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Quantum-mechanical models for photo-ionization: Uni-directional electron re-scattering by a laser pulse

A.D. Bandrauk[a], S. Chelkowski[a], D.J. Diestler[b,c], J. Manz[c], K.-J. Yuan[c,*]

[a] Département de Chimie, Faculté des Sciences, Université de Sherbrooke, Sherbrooke, Quebéc, Canada J1K2R1
[b] Department of Agronomy and Horticulture, University of Nebraska-Lincoln, Lincoln, NE 68583, USA
[c] Institut für Chemie und Biochemie, Freie Universität Berlin, Takustrasse 3, 14195 Berlin, Germany

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ABSTRACT

The interaction of a hydrogen atom with a linearly (z-) polarized laser pulse is described by numerical solution of the three-dimensional Schrödinger equation. Photo-ionization of the ground state, with pulses having three or more optical cycles at photon energies well above the ionization potential and maximum intensity 10^{15} W cm^{-2}, yields symmetric opposite fluxes of electrons preferentially in the $+z$ and $-z$ directions. One-cycle laser pulses can lead to modest symmetry breaking, e.g., more intense electron fluxes in the $+z$ direction than in the $-z$ direction, depending on the carrier-envelope phase (CEP) of the pulse. Production of uni-directional electron fluxes, similar to those observed in Coulomb scattering, calls for laser pulses with stronger intensity, or lower carrier frequency, or shorter duration with appropriate CEP, e.g., half-cycle pulses in the attosecond time domain. Here, uni-directionality is achieved by a four-step mechanism: (i) photo-ionization, (ii) turn-around of the electron, (iii) scattering from the atom's core and (iv) drift in the forward direction.

1. Introduction

This article deals with three closely related topics concerning the interaction of short, intense laser pulses with single atoms:

(T1) *Similarities between electron scattering* [1–3] *and electron re-scattering induced by laser pulses* [4–7]. Our interest in T1 is motivated in part by promising new approaches to the time-dependent imaging of electronic structures, including electron diffraction [8,9] and orbital tomography [10,11], which is based on high harmonic generation (HHG). As a prototype, we consider the simplest system, namely the hydrogen (H) atom, for which Coulomb scattering (e + p → e + p) and photo-ionization (H + $h\nu$ → e + p) by a laser pulse are depicted in Fig. 1. A prominent feature of Coulomb scattering, which is that the distribution of scattered electrons peaks in the forward direction [1], suggests the second topic.

(T2) *Design of laser pulses that induce uni-directional ionization and corresponding symmetry breaking.* A pioneering approach to this goal (i.e., coherent control) was already suggested in 1989 by Kurizki et al. [12] for the induction of uni-directional electron currents in solids. Their prediction was verified experimentally by Dupont et al. [13]. More recently new concepts to achieve uni-directional ionization of atoms based on few-cycle laser pulses have been advanced [14–19,33].

(T3) *Mechanism of uni-directional electron re-scattering*, which is related to the so-called "poor-man's three-step model" for HHG [4]. For an analysis of this model in terms of quantum orbits, see Ref. [6].

We now briefly review previous work on these topics, in the order T2, T1 and T3, pointing out how the current study extends earlier efforts.

(T2) Rapid advances in laser physics have permitted the production of linearly (z-) polarized few-cycle pulses that are well described by a few parameters, such as intensity, duration and carrier-envelope phase (CEP) [20,21]. This development has spurred the exploitation of such pulses to steer atomic and molecular processes in desired directions (with respect to the direction of polarization of the pulse) by appropriate choices of the pulse parameters. A pioneering example is the measurement by Paulus et al. [14] of above-threshold ionization spectra of xenon, which demonstrated that by adjusting the CEP one can control the left to right ratio of the total photo-electron yield (which is dominated by direct one-photon ionization) to a precision of about 10% (greater for the high-momentum components, which arise from

* Corresponding author.
E-mail address: kjyuan@chemie.fu-berlin.de (K.-J. Yuan).

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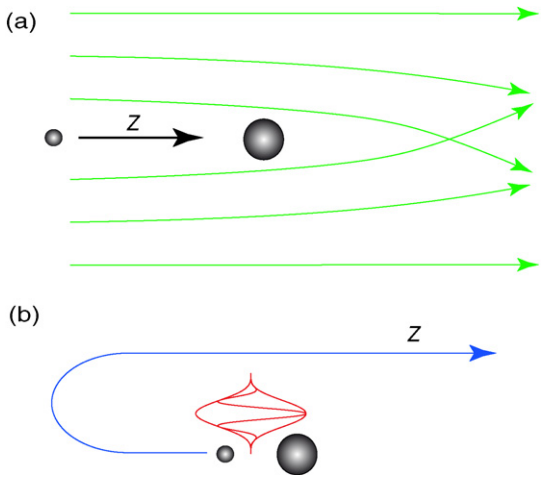

Fig. 1. Analogy between ordinary Coulomb scattering (a) and laser-driven electron re-scattering (b). Schematic trajectories of electron (small sphere) interacting with proton (large sphere) are illustrated by curved arrows. In (a), electron moves along the z-direction toward proton. In (b), linearly z-polarized laser pulse induces (i) ionization, followed by (ii) turn-around and (iii) continuation of motion preferentially in the z-direction.

