClassSTRONG program from [43]. Simulations for the recollision times were run from \( t = 0 \) to a final time of \( t = 6000 \) a.u. with 1000 time steps. For the kinetic energies, the simulations were run from \( t = 1320 \) a.u. to \( t = 4730 \) a.u. with 7000 time steps.

Acknowledgements

We gratefully acknowledge the support of the National Science Foundation under Grant No. PHY- 207209.

References

[1] D. B. Milošević, W. Becker, M. Okunishi, G. Prümper, K. Shimada, and K. Ueda, *Strong-Field Electron Spectra of Rare-Gas Atoms in the Rescattering Regime: Enhanced Spectral Regions and a Simulation of the Experiment*, J. Phys. B: At. Mol. Opt. Phys. **43**, 015401 (2009).
[2] M. B. Gaarde, K. J. Schafer, K. C. Kuklender, B. Sheehy, D. Kim, and L. F. DiMauro, *Strong Species Dependence of High Order Photoelectron Production in Alkali Metal Atoms*, Phys. Rev. Lett. **84**, 2822 (2000).
[3] M. Okunishi, T. Morishita, G. Prümper, K. Shimada, C. D. Lin, S. Watanabe, and K. Ueda, *Experimental Retrieval of Target Structure Information from Laser-Induced Rescattered Photoelectron Momentum Distributions*, Phys. Rev. Lett. **100**, 143001 (2008).
[4] D. Ray et al., *Large-Angle Electron Diffraction Structure in Laser-Induced Rescattering from Rare Gases*, Phys. Rev. Lett. **100**, 143002 (2008).
[5] M. Meckel et al., *Laser-Induced Electron Tunneling and Diffraction*, Science **320**, 1478 (2008).
[6] A. M. Sayler, T. Rathje, W. Müller, K. Rühle, R. Kienberger, and G. G. Paulus, *Precise, Real-Time, Every-Single-Shot, Carrier-Envelope Phase Measurement of Ultrashort Laser Pulses*, Opt. Lett., OL **36**, 1 (2011).
[7] P. Agostini, F. Fabre, G. Mainfray, G. Petite, and N. K. Rahman, *Free-Free Transitions Following Six-Photon Ionization of Xenon Atoms*, Phys. Rev. Lett. **42**, 1127 (1979).
[8] Nye John Frederick, Berry Michael Victor, and Frank Frederick Charles, *Dislocations in Wave Trains*, Proceedings of the Royal Society of London. A. Mathematical and Physical Sciences **336**, 165 (1974).
[9] L. Allen, M. W. Beijersbergen, R. J. C. Spreeuw, and J. P. Woerdman, *Orbital Angular Momentum of Light and the Transformation of Laguerre-Gaussian Laser Modes*, Phys. Rev. A **45**, 8185 (1992).

[10] G. Molina-Terriza, J. P. Torres, and L. Torner, *Twisted Photons*, Nature Phys **3**, 5 (2007).

[11] G. A. Siviloglou, J. Broky, A. Dogariu, and D. N. Christodoulides, *Observation of Accelerating Airy Beams*, Phys. Rev. Lett. **99**, 213901 (2007).

[12] N. Voloch-Bloch, Y. Lereah, Y. Lilach, A. Gover, and A. Arie, *Generation of Electron Airy Beams*, Nature **494**, 331 (2013).

[13] G. A. Siviloglou and D. N. Christodoulides, *Accelerating Finite Energy Airy Beams*, Opt. Lett., OL **32**, 979 (2007).

[14] M. V. Berry and N. L. Balazs, *Nonspreading Wave Packets*, American Journal of Physics **47**, 264 (1979).

[15] O. Matula, A. G. Hayrapetyan, V. G. Serbo, A. Surzhykov, and S. Fritzsche, *Atomic Ionization of Hydrogen-like Ions by Twisted Photons: Angular Distribution of Emitted Electrons*, J. Phys. B: At. Mol. Opt. Phys. **46**, 205002 (2013).

[16] A. Picón, J. Mompart, J. R. V. de Aldana, L. Plaja, G. F. Calvo, and L. Roso, *Photoionization with Orbital Angular Momentum Beams*, Opt. Express, OE **18**, 3660 (2010).

[17] A. Surzhykov, D. Seipt, and S. Fritzsche, *Probing the Energy Flow in Bessel Light Beams Using Atomic Photoionization*, Phys. Rev. A **94**, 033420 (2016).

