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Energy bunching in soft recollisions revealed with long-wavelength few-cycle pulses

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Abstract
Soft recollisions are laser-driven distant collisions of an electron with its parent ion. Such collisions may cause an energy bunching, since electrons with different initial drift momenta can acquire impulses which exactly counterbalance these differences. The bunching generates a series of peaks in the photo-electron spectrum. We will show that this series could be uncovered peak by peak experimentally by means of phase-stabilized few-cycle pulses with increasing duration.

(Some figures may appear in colour only in the online journal)

1. Introduction

In addition to high-energy phenomena such as high-harmonic generation and above-threshold ionization, the interaction of intense long-wavelength (a few μm) laser pulses with atoms also leads to an interesting low-energy structure (LES) in the photo-electron spectrum close to threshold (Blaga *et al* 2009), which could be reproduced with numerical spectra, quantum mechanically (Blaga *et al* 2009) as well as classically (Quan *et al* 2009, Liu and Hatsagortsyan 2010, Lemell *et al* 2012). Recently, soft recollisions, where the electron turns around beside the ion, have been uncovered as the origin of the LES with the analytical prediction that a series of low-energy peaks should exist (Kästner *et al* 2012). In addition, the number of peaks increases with the number of laser cycles contained in the full pulse. However, those peaks are difficult to be accessed experimentally since they sit on top of a large background signal which strongly varies at low energies towards threshold. Moreover, there may be in addition low-energy features depending specifically on individual potential properties.

These difficulties could be overcome if a few-cycle pulse is used for the measurement. In addition, such a pulse should also resolve the generation of the LES peak by peak with successively increasing pulse length. However, using ultra-short pulses will shift the peak positions compared to the analytically determined ones. Moreover, the carrier-envelope phase (CEP), i.e. the phase difference between the maximum of a cycle and the maximum of the envelope (Paulus *et al* 2001, Baltuska *et al* 2003) becomes relevant for the peak positions. For these reasons, we provide in the following numerical photo-electron spectra for such pulses as a guideline for future experiments.

2. Effect of soft recollisions on long-wavelength photo-electron spectra

Before addressing few-cycle pulses directly, we briefly review the origin of the series of peaks (Kästner *et al* 2012) which we have identified as the LES and discuss how these peaks add to the other characteristics of the photo-electron spectrum generated with intense pulses. Typically, upon interaction with a strong oscillating laser field $F(t) = F_0 \cos(\omega t)$, characterized by the peak amplitude F_0 and frequency ω, the bound electrons leave the ion through tunnelling ionization at different phases $\varphi' = \omega t'$ of the laser field[3] with a momentum $p_{\mathrm{direct}}(\varphi') = (F_0/\omega)\sin\varphi' + \Delta p$. Hereby Δp, which accounts

[3] We denote all initial variables with a single prime and all final quantities with a double prime.

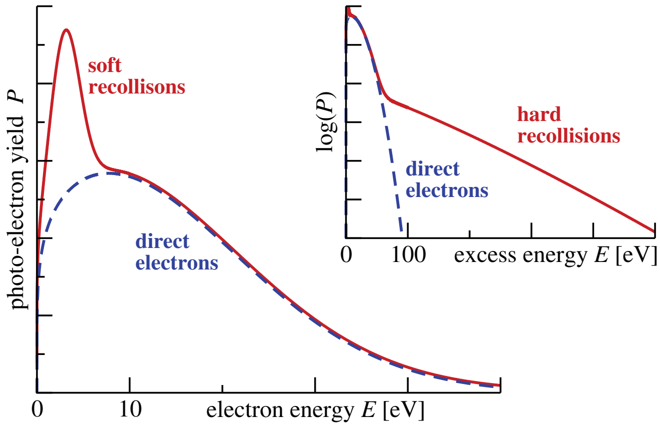

Figure 1. Schematic photo-electron spectrum $P(E)$ of atoms in intense long-wavelength laser pulses. The blue dashed line indicates direct photo-ionization, and the solid red line shows the spectrum including recollisions. At low energies, soft recollisions induce a pronounced structure, the so-called LES, which is shown on a linear scale (left graph). Hard recollisions extend the spectrum to higher energies, which becomes visible only on a logarithmic scale (right).

