Attosecond Streaking in the Low-Energy Region as a Probe of Rescattering

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(Received 22 February 2011; revised manuscript received 31 August 2011; published 26 October 2011)

The dynamics of low-energy photoelectrons (PEs) ionized by a single attosecond pulse in the presence of an intense infrared (IR) laser field is investigated. Whereas attosecond streaking usually involves momentum shifts of high-energy PEs, when PEs have low initial kinetic energies, the IR field can control the continuum-electron dynamics by inducing PE scattering from the residual ion. A semiclassical model is used to show that particular PE trajectories in the continuum involving electron-ion scattering explain the interference patterns exhibited in the low-energy PE spectrum. We confirm the effects of the trajectories by means of a full quantum simulation.

DOI: 10.1103/PhysRevLett.107.183001

PACS numbers: 33.20.Xx, 03.65.Sq, 34.50.Rk, 42.25.Hz

The rescattering process [1,2] in strong field physics leads to many phenomena from which structural and dynamical information on the target system can be extracted. Recently, photoelectrons (PEs) undergoing laser-driven recollision were utilized to retrieve such information for both atoms [3] and molecules [4]. Very recently, an analysis of high-order harmonic generation (HHG) (from the photorecombination of returning electrons ionized from aligned N₂ molecules) was shown to provide experimental images of the attosecond (as) wave packet created in the initial ionization process [5]. An underlying premise of such imaging is knowledge or control of the continuum electron's trajectories in the strong infrared (IR) laser field. Thus, in Ref. [3], only the most energetic PEs were used in the analysis owing to the simplicity of their trajectories. Also, in Ref. [5], only short electron trajectories were selected for the tomographic reconstruction of the molecular orbital. In general, however, the dynamics of a continuum electron in the combined fields of both a strong IR laser field and an atomic or molecular potential is quite complicated. In particular, higher-order return trajectories have been shown to play an important role in above threshold ionization [6] and in HHG [7]. Control of the continuum-electron dynamics, moreover, is difficult when both the initial ionization of the active electron and its further acceleration in the continuum are governed by the same strong IR laser field.

The use of an IR field to assist ionization of atoms by an extreme ultraviolet (XUV) attosecond pulse train (APT) has been actively investigated (see, e.g., Refs. [8–12]). By fixing the phase of the IR field relative to the XUV APT, ionization of electrons can be controlled and information on transiently bound states can be obtained. Owing to the use of APTs, the bandwidth of the XUV-produced electron spectrum is quite narrow (typically $\ll 1 \text{ eV}$ [12]). Interpretation of the results is complicated owing to the

many electron wave packets produced by the APT. A similar investigation of picosecond IR laser ionization of the Li atom in the presence of a microwave field has recently been carried out [13]. Note that control of ionization is not limited to the use of an APT. Very recently, a study of multiphoton ionization of He by long two-color XUV pulses in the presence of an IR field showed how ionization amplitudes can be controlled to produce electromagnetically induced transparency in the XUV regime [14].

The production of single attosecond pulses (SAPs) [15–17] in the XUV regime enables a new class of experiments, in which a *broadband* electron wave packet is created in the continuum by a SAP and then steered (SAPs) [of control over electron motion. For PEs with high initial momenta, interaction with the parent ion can be ignored; the IR field thus simply shifts PE momenta by **A**

 $_{\rm L}(t_i)$, the

IR field's vector potential at the time of ionization t_i [19]. Attosecond streaking, which is based on this principle, has enabled measurements of electron dynamics with a time resolution approaching the atomic unit of time [20].

In this Letter, we investigate the rescattering of lowenergy PEs resulting from ionization by a few-cycle (and hence broadband) SAP in the presence of an IR laser field that is intense enough to deflect some of the PEs so that they revisit the parent ion one or more times. By analyzing the resulting interference patterns in the PE spectrum along the direction of laser polarization, we are able to identify the most important continuum-electron trajectories, thus elucidating the rescattering process. For a short IR laser pulse, we find that interference can occur between direct outgoing electrons and backscattered ones; also a hump structure may occur due to low-energy forward scattering. For a longer IR pulse, higher-order returns manifest themselves by spectra having more complex terference patterns, e.g., with substructures. Low-er attosecond streaking is thus shown to provide r means to study and control rescattering electror in the field of an ion. These results indicate the such broadband XUV + IR field investign tering phenomena for holographic ima molecules, in which the target is "so tered electrons and the directly ior" "reference" beam.

