Attosecond Streaking in the Low-Energy Region

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Abstract. The low-energy photonelectron (PEs) ionized by a single attosecond pulse can be controlled by a moderately intense infrared field (IR). The electric field of the IR pulse can drive part of the PEs back to the parent ion and induce multiple rescattering of the electrons. Interesting interference patterns are observed in the photoelectron momentum distributions, which are formed by the rescattered electrons and the directly ionized PEs. By analyzing the interference patterns with a simple semiclassical model, which considers the particular PE trajectories incorporating the rescattering with the core, we demonstrate that the low-energy attosecond streaking offers a promising method of holographic imaging of atomic and molecular potential. In addition, we show that neither strong field approximation (SFA) or Coulomb-Volkov approximation (CVA) is able to reproduce these interesting structures at the low energy region.

1. Introduction
The availability of laser pulses in the extreme ultraviolet (XUV) range with durations from tens to hundreds of attoseconds [1, 2] offers a new route to the direct observation of the fundamental dynamics of atoms and molecules on their natural time scale. By synchronizing an XUV and a few-cycle infrared (IR) pulse to interact with atoms and molecules, one can characterize both attosecond [3, 4] and few-cycle IR laser pulses [5], and most importantly measure and control ultrafast electron dynamics [6, 7, 8]. Previously, many pioneering theoretical and experimental studies have been carried out to investigate the most important rescattering process in strong IR fields. The rescattering picture is successfully used to explain the underlying physical mechanisms of many strong field phenomena including high order above-threshold ionization (ATI) [9], high-order harmonic generation [10], and nonsequential double ionization [11]. At the same time, many of these processes can be used to extract the structural and dynamical information on the target systems [12, 13]. Very recently, the holographic imaging by the electronic wavepacket in a strong mid-infrared laser field was demonstrated, which offers the opportunity to analyze both spatial and temporal information of the ion and the photoelectrons in subfemtosecond temporal and angstrom spatial resolution [14, 15]. Further control of the electronic dynamics requires that the creation and acceleration of the continuum electron be decoupled; this is difficult by using the rescattering process in a single IR or mid-IR field since the same laser field governs both events.

Decoupling can be achieved by using an XUV single attosecond pulse (SAP) [16, 17, 18] to create an electron wavepackets at a well-defined phase of a synchronized IR field which controls the subsequent dynamics of the wavepackets. Different from the case of an XUV attosecond pulse train (APT), a SAP can produce a broadband electron wavepackets. By using a slightly
higher IR intensity than that used in the conventional attosecond streaking [19], part of low-energy photoelectrons (PEs) can be driven back by the IR electric field to the ionic core several times. These rescattered PEs may interfere with the direct ionized electrons with the same energy and form very interesting interference patterns in the PE spectra [20, 21]. By varying the duration of the IR pulse, one can control the continuum-electron trajectories and the interference patterns. In this work, we analyze the time-dependent Schrödinger equation (TDSE) results with a semiclassical method and show that this type of low-energy attosecond streaking may be used for a holographic imaging of atoms and molecules, in which the rescattered electrons collide with the atomic core and encode its structural and dynamical information while the direct electrons serve as a “reference” beam. Atomic units are used throughout unless otherwise stated.

2. Theoretical Method

The time-dependent Schrödinger equation for a He atom (within the single-active electron approximation) interacting with a SAP and an IR pulse is given by

\[ i \frac{\partial}{\partial t} \psi(\mathbf{r}, t) = \left[ -\frac{1}{2} \nabla^2 + V(\mathbf{r}) + H_I(t) \right] \psi(\mathbf{r}, t), \tag{1} \]

where \( V(\mathbf{r}) \) is a model potential [20] which accurately reproduces the He ionization potential. We numerically solve Eq. (1) in the velocity gauge with \( H_I(t) = \mathbf{p} \cdot \mathbf{A}(t) \), where \( \mathbf{A}(t) \) is the total vector potential of both pulses. Both the SAP and IR pulse are polarized along \( z \)-axis.