electron re-scattering). At about the same time, Hu and Starace [15] conducted a systematic theoretical investigation of laser-pulse scenarios in the frequency domain of multi-photon ionization that might permit even more precisely controlled (i.e., in the ideal case uni-directional) electron detachments from a model hydride anion H^-. In particular, they showed that this demanding goal calls for very short, intense pulses (preferably with duration of just a single optical cycle) with appropriate CEP. They also demonstrated that uni-directional ionization can be achieved by a combination of two corresponding half-cycle pulses, both with the same duration and intensity but with opposite directions of the electric field and separated by an appropriate time delay. Very recently Peng and Starace [17,18] simulated photo-ionization of the H atom by accurate solutions of the time-dependent Schrödinger equation in the velocity gauge. They examined two- or more-cycle pulses with appropriate CEP and carrier frequencies ω corresponding to photon energies well above the ionization potential ($\hbar\omega \geq 36\,\mathrm{eV} = 1.3\,E_h \gg IP = -E_{1s} = 0.5E_h$, i.e., a single photon suffices to ionize the H atom). An important result is a scaling law for the asymmetry of the photo-electron distribution that depends approximately on the scaling parameter $\sqrt{I_{max}}/\omega$, where I_{max} is the maximum intensity. Specifically, in the domain $\sim 10^{12} < I_{max} < 10^{15}\,\mathrm{W\,cm^{-2}}$ single-photon ionization dominates, yielding nearly symmetric distributions. Increasing the intensity gives rise to two-photon processes, which break the symmetry, albeit with asymmetry factors below 10% (i.e., still well below the ideal goal of uni-directionality). From the scaling parameter $\sqrt{I_{max}}/\omega$ we deduce that the asymmetry should also increase with decreasing ω. These results are in accord with the general theory of the influence of the CEP [22], i.e., asymmetries in electron ionization processes reflect transition amplitudes involving both odd and even numbers of photons.

Extending the previous pioneering investigations [15,17,18], we search in more promising domains of the laser-pulse parameters for pulses that might achieve uni-directional ionization of the H atom. In particular, we employ linearly (z-) polarized pulses, whose z-component of the electric field is denoted by $E(t)$, and look for the trends that result from: increasing the maximum electric field strength E_{max}, or corresponding maximum intensity $I_{max} = c\varepsilon_0 E_{max}^2$ (where ε_0 is the electric permittivity of the vacuum and c is the speed of light) beyond $10^{15}\,\mathrm{W\,cm^{-2}}$; decreasing the pulse duration t_p up to the limit of a single cycle (with concomitant variations of the CEP η); decreasing ω up to the domain which corresponds to a photon energy just above the ionization potential ($\hbar\omega = 0.52E_h$, i.e., still in the domain where a single photon suffices to ionize the H atom). Moreover, in order to go even below the one-cycle limit, we design a so-called half-cycle laser pulse comprising an initial short, strong half-cycle component with uni-directional (positive) electric field, followed by a second long, weak half-cycle. This sequence is necessary to fulfill the condition that must be satisfied by all laser pulses: $\int_0^{t_p} dt\, E(t) = 0$ [6]. Our specific design of the half-cycle pulse, which is motivated by experimental approaches to half-cycle pulses (see footnote '1') [23], differs from the preceding scenario of two opposite half-cycle components with same duration and electric field strength [15].

(T1) Our goal of achieving uni-directional photo-ionization of the H atom $H + h\nu \rightarrow e + p$ is also motivated by the analogous dominant uni-directionality of Coulomb scattering $e + p \rightarrow e + p$, as schematized in Fig. 1(a). In the case of Coulomb scattering the incident electron approaches the atomic core (i.e., the proton) along the z-axis from the asymptotic region ($z \rightarrow -\infty$) with relative kinetic energy T. The electron then scatters with cylindrical symmetry into various angles (θ) with respect to the z-axis, according to the celebrated Rutherford formula for the differential cross-section [1]:

$$\frac{d\sigma}{d\Omega} = \left(\frac{1}{4\pi\varepsilon_0}\frac{e^2}{4T}\right)^2 \frac{1}{\sin^4(\theta/2)} \tag{1}$$

That is, the electron scatters preferentially in the forward direction (positive z, $\theta = 0$), as illustrated in Fig. 2. Note that the angular distribution is independent of T, whereas the efficiency of scattering increases quadratically with decreasing T. The total cross-section is

$$\sigma \propto 1/T^2 \tag{2}$$

Recently, the group of Lin and coworkers [24,25] compared the angular dependence (Eq. (1)) for Coulomb scattering with the intensities on so-called back-re-scattering ridges (BRR) representing electrons that have been re-scattered in the backward direction by the target ion. The agreement is good, although the comparison is restricted to the domain of backward scattering. The predicted BRR have been verified by the same theoretical group in collaboration with experimental groups [26,27]. In contrast with these earlier investigations [24,26,27], we focus on the comparison of electron scattering and re-scattering in the dominant forward direction.

(T3) Anticipating that we should be able to design a laser pulse that achieves forward electron re-scattering analogous to Coulomb scattering, we analyze the overall process in terms of sequential steps. As a working hypothesis, we assume that the laser-induced process involves three steps, the first two of which coincide with

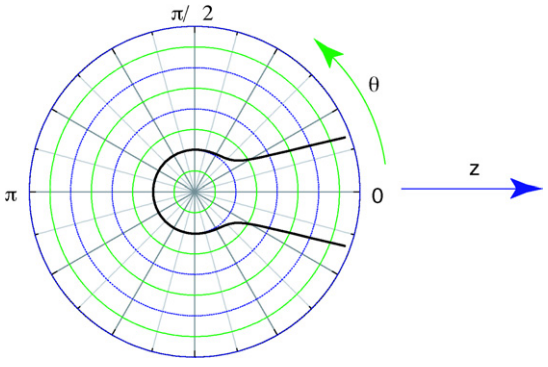

Fig. 2. Polar plot of differential cross-section for Coulomb scattering, according to Eq. (1). Most electrons are scattered in the forward direction ($\theta = 0$).

those of Corkum's "simple man's three-step model" [4]: (i) ionization of the ground-state atom (via tunneling through the Coulomb barrier, or escape over the barrier) and (ii) turn-around of the electron. In addition, Corkum's model invokes another step for HHG, namely "re-collision", in which the electron is envisioned to linger in the vicinity of the nuclear core. However, the observed small yields of high-harmonic photons suggest that the re-colliding electron has a low probability of being trapped. Rather it scatters away from the core with large probability. Thus, we take as the third step of our hypothetical scheme (iii) (forward-) scattering of the returning electron from the atom's core (i.e., from the proton in the present case of H; see Fig. 1(a)). We refer to this three-step sequence simply as "re-scattering", since the laser-driven electron originates from the target itself. The challenge is to analyze the overall process of forward re-scattering in terms of steps (i)–(iii), and possibly others.