We solve the time-dep (TDSE) numerically for SAP and an IR laser pul polarized and to or active electron an a.u.) is

Since we focus on low-energy PEs, we employ a SAP similar to that produced in Ref. [15], i.e., 126 as (full width at half maximum) with a central frequency of 36 eV, but having a slightly higher peak intensity of 10^{12} W/cm². For our low-energy attosecond streaking investigation, the SAP is placed at a zero of the IR laser electric field, wh results in the maximum momentum shift of the ele that is promoted to the continuum by the SAP fr initial state. The IR laser intensity is chosen to be to affect the bound electron, but strong enoy continuum electrons. The spatial discretized he time evolution of the TDSE follow the retions of Ref. [22]: The grid spacing is 0.05 a.u me step is 0.02 a.u. The PE spectra are obtain ecting the the field-free final wave function onto energy eige boundary con-Hamiltonian that satisfy the incor dition [23]. Note that unlike ip ons of low-energy electrons produced by an A le presence of an IR field, our use of a SAP result roadband initial ionized electron momentum dist , providing greater oppord-induced interference phetunity for different frequency of our SAP is rather nomena. Since the far above the ion threshold, the probability for excitation of Ryd states is found to be <15% of the ionization pr fity; i.e., they do not play a large role in the pher ha described below.

IR pulse and A(t) is the corresponding vector potential.

Consider of st a 2-cycle IR laser pulse (with $\lambda = 750$ nm and $I_0 = 2 \times 10^{13}$ W/cm²) having a trapezoidal envelope composed of a one-cycle flat top and a half-cycle turn on an our off, as shown in Fig. 1(a). [Note that use of a

pulse shape gives qualitatively similar numerical but the results are easier to analyze for a trapezoidal c shape.] For the SAP and IR pulse intensities chosen, inization is almost entirely due to the SAP, while the IR field only affects the motion of PEs ionized by the SAP. The photoelectron energy spectra resulting from ionization of He by the SAP in the presence of the IR pulse for photoelectrons having negative ($p_z < 0$) and positive ($p_z > 0$) momenta along the laser polarization axis are shown, respectively, by the solid (red) lines in Figs. 1(b) and 1(c). One observes two distinctive features of the PE spectrum: (1) An oscillatory pattern for $p_z < 0$ [Fig. 1(b)]; (2) A hump structure at low energies for $p_z > 0$ [Fig. 1(c)]. We show below that reencounters of some PEs with the He⁺ ion are responsible for these two features.

To explain the oscillation patterns, we employ a simple semiclassical model in terms of trajectories [24]. First, consider the classical trajectories of PEs that are initially ejected by the SAP with momenta $p_z(0) < 0$ along the laser polarization axis. After ionization, they are decelerated by the IR field. So whereas the low kinetic energy part of these PEs will reverse direction and revisit the helium ion at a later time [25], the high-energy (direct) part simply quivers away with reduced negative momentum. If back-scattering occurs [Fig. 1(b), inset], the final energies of the backscattered electrons and those direct electrons having

the same negative momentum will overlap; i.e., interference occurs between these two pathways (direct and backscattered). Second, the oscillations due to interference are controlled by the time evolution phase. The quantum phase acquired by a free electron in an electromagnetic field is given by the Volkov phase, $e^{-iS_{\mathbf{p}}(t)}$, where $S_{\mathbf{p}}(t)$ denotes the semiclassical action of the trajectory,

$$S_{\mathbf{p}}(t) = \frac{1}{2} \int_0^t d\tau [\mathbf{p} + \mathbf{A}_{\mathrm{L}}(\tau)]^2, \qquad (2)$$

with $\mathbf{p} + \mathbf{A}_{\rm L}$ being the classical momentum of the PE. The reflected electrons are found (see below) to obtain an additional phase of π upon reflection (analogous to the $\lambda/2$ phase change upon reflection in optics). We assume that the reflection probability equals unity. Despite the simplicity of this semiclassical model of two interfering pathways for photoelectrons having $p_z < 0$, this model reproduces the frequency of the oscillations (i.e., the spacing between adjacent peaks) very well over a wide range of energies [cf. Fig. 2(a)]. (Qualitatively, one expects the agreement to improve for longer wavelengths owing to the greater quiver amplitude.) The exact *positions* and *amplitudes* of the interference fringes, of course, require knowledge of the quantum reflection probability. Our simple model's success indicates that the interference *spacings*

depend largely on the IR laser pulse. Note also that the interference between the direct PEs and the backscattered ones is similar to that in optical holography. The reflected PEs carry information about the parent ion in the scattering process. And the direct PEs serve as the reference wave. Therefore, the interference pattern can be viewed as an electron hologram, within which information on the parent ion is encoded.