[18] M. Babiker, C. R. Bennett, D. L. Andrews, and L. C. Dávila Romero, *Orbital Angular Momentum Exchange in the Interaction of Twisted Light with Molecules*, Phys. Rev. Lett. **89**, 143601 (2002).

[19] C. T. Schmiegelow, J. Schulz, H. Kaufmann, T. Ruster, U. G. Poschinger, and F. Schmidt-Kaler, *Transfer of Optical Orbital Angular Momentum to a Bound Electron*, Nat Commun **7**, 12998 (2016).

[20] H. M. Scholz-Marggraf, S. Fritzsche, V. G. Serbo, A. Afanasev, and A. Surzhykov, *Absorption of Twisted Light by Hydrogen-like Atoms*, Phys. Rev. A **90**, 013425 (2014).

[21] A. Surzhykov, D. Seipt, V. G. Serbo, and S. Fritzsche, *Interaction of Twisted Light with Many-Electron Atoms and Ions*, Phys. Rev. A **91**, 013403 (2015).

[22] S. A.-L. Schulz, S. Fritzsche, R. A. Müller, and A. Surzhykov, *Modification of Multipole Transitions by Twisted Light*, Phys. Rev. A **100**, 043416 (2019).

[23] M. Zürch, C. Kern, P. Hansinger, A. Dreischuh, and C. Spielmann, *Strong-Field Physics with Singular Light Beams*, Nature Phys **8**, 743 (2012).

[24] D. Gauthier et al., *Tunable Orbital Angular Momentum in High-Harmonic Generation*, Nat Commun **8**, 14971 (2017).

[25] G. Gariepy, J. Leach, K. T. Kim, T. J. Hammond, E. Frumker, R. W. Boyd, and P. B. Corkum, *Creating High-Harmonic Beams with Controlled Orbital Angular Momentum*, Phys. Rev. Lett. **113**, 153901 (2014).

[26] R. Géneaux, A. Camper, T. Auguste, O. Gobert, J. Caillat, R. Taïeb, and T. Ruchon, *Synthesis and Characterization of Attosecond Light Vortices in the Extreme Ultraviolet*, Nat Commun **7**, 1 (2016).

[27] S. Gemsheim and J.-M. Rost, *High-Order Harmonic Generation with Twisted Electrons*, Phys. Rev. A **100**, 043408 (2019).