2. Model and methods

The Schrödinger equation describing the interaction of the z-polarized laser pulse with the H atom is expressed in relative coordinates as

$$i\hbar\frac{\partial\Psi(r,\theta,t)}{\partial r} = H(t)\Psi(r,\theta,t) \tag{3}$$

where

$$H(t) = -\frac{\hbar^2}{2m_e r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial}{\partial r}\right) + \frac{\mathbf{L}^2}{2m_e r^2} - \frac{e^2}{4\pi\varepsilon_0 r} + er\cos\theta E(t) \tag{4}$$

is the Hamiltonian in the length gauge, $\mathbf{L}$ is the angular momentum, and $E(t)$ is the z-component of the electric field associated with the pulse. The initial state of the H atom is

$$\Psi(r,\theta,0) = \psi_{1s}(r) = N\exp\left(\frac{-r}{a_0}\right)Y_{00}(\theta,\phi) \tag{5}$$

where $N = 1/\sqrt{\pi a_0^3}$ is the normalization constant, a_0 is the first Bohr radius, and $Y_{00} = 1/\sqrt{4\pi}$ is the spherical harmonic.

The cylindrical symmetry of the system suggests that the wavefunction be expanded in terms of spherical harmonics with the magnetic quantum number m fixed at $m = 0$. Accordingly, we write:

$$\Psi(r,\theta,t) = \sum_{l=0}^{l_{\max}} R_l(r,t)\,Y_{l0}(\theta,\phi) \tag{6}$$

Here $l_{\max}$ is the maximum azimuthal quantum number taken into account. We also define the "excited" component of the wavefunction as

$$\Psi_{\rm ex}(r,\theta,t) \equiv \Psi(r,\theta,t) - R_0(r,t)Y_{00}(\theta,\phi) = \sum_{l=1}^{l_{\max}} R_l(r,t)Y_{l0}(\theta,\phi) \tag{7}$$

with corresponding probability density

$$\rho_{\rm ex}(r,\theta,t) = \left|\Psi_{\rm ex}(r,\theta,t)\right|^2 \tag{8}$$

Inserting Eq. (6) into Eq. (3) and operating with $\int \mathrm{d}\Omega\, Y_{l0}^*(\theta,\phi)\ldots$ on both members of the resulting equation yield a set of coupled differential equations for the time-dependent radial functions $R_l(r,t)$. These are solved numerically by the method of DeVries [28] (see also Ref. [29]). The atom is taken to be confined to an infinite spherical square well so that the radial functions are determined over the finite domain $0 \le r \le r_{\max}$ and satisfy the boundary condition $R(r_{\max},t) = 0$. The radial and temporal coordinates are discretized and the radial- and time-step sizes are set to $\Delta r = 0.125a_0$ and $\Delta t = 0.03\hbar/E_{\rm h}$, respectively; $r_{\max} = 256a_0$; $l_{\max} = 80$. To prevent spurious effects due to the reflection of the wavepacket from the boundary at $r = r_{\max}$, we multiply $\Psi(r,\theta,t)$ is by a "mask function" [30]:

$$g(r) = \begin{cases} 1, & r < r_1 \\ \{\cos[\pi(r-r_1)/2(r_{\max}-r_1)]\}^{1/8}, & r_1 < r < r_{\max} \end{cases} \tag{9}$$

For all results reported here we set $r_{\max} - r_1 = 32a_0$.

The wavepacket $\Psi(r,\theta,t)$ generates a radial flux (electron current density) given by

$$j_0(\theta,t) = \frac{\hbar}{2im}\left[\Psi^*(r,\theta,t)\frac{\partial\Psi(r,\theta,t)}{\partial r} - \frac{\partial\Psi^*(r,\theta,t)}{\partial r}\Psi(r,\theta,t)\right]_{r=r_0} \tag{10}$$

with the corresponding differential yield:

$$\frac{\mathrm{d}Y(\theta)}{\mathrm{d}\Omega} = \int_0^\infty \mathrm{d}t j_0(\theta,t) \tag{11}$$

and total yield:

$$Y = \int \mathrm{d}\Omega\frac{\mathrm{d}Y}{\mathrm{d}\Omega} \tag{12}$$

The radial flux is calculated at $r_0 = 223a_0$.

The electric field $E(t)$ is given by

$$E(t) = -\frac{\partial A}{\partial t} \tag{13}$$

where the z-component of the vector potential A satisfies the relations:

$$A(0) = A(t_{\rm p}) = 0 \tag{14}$$

and we assume that A vanishes for $t < 0$, the initial instant, and for $t > t_{\rm p}$, the final instant. Eqs. (13) and (14) ensure that the condition [6,31]:

$$\int_0^{t_{\rm p}} \mathrm{d}t\, E(t) = 0 \tag{15}$$

is satisfied automatically. We model the vector potential for few-cycle ($n_{\rm cy} = 1$ or 3 in the subsequent applications) pulses by

$$A(t) = \begin{cases} A_0\sin^4(\pi t/t_{\rm p})\sin(\omega t+\eta), & 0 < t < t_{\rm p} \\ 0, & \text{elsewhere} \end{cases} \tag{16}$$

where ω corresponds to a wavelength of $2\pi c/\omega$, the time for an optical cycle is $\tau = 2\pi/\omega$ and the duration of the laser pulse is $t_{\rm p} = n_{\rm cy}\tau$ for an $n_{\rm cy}$-cycle pulse. To explore the influence of the CEP, we set $\eta = 0$ or $\eta = \pi$. The $\sin^4$ carrier envelope in Eq. (16) is used so that the electric field (Eq. (13)), as well as its derivative $\partial E/\partial t$, vanish at the beginning ($t = 0$) and end ($t = t_{\rm p}$) of the pulse (i.e., the electric field is switched on and off smoothly [32]). In contrast, a $\sin^2$ envelope for the vector potential, which was employed, for example, by Peng and Starace [17,18], leads to rather sudden switching on and off. Note that the pulse is shorter for the $\sin^4$ compared with the $\sin^2$ envelope, as estimated from the full width at half maximum (FWHM). In the case of the multi-cycle pulses, where the envelope of $E(t)$ resembles that of $A(t)$, we infer that FWHM = 0.261 $t_{\rm p}$ and 0.364 $t_{\rm p}$ for $\sin^4(\pi t/t_{\rm p})$ and $\sin^2(\pi t/t_{\rm p})$ envelopes, respectively.