Consider now the PE spectrum in Fig. 1(c). Upon ionization by the SAP, a PE having an initial momentum $p_z(0) > 0$ along the laser polarization axis will then be accelerated by the first half-cycle of the remaining single cycle of the IR field and will not revisit the helium ion. If we ignore the effect of the He ion potential on the PE, the final momentum of the PE will be shifted to a larger positive momentum, with the shift given by the vector potential of the IR laser pulse at the time of ionization (cf. the analysis in Ref. [19]). We illustrate the prediction



influence of a 4-cycle IR pulse. If backscattering occurs at the second encounter, these (forward and then backscattered) PEs [Fig. 3(c) inset] can interfere with both the forward-scattering-only PEs [Fig. 1(c) inset] and the PEs initially emitted with $p_z(0) > 0$. As shown in Fig. 2(b), the energy separations of the interference structures in the PE spectrum in Fig. 3(c) can be reproduced quite accurately by the semiclassical model, thus explaining their origin as due to interference between three different continuum PE quantum trajectories.

The influence of higher-order electron trajectories on the PE spectrum for $p_z < 0$ is also evident. As shown in Fig. 3(b), for the 4-cycle IR laser pulse case substructures appear on the interference peaks observed in the 2-cycle IR laser case. This indicates that additional PE trajectories with lower amplitudes contribute to the interference between the directly ionized and the backscattered PEs. In the inset of Fig. 3(b), we show one such possible higher-order trajectory in which PEs experience forward scattering from the He⁺ ion twice.

To confirm the above semiclassical trajectory interference explanation, we have performed another TDSE calculation in which we solve the time-integral equation [30] and decompose the contributions of the wave packets launched by the SAP with initial momenta along the positive and negative directions of the laser polarization axis, for the same laser parameters used in Fig. 1. We find that the PEs with $p_z < 0$ originate from (a) *direct ionization* of the PEs with initially negative momentum ($p_z(0) < 0$), (b) PEs with initially negative momenta that influence of the IR field and *forward scatter*. Figure 4(b) shows that the hump at low energies originates mainly from the forward-scattered PEs. To confirm this origin of the low-energy peak, we analyzed the time-dependent electron wave packet, which shows a return to, and subsequent exit from, a sphere around the parent ion [32].

In summary, we have investigated theoretically lowenergy photoelectron spectra for ionization of an atom by a few-cycle SAP in the presence of an intense IR laser pulse, i.e., low-energy attosecond streaking. The interference patterns in the photoelectron spectra reveal the quantum trajectories of the continuum electrons. The IR laser field is shown to provide a remarkable degree of control over the continuum-electron dynamics in the low-energy region. A short IR laser pulse can guide some initially ionized electrons back to the parent ion from which they rescatter and interfere with directly ionized electrons, thus providing a kind of holographic imaging of the ionic potential. By increasing the IR laser pulse length, multiple returns of PEs have been shown to arise. Further control of electron dynamics in the continuum will require waveform shaping of the driving laser field, e.g., by adding a second harmonic of the fundamental IR laser. Although this proposed low-energy attosecond streaking is experimentally feasible at present, a full quantum-mechanical description of the process remains a theoretical challenge. The dynamics of low-energy continuum electrons is dominated by both the strong IR laser field and the ionic core potential. Thus any analytic quantum theory of low-energy attosecond streaking that neglects this potential (such as, e.g., [33]) cannot describe the low-energy PE spectrum. Any such theory must treat not only the IR laser field but also (multiple) scattering from the ionic potential.

We gratefully acknowledge fruitful discussions with T. Morishita and O. I. Tolstikhin. This work was supported by the National Natural Science Foundation of China under Grants No. 11174016, No. 10974007, No. 10821062, and by the U.S. Department of Energy under Grant No. DE-FG02-96ER14646. X. M. T. was supported by the Japan Society for the Promotion of Science.

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