In brief, TDSE (1) has been solved numerically in spherical coordinates using the finite difference method and split-operator propagator. The ground state wavefunction is calculated by an imaginary time propagation of an arbitrary trial wavefunction without any external fields. After the end of the laser pulses, the probability of a photoelectron with an asymptotic momentum \( \mathbf{p} \) can then be obtained by a projection of the final wave function onto the scattering states of the field-free Hamiltonian, i.e.,

\[ T(\mathbf{p}) = T(p, \theta, \varphi) = \left| \left\langle \Psi^-_{\mathbf{p}}(\mathbf{r}) \right| \Psi(\mathbf{r}, t_f) \right|^2. \tag{2} \]

For a linearly polarized pulse, \( T(\mathbf{p}) \) has an azimuthal symmetry about \( \varphi \). The corresponding azimuthal angle integrated differential ionization probability is given by

\[ P(E, \theta) = 2\pi \sqrt{2E} T(p, \theta, \varphi = 0), \tag{3} \]

where \( E \) is the electron energy.

We use a 126 as (FWHM) SPA with a central frequency of 36 eV and a peak intensity of \( 10^{12} \) W/cm\(^2\). The SPA is placed at a zero of the IR laser electric field to obtain the maximum momentum shift. The IR laser intensity, which will be used in the present work, is able to drive the low-energy electrons back to the core but is too weak to cause significant ionization.

For comparison, we also present the results of strong field approximation (SFA) [3], which neglects the Coulomb potential effects on the electron motion after its ionization. In addition, the Coulomb-Volkov approximation (CVA) [22, 23] is used for comparison, which considers the Coulomb effects in the final state by a Coulomb-distorted Volkov wave function. Within the first order of SFA, the ionization amplitude can be given by

\[ M_{\text{SFA}}(\mathbf{p}) = -i \int_{-\infty}^{\infty} dt \left\langle \psi^N_{\mathbf{p}}(t) \right| \mathbf{r} \cdot \mathbf{E}_{\text{SAP}}(t) \left| \psi_i(t) \right\rangle, \tag{4} \]

where

\[ \psi^N_{\mathbf{p}}(t) = (2\pi)^{-3/2} \exp \left\{ i \left( \mathbf{p} + \mathbf{A}_L(t) \right) \cdot \mathbf{r} - \frac{i}{2} \int_{-\infty}^{t} (\mathbf{p} + \mathbf{A}_L(t'))^2 dt' \right\}, \tag{5} \]
Figure 1. (Color online) (a) Momentum spectra of the photoelectrons by TDSE with the SAP alone. When an additional 2-cycle IR pulse is applied, the corresponding momentum spectra from different methods are compared: (b) SFA results, (c) CVA results and (d) TDSE results. Panel (e) shows the TDSE results when the duration of IR pulse is increased to 4 cycles.

is the Volkov state and $E_{\text{SAP}}(t)$ is electric fields of the SAP, $A_L(t)$ is the vector potential of the IR field. If one substitutes the Volkov state by the Coulomb-distorted Volkov state

$$\psi_{\text{CV}}^-(p,t) = \varphi^-_{p}(r) \exp \left\{ iA_L(t) \cdot r - \frac{i}{2} \int_{-\infty}^{t'} (p + A_L(t'))^2 dt' \right\},$$

where $\varphi^-_{p}(r) = \exp \left( \frac{-i}{\hbar} \right) \Gamma (1 + iv) \exp (ip \cdot r) \frac{1}{\Gamma (1 - iv)} F_1 (-iv; 1; -ipr - ip \cdot r)$ is the Coulomb continuum function, one obtains the transition amplitude under the Coulomb-Volkov approximation

$$M_{\text{CVA}}(p) = -i \int_{-\infty}^{\infty} dt \left\langle \psi_{\text{CV}}^-(p,t) | r \cdot E_{\text{SAP}}(t) | \psi_i(t) \right\rangle.$$  

With the ionization amplitude given by Eq. (4) in SFA or by Eq. (7) in CVA, the photoelectron momentum spectra Eq. (2) or energy spectra (3) can be simply evaluated by taking the square of the corresponding amplitude.