Alternatively, we employ a *half-cycle* pulse having an initial short, strong positive electric field ($0 \le t \le t_1$) followed by a long, weak negative one ($t_1 \le t \le t_2$). The strong- and weak-field components of the vector potential are defined by

$$A(t) = \begin{cases} -A_0\sin^4(\pi t/2t_1), & 0 \le t \le t_1 \\ -A_0\sin^4\left[\dfrac{\pi(t-t_1)}{2(t_2-t_1)} + \pi/2\right], & t_1 < t \le t_2 \end{cases} \tag{17}$$

For few-cycle pulses we express the intensity in terms of the electric field by $I_0 = c\varepsilon_0 E_0^2$, where $E_0 = A_0\omega$. For the half-cycle pulse the maximum of the electric field $E_{max} = (3\sqrt{3}/16)(2\pi A_0/t_1)$ occurs at $t = 2t_1/3$. In practice we set $E_{max} = 0.169E_h/ea_0$, which corresponds to the intensity $I_{max} = 2 \times 10^{15}\,W\,cm^{-2}$.

3. Results

We begin with a systematic investigation of the dependence of the differential ion yield $dY/d\Omega$ on the electric field of the laser pulse. The results are restricted, however, to some important

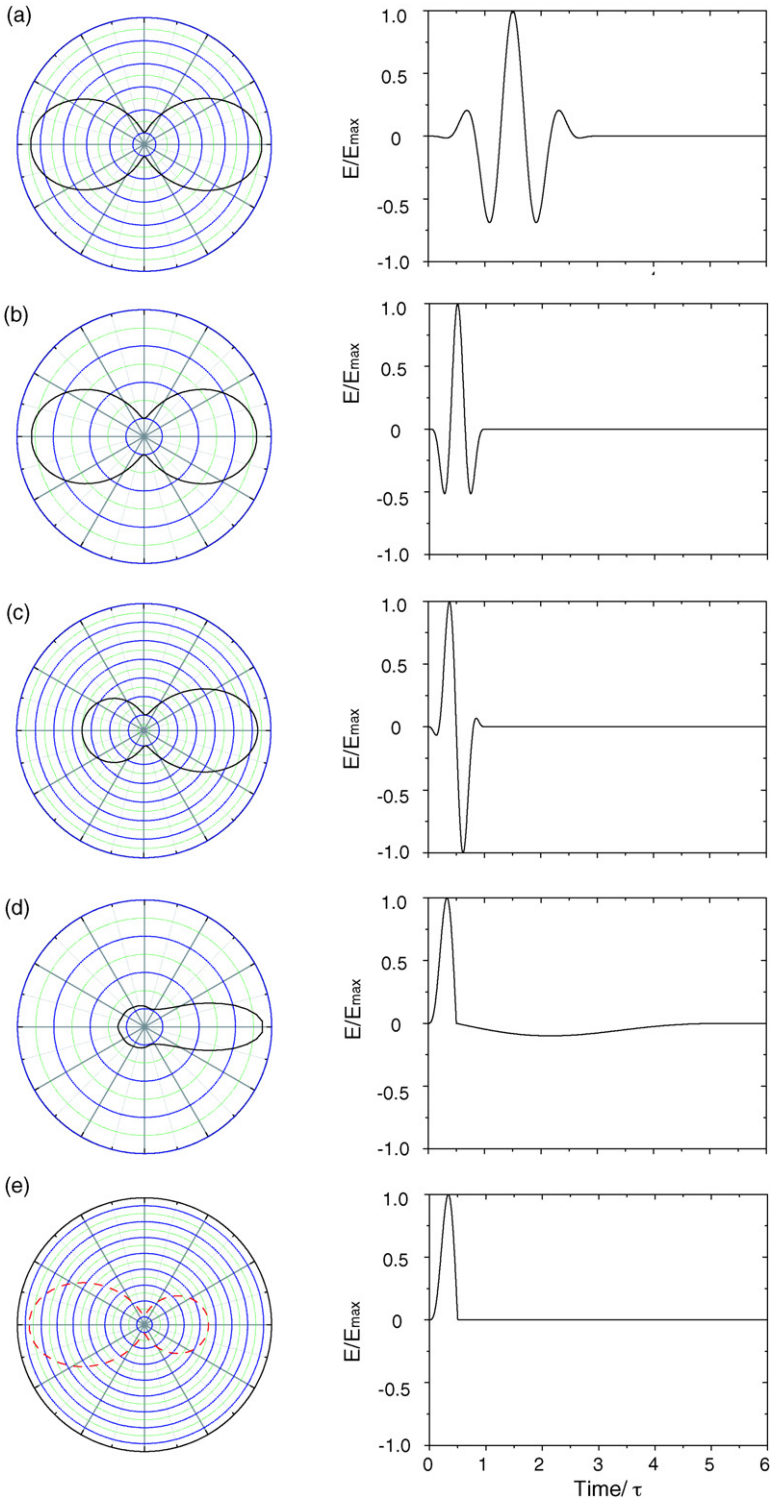

Fig. 3. Differential yields $dY/d\Omega$ (left panels, polar plots with notations as in Fig. 2) for photo-ionization of H atom (1s) by z-polarized laser pulses (right panels). (a) Three-cycle pulse ($n_{cy} = 3$, $\eta = 0$), (b) one-cycle pulse ($n_{cy} = 1$, $\eta = 0$), (c) one-cycle pulse ($n_{cy} = 1$, $\eta = \pi$) and (d) half-cycle pulse. Pulses for (a)–(c) are specified by Eqs. (13)–(16) with parameters $E_{max} = 0.169\,E_h/ea_0$ ($I_{max} = 2 \times 10^{15}\,W\,cm^{-2}$), $\hbar\omega = 0.55\,E_h$, $t_p = n_{cy}\tau$, $\tau = 276$ as. The half-cycle pulse (d) is specified by Eq. (17), with parameters $E_{max} = 0.169\,E_h/ea_0$, $t_1 = \tau/2$, $t_2 = 5\tau$, $\tau = 276$ as. Artificial half-cycle pulse (e) consists of only initial strong short positive component of pulse shown in (d).