for the Coulomb attraction during release, depends negligibly on φ'. All these events are well described by the strong-field approximation (which usually assumes a zero-range potential with $\Delta p = 0$), which essentially states that the electrons reach the detector *directly* with the unchanged initial momentum $p_{\text{direct}}(\varphi')$. In connection with the φ'-dependent tunnelling probabilities, this leads to the typical well-known shape of the spectrum, see the blue dashed line in figure 1. For specific initial laser phases φ', the electron returns to the nucleus executing a hard recollision by hitting the nucleus head-on. Thereby the electron can acquire an energy up to $10E_{\text{pond}}$ with the ponderomotive energy $E_{\text{pond}} = F_0{}^2/(2\omega)^2$ which leads to a high-energy component in the photo-electron spectrum albeit of exponentially small magnitude as shown in the right panel of figure 1. This phenomenon is known as the plateau in above-threshold ionization (Paulus *et al* 1994).

Complementarily, an electron released around a different specific phase φ' of the laser can be driven aside the nucleus with *minimal* kinetic energy in the vicinity of the nucleus. This amounts to a low-energy collision in contrast to the high-energy recollision discussed before and the peaks arising from this soft recollision appear at very low energy in the photo-electron spectrum, see the left graph of figure 1. The peaks appear since the soft recollision has a focusing effect on the final electron energy which we have called electron-energy bunching (Kästner *et al* 2012). The soft recollision can be thought of giving the laser-driven electron a small impulse δp through the interaction with the ion potential changing the initial momentum $p_{\text{final}} = p_{\text{direct}}(\varphi') + \delta p(\varphi')$. The impulse depends through the turning point on the strong-field trajectory and thereby on the initial phase φ'. If the impulse δp exactly compensates the change of the direct momentum, i.e. $\mathrm{d}(p_{\text{direct}} + \delta p)/\mathrm{d}\varphi' = 0$, a low-energy peak in the photo-electron spectrum emerges. Since soft recollisions occur at times $t_k = (k+1/2)T$, with $T = 2\pi/\omega$ the period of the laser, as shown for $k = 1, 2$ in figure 2, a series of peaks can emerge provided the laser pulse is long enough. Obviously, the peak positions are given approximately by (Kästner *et al* 2012)

$$p_k = \frac{F_0}{\omega}\frac{2}{(2k+1)\pi} \tag{1}$$

and the corresponding energies read

$$E_k = \frac{8}{(2k+1)^2\pi^2}E_{\text{pond}}. \tag{2}$$

The experimentally observed peaks (Blaga *et al* 2009) correspond to $k = 1$, i.e. $E \approx E_{\text{pond}}/10$. As we will see below, a pulse envelope can shift the peak position with respect to equation (2), where an infinite pulse was assumed.

3. Computation of the photo-electron dynamics

3.1. Hamiltonian

To obtain a photo-electron spectrum from illumination of atoms with few-cycle pulses, we classically propagate electrons in the field of a laser pulse, linearly polarized along $\hat{z}$, and in the ionic Coulomb potential, after the electrons have been released according to a tunnel probability, described below. The Hamiltonian of the electron in cylindrical coordinates ρ and z reads (atomic units are used unless stated otherwise)

$$H = \frac{p_\rho^2}{2} + \frac{p_z^2}{2} - \frac{1}{\sqrt{\rho^2+z^2}} + zF(t). \tag{3}$$

The time evolution of the few-cycle pulse $F(t)$ with a Gaussian envelope is defined through the vector potential $A(t)$:

$$F(t) = -\frac{\partial}{\partial t}A(t) \tag{4a}$$

$$A(t) = -A_0\exp(-2\ln 2(t/nT)^2)\sin(\omega t + \phi), \tag{4b}$$

with $A_0 = F_0/\omega$ being the amplitude of the vector potential, $T = 2\pi/\omega$ the laser period, n the number of cycles and ϕ the CEP. Expression (4) guarantees $\int \mathrm{d}t\, F(t) = 0$.