3. Results and Discussion
Firstly, for $\lambda = 750$ nm and $I_0 = 2 \times 10^{13}$ W/cm$^2$, we consider a 2-cycle trapezoidal IR laser pulse comprising a one-cycle flat top and a half-cycle turn on and turn off respectively. At this intensity, the IR field is strong enough to steer the low energy continuum electrons but is too weak to ionize the bound state significantly. Fig. 1 shows the corresponding momentum spectra calculated by TDSE, SFA and CVA respectively. From the comparisons, one notices that all the three methods can give a global shift of the spectra by the value of the IR vector potential $A(t_i)$ at the ionization time $t_i$ by the SAP. We can see that the TDSE result [Fig. 1(d)] gives very interesting interference patterns in $p_z < 0$ and a hump structure in $p_z > 0$ along the direction of the laser polarization. In contrast, because of neglecting the ionic potential on the electron motion after its ionization, SFA [Fig. 1(b)] only shows the global momentum shift. By taking account the Coulomb effects into the final wave function, CVA [Fig. 1(c)] is able to reproduce a similar hump structure along $p_z > 0$. But because both SFA and CVA neglect the further interaction of the electron with the parent ion, they cannot describe the rescattering effects, which generates the interesting interferences in $p_z < 0$. Moreover, when the duration of the IR field is increased to 4 cycles, the TDSE momentum spectra [Fig. 1(e)] show more complex interference patterns, but SFA and CVA results (not shown for the 4-cycle case) remain almost the same with those of the 2-cycle case, which further confirms that rescattering with the core plays an important role in forming the interference structures.
In the following, we focus on the electron energy spectra along the laser polarization axis [i.e., $\theta = 0$ or $\pi$ in Eq. (3)]. We perform various TDSE calculations with a same peak IR vector potential by changing the wavelength and peak intensity of the IR field. In this way, the momentum shift caused by the IR field keeps the same and we can investigate the influence of different IR laser parameters on the low-energy part of the energy spectrum. As seen in Fig. 2 (a), with the increase of the IR wavelength more oscillating patterns appear along $p_z < 0$ direction and a more remarkable hump structure is observed along $p_z > 0$ direction [Fig. 2(b)]. Obviously, in a longer wavelength IR field, the ionized electrons will spend more time to come back to the ion core. Comparing with in a shorter wavelength IR field, the influence of Coulomb potential on the ionized electrons in a longer wavelength IR field plays a more significant role especially in the low-energy part of the energy spectra.

In order to give deeper insights into the origin of the interference structures in the energy spectrum, we employ a simple semiclassical model in terms of classical trajectories [24], which is based on the following assumptions: (1) the oscillations due to interference are controlled by the time evolution phase and this quantum phase of the continuum electron accumulated in the IR field is described by Volkov phase, i.e., $e^{-iS_p}$, where $S_p(t) = \frac{1}{2} \int_0^t d\tau [p + A_L(\tau)]^2$ denotes the semiclassical action of the trajectory; (2) if the PEs undergo backscattering, they will obtain an additional phase of $\pi$ upon reflection; (3) the reflection probability of electrons with different energy equals unity. Firstly, we focus on a 750 nm 2-cycle IR field. Considering the classical trajectories of PEs that are initially ejected by the SAP with momenta $p_z(0) < 0$ along the laser polarization direction. From then on they are decelerated by the IR field. The high-energy part of these PEs just directly go to the detector with reduced negative momenta, whereas some of the low-energy PEs will be driven back to the ion core as the IR field changes direction and some of them will backscatter at the core [Fig. 3(a) inset]. If these backscattering PEs and the direct outgoing high energy PEs have same final momentum, interference will occur between these two trajectories. Despite the reflection probability is assumed to be unity, our simple semiclassical model reproduces the spacing between adjacent interference peaks very well over a wide range of energies [Fig. 4(a)]. These backscattering PEs carrying information of the ion potential may serve as the signal wave. Meanwhile the direct PEs can serve as the reference wave. Therefore, the interference pattern can be viewed as an electron hologram, within which information of the parent ion is encoded. Considering the energy spectra with $p_z > 0$ along the laser polarization direction, if the PEs obtain a initial $p_z(0) > 0$ momenta after SAP ionization, they will be accelerated by the IR field and never return back to the core. Their momenta will be simply shifted to a larger positive momentum by the vector potential of the IR laser pulse at the time.
of ionization. In [Fig. 3(b)] we perform SFA calculation (blue dashed line), which can properly describes the direct outing PEs discussed above. One notices that SFA and TDSE results agree very well at the high-energy part of the spectrum. But discrepancy obviously exists in the low-energy region, in which a hump structure is observed in the TDSE calculation but is absent in SFA result. As discussed above, some of the ionized PEs with initial momenta \( p_z (0) < 0 \) can undergo forward scattering [Fig. 3(b) inset]. The interference between these forward scattering and the direct outgoing PEs contribute to the hump structure in the low-energy regime. Thus, this hump structure is reminiscent of unexpected peaks observed in intense mid-IR laser field by the Coulomb potential effects on the low energy electrons [25, 26, 27].