domains of the pulse parameters, whose choice has been motivated in the Introduction. In particular, we examine the effects of pulses with I_{max} in the range 2×10^{15}–2×10^{16} W cm^{-2}, corresponding to E_{max} in the range 0.169–0.534 E_h/ea_0. Moreover, we employ photon energies in the domain $0.5E_h < \hbar\omega < E_h$ (i.e., just above the ionization potential IP = $-E_{1s} = 0.50E_h$) such that absorption of a single photon induces ionization.

Let us consider first results for multi-cycle laser pulses. As typical example, Fig. 3(a) shows the differential ion yield achieved by a three-cycle pulse with photon energy $\hbar\omega = 0.55E_h$ and intensity $I_{max} = 2 \times 10^{15}$ W cm^{-2}. It is nearly symmetric with essentially equal fluxes of electrons in opposite directions (i.e., not at all like that characteristic of Coulomb scattering; see Fig. 2). Similar results are obtained for other multi-cycle pulses. We surmise that uni-directional photo-ionization requires shorter pulses. Fig. 3(b) and (c) display results for single-cycle pulses with the same photon energy, $\hbar\omega = 0.55E_h$, but with different phases, $\eta = 0$ and $\eta = \pi$, respectively. The corresponding differential ion yields clearly are nearly symmetric and asymmetric, respectively. We conclude that a single-cycle laser pulse may break the symmetry of the electron flux, depending on the phase. Other effects of symmetry breaking by a single laser pulse have been discovered previously (see, for example [33,34]). The degree of symmetry breaking, however, is rather modest (i.e., even though most of the electrons are emitted in the forward direction, the fraction driven backward remains significant). This observation accords with the analysis of Peng and Starace [17,18]. That is, for the present laser parameters dominant single-photon transitions to the ionization continuum essentially dictate an overall symmetric differential ionization yield, with rather modest effects of symmetry breaking due to small contributions from two-photon processes, depending on the CEP.

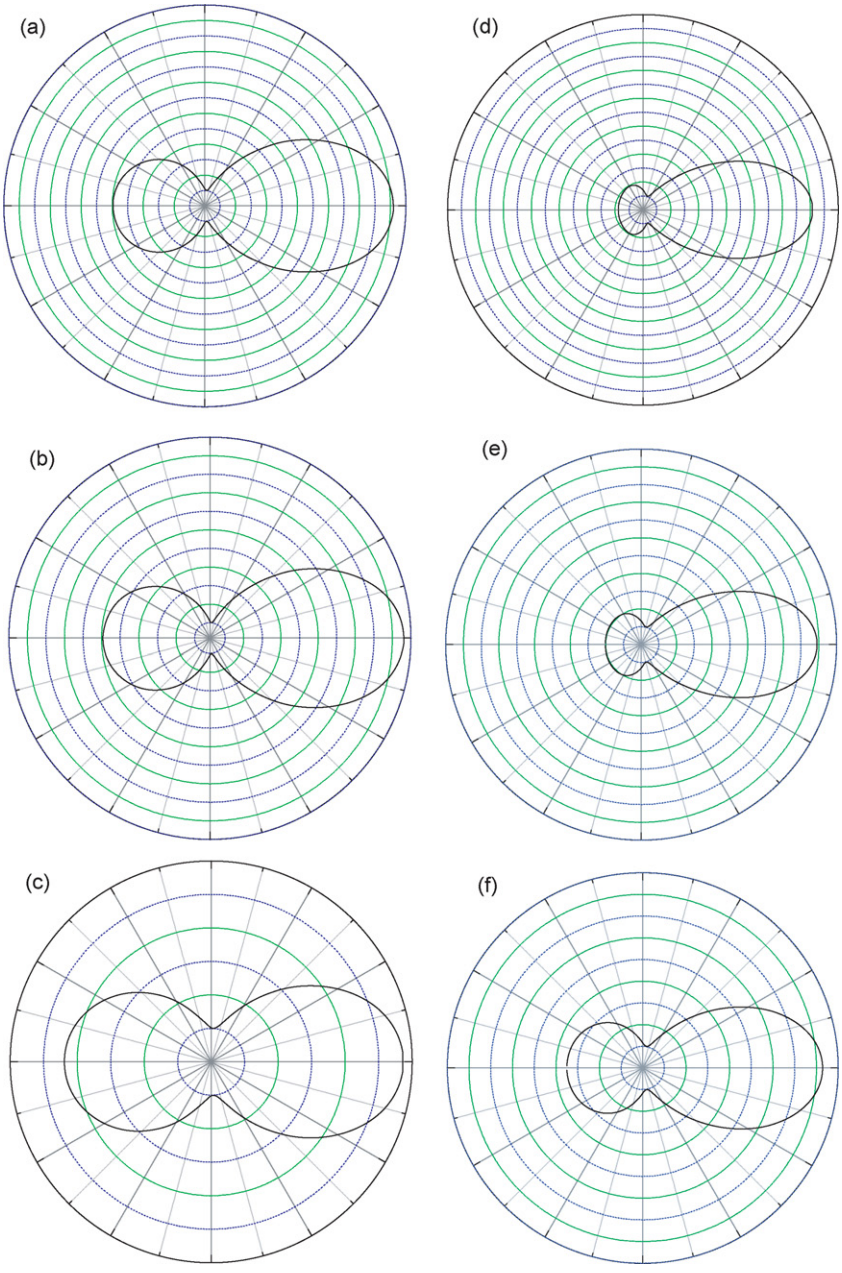

Fig. 4. Differential ionization yields $dY/d\Omega$ for one-cycle laser pulses with same parameters as in Fig. 3c, except $\hbar\omega = 0.52$, 0.6, 1.0 E_h for (a) and (d), (b) and (e), and (c) and (f), respectively. I_{max} is 2×10^{15} for (a)–(c) and 2×10^{16} W cm^{-2} for (d)–(f). Notations as in Fig. 2.