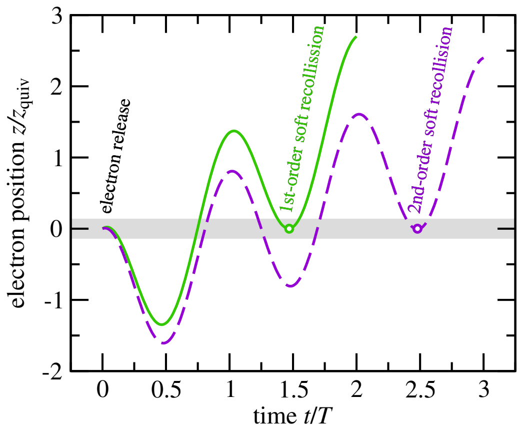

Figure 2. Sketch of a first-order (solid green line) and second-order (dashed violet line) soft recollision. Shown is the electron position z along the laser polarization axis in units of the quiver amplitude $z_{\rm quiv} = F_0/\omega^2$ as a function of time t in units of the laser period T for an electric field $F(t) = F_0 \cos(\omega t)$. Note that the recollisions occur at finite ρ.

3.2. Initial conditions for trajectories

The probability for tunnelling at time t', where $F' = |F(t')|$, with a transverse momentum of $p'_\rho = p_\rho(t')$ is given by (Shvetsov-Shilovski *et al* 2009)

$$W(F', p'_\rho) \propto \frac{1}{F'^2} \exp(-2(2E_{\rm ip} + p'^2_\rho)^{3/2}/3F') \times \frac{p'_\rho}{\sqrt{1 + p'^2_\rho/2E_{\rm ip}}}, \quad (5)$$

with an additional Jacobian factor p'_ρ. $E_{\rm ip}$ is the atomic ionization potential. In principle, one could uniformly sample possible initial conditions for t' and p'_ρ. This, however, would be extremely inefficient due to the exponential weight in equation (5). Most of the propagated trajectories would barely contribute due to their tiny weight. It is more efficient to propagate trajectories with the same weight. In order to do so, we define a grid of initial conditions with grid positions of $t'_l = l \times \delta t$, where $|l| \leqslant L/2$ in time and $p'_m = m \times \delta p$, where $0 \leqslant m \leqslant M$ in momentum. The resulting $L \times M$ boxes in the (p'_ρ, t') plane are now connected by some (arbitrary) path. With the definition

$$X_j \equiv \sum_{i=1}^{j}{}' W(|F(l_i \times \delta t)|, m_i \times \delta p), \quad (6)$$

where the prime indicates that the sum is calculated along the pre-defined path, we obtain the probability

$$P_j = X_j / X_{L\times M}, \quad (7)$$

which is a monotonic function between 0 and 1 in terms of the index j. Now we pick a uniform random number between 0 and 1, which corresponds by means of equation (7) to some j and thus (again using the pre-defined path through the initial conditions) to $t' = l_j \times \delta t$ and $p'_\rho = m_j \times \delta p$. For the results presented below, we have used $L = {\rm int}(6nT/\delta t)$, $M = 1000$, $\delta t = 0.5$ au and $\delta p = 0.001$ au.

Electrons are propagated until $t = 3nT$, where the laser field is sufficiently weak to be neglected. With the position and the momentum at this time one can easily calculate the asymptotic momenta p''_z and p''_ρ.

4. Photo-electron spectra from few-cycle pulses

In the following, we present calculations for argon atoms ($E_{\rm ip}$ = 0.5792 au) and use laser pulses with a wavelength of $\lambda = 2\ \mu$m (ω = 0.0228 au) and an intensity $I = 10^{14}$ W cm^{-2} (F_0 = 0.0534 au, A_0 = 2.342 au). Thus, the ponderomotive energy is $E_{\rm pond}$ = 37.3 eV and the quiver amplitude $z_{\rm quiv}$ = 54.3 Å. Figure 3 shows photo-electron spectra for pulses with an increasing number n of cycles and two CEPs ϕ for each pulse length. The figure shows spectra obtained from averaging over intensities in a laser beam with a Gaussian focus (Augst *et al* 1991). Non-averaged spectra for the corresponding peak intensities are qualitatively the same, but show sharper structures at slightly higher energies.