Increasing the duration of the IR field from 2-cycle to 4-cycle, as expected, the energy spectra appear more complex interference structures [Fig. 3(c) and (d)]. In a 4-cycle IR field the interference between multiple return trajectories and that of single return or direct trajectories form finer structures in the low-energy part of the spectrum. For the energy spectrum along \( p_z < 0 \) direction, we show one such possible higher-order trajectory in which PEs experience forward scattering twice from the ion core in the inset of Fig. 3(c). This twice forward scattering trajectory interferes with directly ionized electron and the backscattered electron trajectory [Fig. 3(a) inset], contributing to the substructures imposed on the interference peaks observed in the 2-cycle IR laser case. The influence of higher order electron trajectories on the energy spectrum for \( p_z > 0 \) is also evident. Some PEs experiencing forward scattering [Fig. 3(b) inset] may revisit the ion a second time under the influence of a 4-cycle IR pulse and backscatter [Fig. 3(d) inset]. These backscattering PEs can interfere both with the only forward-scattering PEs and that of directly ionized PEs. By considering these three types of trajectories, our semi-classical model reproduces the interference spacings accurately [Fig. 4(b)].

To confirm the above semi-classical trajectory interference explanation, for the laser parameters used in Fig. 3(a) and (b), we also performed time-integral TDSE calculation [28], which can decompose the contributions of the PEs with positive and negative initial momentum after SAP ionization. We find that the energy spectrum with \( p_z < 0 \), as seen in Fig. 4(c), originates mainly from the interference between the direct ionization PEs with initially negative momentum \( p_z (0) < 0 \) and the PEs with initially negative negative momentum but backscattered at
Figure 4. (Color online) (a) and (b): Comparisons of peak positions of the interference pattern in the PE spectrum for: $p_z < 0$, 2-cycle IR pulse case and $p_z > 0$, 4-cycle IR pulse case. In each frame, TDSE results (open symbols) are compared with predictions of the semiclassical model (filled symbols). (c) and (d): energy spectrum calculated by time-integral TDSE for negative and positive momenta respectively. The laser parameters are same as those used in Fig. 3(a) and (b).

a later time. For energy spectra with $p_z > 0$, the hump structure is primarily caused by the interference of direct ionization PEs with initially positive momentum ($p_z(0) > 0$) and the PEs with initially negative momentum ($p_z(0) < 0$) driven back to the core and forward-scattered [Fig. 4(d)]. This time-integral TDSE calculation confirms our semi-classical model, in which the low-energy spectra are explained as the interference between different continuum PE trajectories.

4. Conclusion
In conclusion, we have shown that our low-energy attosecond steaking method provide an ideal means to steer and control rescattering electron dynamics. By comparing the TDSE results with that of SFA and CVA, we have demonstrated that Coulomb potential and the rescattering processes play significant role in formation of the low-energy interference patterns. By applying a semiclassical model to analyze the energy spectra, we have found that the interference patterns in the photoelectron spectra reveal the quantum trajectories of the continuum electrons and this low-energy streaking method has the potential to holographic imaging of the target atoms.

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