An extended search for enhancement of this type of symmetry breaking by a single-cycle laser pulse with phase $\eta = \pi$, similar to that shown in Fig. 3(c), but with varying photon energies, is documented in Fig. 4. Lower photon energies apparently favor symmetry breaking over higher photon energies, but the effect remains modest. This agrees with the scaling law discovered by Peng and Starace [18]. Note that the one-cycle pulse with phase $\eta = \pi$ cannot be extended to photon energies significantly smaller than $\hbar\omega = 0.52E_h$ (this limiting case is illustrated in Fig. 4(a)), because lower carrier frequencies produce populations of Rydberg states at the expense of photo-ionization. This suggests that one should use even shorter laser pulses in order to achieve the goal of uni-directional photo-ionization.

The scaling law [18] indicates that the effects of two- or multi-photon processes and, therefore, the effects of symmetry breaking, should increase as the 3/2 power of the maximum intensity I_{max}. Moreover, the asymmetries should scale as the parameter $\sqrt{I_{max}/\omega}$ (i.e., they should also increase with decreasing frequency), in accord with the trend which can be seen in Fig. 4(a)–(c). Thus, Fig. 4(a)–(f) compare the results for intensities 2×10^{15} and 2×10^{16} W cm^{-2}, for the same photon energies, $\hbar\omega = 0.52$, 0.6 and 1.0 E_h, respectively. As expected, the largest asymmetry is observed in Fig. 4(d) for the case with highest intensity $I_{max} = 2 \times 10^{16}$ W cm^{-2} and lowest carrier frequency $\omega = 0.52\, E_h/h$. These results may be considered as empirical confirmation of the scaling law [18] extended to the extreme situation of photon energy just above the ionization potential, and rather large intensity. Comparison of Figs. 4(d) and 2 reveals similar behaviors of dominant forward laser-induced electron re-scattering and Coulomb scattering, except for slightly larger widths of the re-scattering distributions.

We now seek a scenario that generates even narrower laser-induced differential ionization yields than those shown in Fig. 4(d). For this purpose we follow the advice deduced from Ref. [15], i.e., we choose an even shorter laser pulse. Accordingly, we design a half-cycle pulse consisting of a short, strong positive component (which has the same E_{max} and the same half-period $t_1 = \tau/2$ as the symmetry-breaking pulse that produces the flux shown in Fig. 3(c)), followed by a compensating long, weak negative component [see Eq. (17)]. The plot of $dY/d\Omega$ displayed in Fig. 3(d) demonstrates the desired uni-directional flux of photoelectrons in the forward direction (compare with Fig. 2 for Coulomb scattering). Contour plots of ρ_{ex} defined in Eq. (8) are exhibited in Fig. 5. These "snapshots" are quite surprising in that they appear to contradict our working hypothesis of a three-step mechanism for electron re-scattering. The dominant effect of the initial short, strong positive component of the half-cycle pulse is to create an "excited" component having a rather symmetric shape reminiscent of a p-orbital. This initially symmetric distribution, which reflects the dominance of single-photon ionization [18], is followed by a rapid, nearly symmetric dispersal of the wavepacket. The uni-directionality of the emitted flux of electrons becomes clear only after the passage of the long, weak component of the half-cycle pulse, i.e., the strong asymmetry, which is obvious from Fig. 3(d), is imposed by the second long, weak component of the half-cycle pulse, not by the first short, strong component. This finding may be regarded as an extension of the discovery of uni-directionality induced by two separate half-cycle pulses with opposite electric fields and equal intensity profiles [15] to non-equivalent ones.

In order to test the working hypothesis further, we plot in Fig. 6 the corresponding expectation value $\langle z(t)\rangle$. These confirm that initially the short, strong positive component of the half-cycle pulse drives the electron away from the nucleus, first in the negative z-direction and then back toward the nucleus, which it overshoots in the positive z-direction. Over this initial period the behavior of $\langle z(t)\rangle$ is in accord with our proposed three-step mechanism. Subsequently, however, the evolution in the forward direction is dominated by oscillations of the excited wavepacket in the Coulomb well, albeit with a systematic increase in $\langle z(t)\rangle$, which reflects the flux of electrons in the forward direction.

To shine additional light on the mechanism, we carried out complementary model simulations for an artificial half-cycle pulse consisting of the same short, strong positive component, but no compensating long, weak negative component (i.e., the latter is switched off; see Fig. 3(e)). We are aware that this artificial pulse does not fulfill the condition in Eq. (15), but it is nevertheless useful