Not surprisingly, the most dramatic dependence on ϕ is seen for $n = 1/2$ in figures 3(a) and (b). The electron emission is preferentially in the upward direction ($p''_z > 0$). Although this occurs for both CEPs, the reason behind it is different in both cases. The highest probability for tunnelling is at the maxima of the electric field $F(t)$, cf the green shaded regions in the insets of figure 3. For $\phi = 0$, there is one global maximum at $t_{\max} = 0$. For a strong-field trajectory

$$z(t, t') = z' - A(t')[t - t'] + \int_{t'}^{t} {\rm d}\tau\, A(\tau) \quad (8a)$$

$$p_z(t, t') = A(t) - A(t'), \quad (8b)$$

the drift momentum is given by $-A(t')$. Thus for $n = 1/2$ and $\phi = 0$ one would expect an up-down symmetry since $A(t')$ is positive and negative for $t' \approx 0$. However, electrons, which are released towards negative z (due to the negative electron charge a positive field corresponds to a force pointing in the negative direction) acquire an impulse Δp from the Coulomb potential which generates a positive offset for the drift momentum. Therefore upward-emitted electrons are in the majority. This holds for all pulse lengths (figures 3(a), (c) and (e)), whereby the difference gets weaker for longer pulses. For a CEP of $\phi = \pi/2$, the energy distribution is shifted towards higher energies, see figure 3(b). Here the electric field has two maxima at $t_{\max} \approx \pm 0.174T$. Since the vector potential is negative for both times, namely $A(t_{\max}) \approx -0.389A_0$, most of the electrons start with a positive drift momentum and the electrons are driven upwards. The maximum of the distribution is expected around $E \approx 0.302E_{\rm pond} \approx 11.3$ eV, i.e. outside the region shown in figure 3(b). Again the asymmetry[4] seen for the shortest pulse holds also for the longer pulses (figures 3(d) and (f)), but becomes less and less distinct.

Most important, for the recollisions we are interested in, is that the spectra for $n = 1/2$ are featureless at small energies. This changes with $n = 1$ where a low-energy peak shows up around $E \approx 1$ eV, see figures 3(c) and (d). Apparently, now the pulse is sufficiently long, that the electron can be driven back at about 3/2 cycles after its release. Figure 2 shows a typical trajectory (solid green line), which is released at the maximum of the field ($t \approx 0$) and returns to the nuclei, which we call

[4] The CEP dependence of this asymmetry has recently been measured (Bergues *et al* 2011).