[28] A. S. Maxwell, G. S. J. Armstrong, M. F. Ciappina, E. Pisanty, Y. Kang, A. C. Brown, M. Lewenstein, and C. F. de M. Faria, *Manipulating Twisted Electrons in Strong-Field Ionization*, Faraday Discuss. **228**, 394 (2021).
[29] L. Rego, J. S. Román, A. Picón, L. Plaja, and C. Hernández-García, Nonperturbative Twist in the Generation of Extreme-Ultraviolet Vortex Beams, Phys. Rev. Lett. 117, 163202 (2016).
[30] B. Böning, W. Paufler, and S. Fritzsche, Above-Threshold Ionization by Few-Cycle Bessel Pulses Carrying Orbital Angular Momentum, Phys. Rev. A 98, 023407 (2018).
[31] D. Seipt, R. A. Müller, A. Surzhykov, and S. Fritzsche, Two-Color above-Threshold Ionization of Atoms and Ions in XUV Bessel Beams and Intense Laser Light, Phys. Rev. A 94, 053420 (2016).
[32] W. Paufler, B. Böning, and S. Fritzsche, Strong-Field Ionization with Twisted Laser Pulses, Phys. Rev. A 97, 043418 (2018).
[33] A. A. Silaev, M. Yu. Ryabikin, and N. V. Vvedenskii, Strong-Field Phenomena Caused by Ultrashort Laser Pulses: Effective One- and Two-Dimensional Quantum-Mechanical Descriptions, Phys. Rev. A 82, 033416 (2010).
[34] Y.-Y. Tian, S.-Y. Li, S.-S. Wei, F.-M. Guo, S.-L. Zeng, J.-G. Chen, and Y.-J. Yang, Investigation on the Influence of Atomic Potentials on the above Threshold Ionization, Chinese Phys. B 23, 053202 (2014).
[35] M. Wollenhaupt, A. Assion, and T. Baumert, Femtosecond Laser Pulses: Linear Properties, Manipulation, Generation and Measurement, in Springer Handbook of Lasers and Optics, edited by F. Träger (Springer New York, New York, NY, 2007), pp. 937–983.
[36] William H. Press, William T. Vettering, Saul A. Teukolsky, and Brian P. Flannery, Numerical Recipes in Fortran, 2nd ed. (Cambridge University Press, 1992).
[37] J. L. Krause, K. J. Schafer, and K. C. Kulander, Calculation of Photoemission from Atoms Subject to Intense Laser Fields, Phys. Rev. A 45, 4998 (1992).
[38] K. J. Schafer and K. C. Kulander, Energy Analysis of Time-Dependent Wave Functions: Application to above-Threshold Ionization, Physical Review A 42, 5794 (1990).
[39] K. J. Schafer, The Energy Analysis of Time-Dependent Numerical Wave Functions, Computer Physics Communications 63, 427 (1991).
[40] A. L. Harris, Tdse.Cn.f, Doi:10.6084/M9.Figshare.21084826, (2022).
[41] A. L. Harris, Ati.Spectrum.Id.f, Doi:10.6084/M9.Figshare.21084841, (2022).
[42] A. L. Harris, Imaginary.Time.Prop.f, Doi:10.6084/M9.Figshare.21084808, (2022).
[43] M. F. Ciappina, J. A. Pérez-Hernández, and M. Lewenstein, ClassSTRONG: Classical Simulations of Strong Field Processes, Computer Physics Communications 185, 398 (2014).
[44] M. V. Frolov, N. L. Manakov, and A. F. Starace, Analytic Formulas for Above-Threshold Ionization or Detachment Plateau Spectra, Phys. Rev. A 79, 033406 (2009).
[45] W. Becker, S. P. Goreslavski, D. B. Milošević, and G. G. Paulus, The Plateau in Above-Threshold Ionization: The Keystoate of Rescattering Physics, J. Phys. B: At. Mol. Opt. Phys. 51, 162002 (2018).
[46] G. G. Paulus, W. Becker, W. Nicklich, and H. Walther, Rescattering Effects in Above-Threshold Ionization: A Classical Model, J. Phys. B: At. Mol. Opt. Phys. 27, L703 (1994).
[47] G. G. Paulus, W. Nicklich, H. Xu, P. Lambropoulos, and H. Walther, Plateau in above Threshold Ionization Spectra, Phys. Rev. Lett. 72, 2851 (1994).
[48] W. Paufler, B. Böning, and S. Fritzsche, Tailored Orbital Angular Momentum in High-Order Harmonic Generation with Bicircular Laguerre-Gaussian Beams, Phys. Rev. A 98, 011401 (2018).
[49] K. Y. Bliokh et al., Theory and Applications of Free-Electron Vortex States, Physics Reports 690, 1 (2017).
[50] S. M. Lloyd, M. Babiker, and J. Yuan, *Interaction of Electron Vortices and Optical Vortices with Matter and Processes of Orbital Angular Momentum Exchange*, Phys. Rev. A *86*, 023816 (2012).

[51] S. Lloyd, M. Babiker, and J. Yuan, *Quantized Orbital Angular Momentum Transfer and Magnetic Dichroism in the Interaction of Electron Vortices with Matter*, Phys. Rev. Lett. *108*, 074802 (2012).

[52] R. Van Boxem, B. Partoens, and J. Verbeeck, *Inelastic Electron-Vortex-Beam Scattering*, Physical Review A *91*, 032703 (2015).

[53] C. Lei and G. Dong, *Chirality-Dependent Scattering of an Electron Vortex Beam by a Single Atom in a Magnetic Field*, Phys. Rev. A *103*, 032815 (2021).

[54] A. L. Harris, *Single and Double Scattering Mechanisms in Ionization of Helium by Electron Vortex Projectiles*, J. Phys. B: At. Mol. Opt. Phys. *54*, 155203 (2021).

[55] A. Plumadore and A. L. Harris, *Electron Spectra for Twisted Electron Collisions*, (2021).

[56] P. Schattschneider, B. Schaffer, I. Ennen, and J. Verbeeck, *Mapping Spin-Polarized Transitions with Atomic Resolution*, Phys. Rev. B *85*, 134422 (2012).

[57] A. Asenjo-Garcia and F. J. García de Abajo, *Dichroism in the Interaction between Vortex Electron Beams, Plasmons, and Molecules*, Phys. Rev. Lett. *113*, 066102 (2014).

[58] S. Majorosi, M. G. Benedict, and A. Czirják, *Improved One-Dimensional Model Potentials for Strong-Field Simulations*, Phys. Rev. A *98*, 023401 (2018).