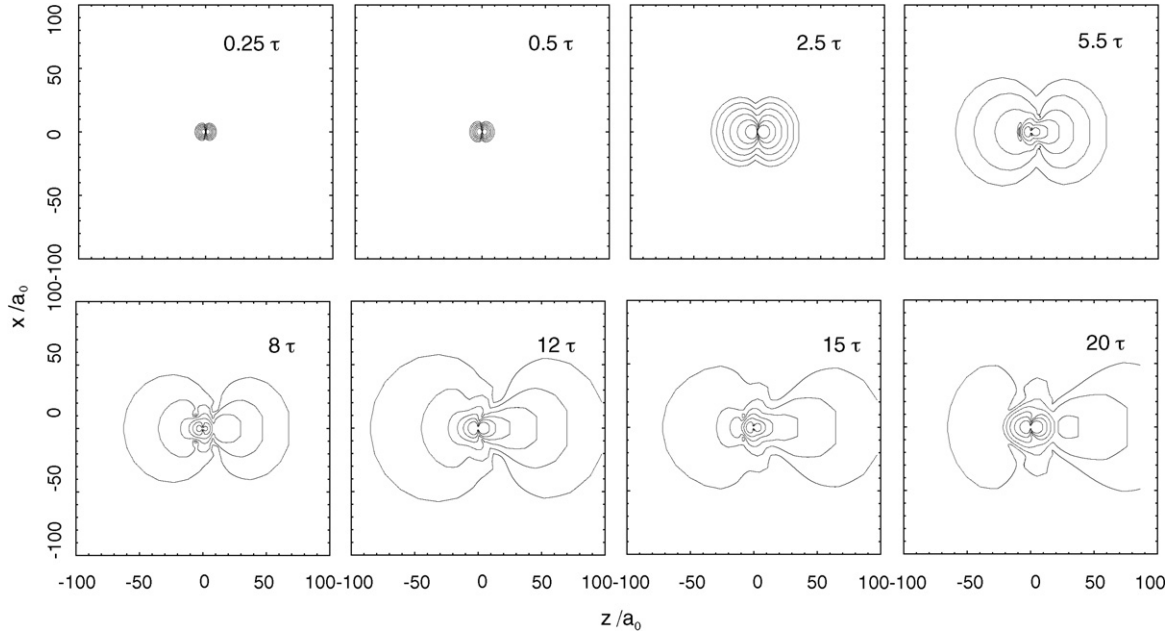

Fig. 5. Contour plots of probability density $\rho_{ex}(x, z)$ (see Eq. (8)) ($x = r\sin\theta, y = 0, z = r\cos\theta$) corresponding to pulse shown in Fig. 3(d). Contours are equidistant on a logarithmic scale.

for our present purpose. Fig. 3(e) shows that the strongly asymmetric forward re-scattering in the case of the full half-cycle pulse is replaced by much less asymmetric back scattering in the case of the single uni-directional component of the full half-cycle pulse. Again, this is in accord with dominant single-photon ionization and small contributions of two-photon processes, induced by the single short, strong component. This result may again be viewed as confirmation of the scaling law [18] applied empirically to a single component of a half-cycle pulse. As indicated in Fig. 6, the effects of the initial short, strong positive components are identical, but the oscillatory evolution of $\langle z(t)\rangle$ with a systematic increase observed for the compensated pulse is reversed for the artificial one! We conclude that uni-directional electron re-scattering by the half-cycle laser pulse (Eq. (17)) is the result of not only steps (i) through (iii) (due to the initial short, strong component) but also of a new step (iv) (due to the long, weak component), namely a gradual drift in the forward direction of the multiply re-scattering electron . It remains appropriate to call the overall mechanism, now consisting of steps (i)–(iv), "electron re-scattering", since step (iv) involves multiple re-scattering of the electron. The decisive role of step (iv) in uni-directionality is supported by Fig. 3(e), which shows the differential yield for the artificial pulse. Although the initial short, strong positive component of the half-cycle pulse is important, it does not per se dictate uni-directionality. We conclude that the net effect of the steps (i)–(iii) is to induce ionization, but step (iv) is necessary for uni-directionality.

In Fig. 7 we compare the dependence of the total cross-section σ for Coulomb scattering and the yield for photo-ionization Y on respectively the incident kinetic energy T and photon energy $\hbar\omega$, where we take the "frequency" of the half-cycle pulse (Eq. (17)) to be $\omega = 2\pi/\tau = \pi/t_1$. Although both σ (Fig. 7(a)) and Y (Fig. 7(b)) decrease with T and $\hbar\omega$, respectively, the causes of the decreases are different. In the case of Coulomb scattering, the faster the incident electron (i.e., the greater T is), the less time it has to interact with the nucleus and consequently the less deflection it suffers. In the case of laser-induced ionization, on the other hand, the greater ω, the less time the electric field acts upon the electron and hence the smaller the ionization yield.

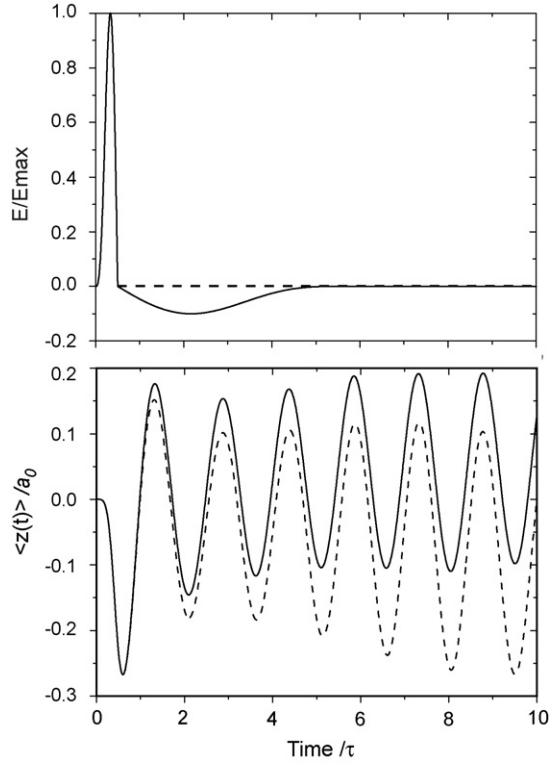

Fig. 6. Comparison of $\langle z(t)\rangle$ for half-cycle pulse in Fig. 3(d) (solid lines) and artificial half-cycle pulse in Fig. 3(e), without long, weak negative electric field component (dashed lines).

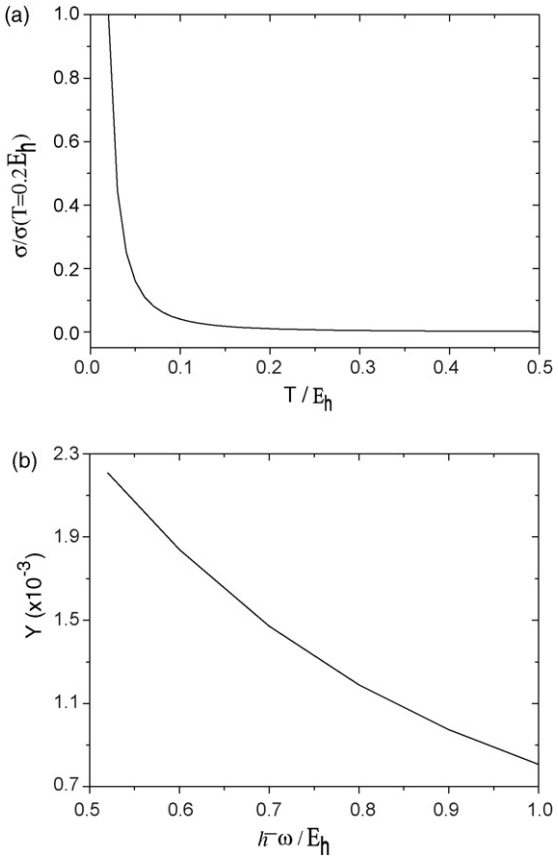

Fig. 7. Total (normalized) cross section for Coulomb scattering vs. incident kinetic energy T (a) (Eq. (2)) and total ionization yield vs. photon energy $\hbar\omega$ (b) achieved by half-cycle pulses with parameters as in Fig. 3(d) (except that $\hbar\omega$ varies).

4. Conclusions

The search for laser pulses that produce a uni-directional flux of photo-electrons, by analogy with ordinary Coulomb scattering, has led us to a rather extreme conclusion: the goal can be achieved approximately by use of pulses having very high intensities, low carrier frequencies, very short durations, and appropriate CEPs. A compelling example is shown in Fig. 4(d), for the case of the pulse parameters $I_{max} = 2 \times 10^{16}\,\mathrm{W\,cm^{-2}}$, $\hbar\omega = 0.52\,E_h$ and $t_p = 2\pi/\omega$, which corresponds to a single cycle. This result agrees with the scaling law of Peng and Starace [18], if we extrapolate to the limit of the frequency domain where a single photon can ionize H (i.e., $\hbar\omega > \mathrm{IP} = 0.5\,E_h$). We showed that a uni-directional flux of photo-electrons can be achieved even more efficiently by use of a less intense half-cycle pulse in the attosecond time domain. We take the half-cycle pulse to consist of a short, strong ($I_{max} = 2 \times 10^{15}\,\mathrm{W\,cm^{-2}}$) component followed by a weak, long one with opposite field strength, in order to satisfy the relation in Eq. (15). It differs from the sequence of two time-delayed half-cycle pulses having the same intensity profile but opposite field strengths, used by Hu and Starace [15]. One might expect that the short, strong component would dominate the long, weak "tail", but this intuitive notion is contradicted by the analysis of the resulting uni-directional ionization.

This surprising result suggests a four-step mechanism for electron re-scattering: (i) photo-ionization; (ii) turn-around of the electron; (iii) scattering of the returning electron from the atom's core; (iv) drift of the multiply re-scattering electron in the forward direction. The first three steps are induced by the initial short, strong positive electric field component of the half-cycle laser pulse. They have been anticipated by analogy with Corkum's three-step mechanism for HHG [4]. Their net effect is ionization, but not yet uni-directionality. The latter is achieved only by the rather unexpected fourth step, i.e., it is due to the compensating long, weak negative electric field component, which exerts a weak, but rather persistent, force that drives the "ionized" component of the wavepacket in the forward direction. The discovery of this effect is not easy, because it is hidden beneath the prominent superimposed phenomenon of multiple electron re-scatterings. In addition to processes (i)–(iv), we observe significant dispersion of the wavepacket, which does not diminish, however, the uni-directional flux of the emitted electrons in the forward direction. Nevertheless, it implies a less pronounced focusing of the re-scattered wavepacket than of the wavepacket that governs ordinary Coulomb scattering (compare Fig. 2 with Fig. 3(d)). In any case the present results, which show good agreement between Coulomb scattering and laser-driven electron re-scattering in the dominant forward direction, complement good agreement previously observed at rather low flux in the backward direction [24].

The analogy between ordinary Coulomb scattering and the laser-induced electron re-scattering that underlies uni-directional photo-ionization is quite limited. For example, in Coulomb scattering the electron begins its journey toward the nuclear core from a great (asymptotic) distance, whereas in "re-scattering" the electron that is eventually emitted starts on its path toward the core from a position very close to the core (i.e., where it turns around in step (ii)). Further, even though the total ionization yield decreases with increasing photon energy, in analogy with the decrease in the total cross-section for Coulomb scattering with increasing incident kinetic energy, the underlying causes of the fall-offs are different. We conclude that Coulomb scattering and laser-driven electron re-scattering are rather different processes, at least for the present domain of photon energies $\hbar\omega > \mathrm{IP}$. We therefore expect that the approaches to the imaging of electronic structures by means of electron diffraction [8,9] and orbital tomography [10,11] should yield complementary information. It remains to search for closer analogies in different domains of the laser-pulse parameters. For example, use of laser pulses with higher intensities and lower frequencies ($\hbar\omega < \mathrm{IP}$) should increase ionization yield and lead to greater excursions of the electron from the core, in which case the re-scattering would be more like ordinary Coulomb scattering.

The results of the present study should stimulate additional investigations of uni-directional electron re-scattering in more complex atoms and molecules. Furthermore, they suggest that it would be useful to search for mechanisms of HHG where Corkum's three-step scheme may need to be extended. Last, but not least, the results call for experimental applications. It is encouraging that the production of attosecond one-cycle laser pulses is feasible [7,21]. But the preparation of attosecond *half-cycle* pulses presents a challenge.

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