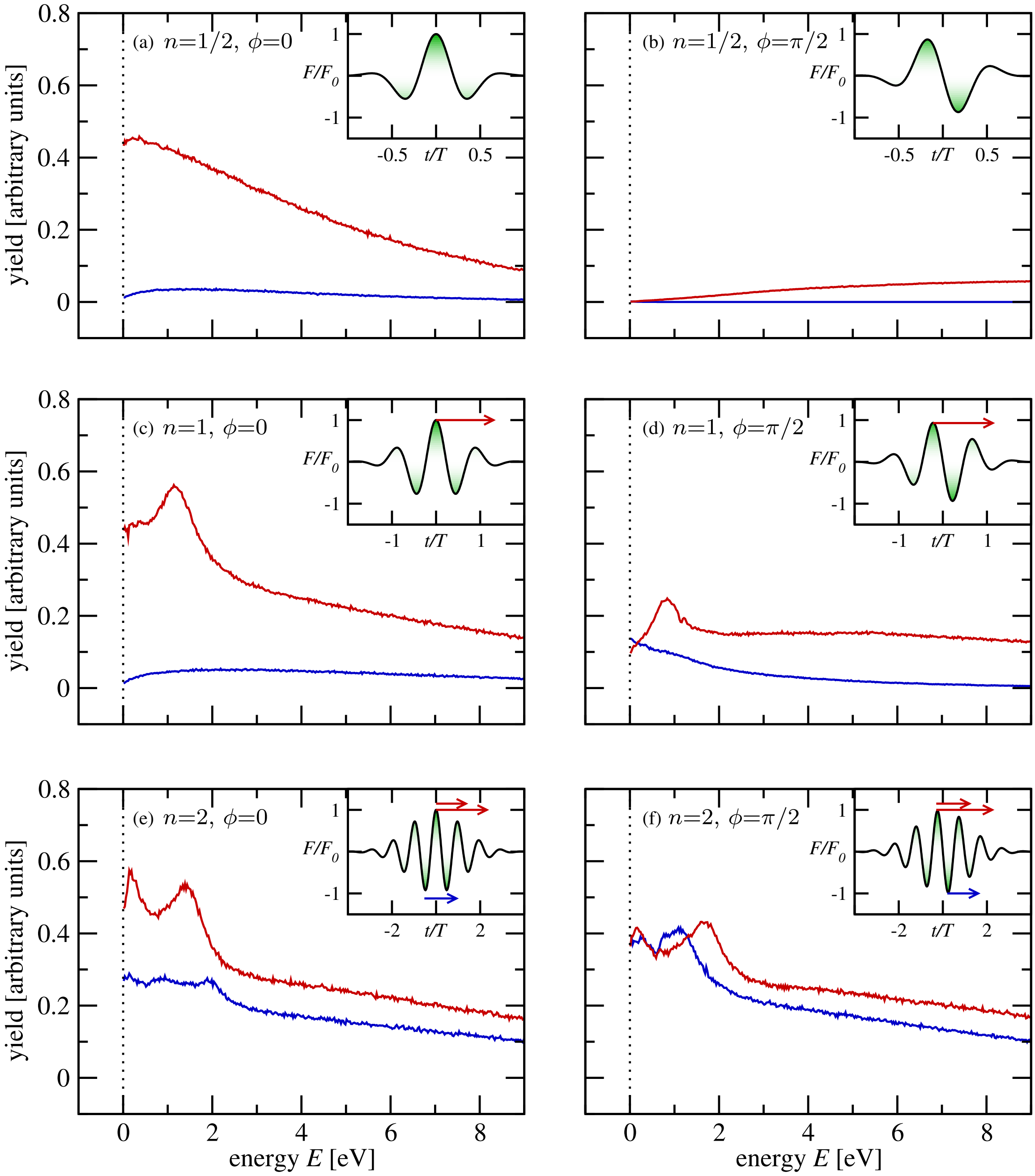

Figure 3. Photo-electron spectra for energies $E = p_\rho''^2/2 + p_z''^2/2$ of electrons detected upwards $\theta'' < 5^\circ$ (red line) and downwards $\theta'' > 175^\circ$ (blue line) with $\tan(\theta'') \equiv p_\rho''/p_z''$. Spectra are shown for pulses with $n = 1/2$, 1 and 2 cycles (top to bottom row) and two CEPs $\phi = 0$ (left column) and $\phi = \pi/2$ (right column), corresponding to sin-like and cos-like pulses, respectively. The insets show the time-dependent electric field $F(t)$. The arrows indicate the time span from tunnelling to recollision. Each spectrum has been obtained by averaging over a Gaussian laser focus.

a *first-order soft recollision*. Such trajectories are started at $t \approx 0$ for $\phi = 0$ and at $t \approx -T/4$ for $\phi = \pi/2$. The time span for the laser-driven dynamics from release to recollision is marked by arrows (with the color marking the direction of tunnelling) in the insets of figure 3. Since for few-cycle pulses the electric field during the dynamics depends on ϕ, the peak positions for $\phi = 0$ and $\phi = \pi/2$ are different. This shows that the expression in equation (2) should be used with care when applied to ultra-short pulses. In any case it gives a good estimate for the peak locations. A further increase of the pulse duration shows for $n = 2$ and $\phi = 0$ (figure 3(e)) a different spectrum. For the first time one observes a single peak at $E \approx 2$ eV for the downward-emitted electrons as well as a double peak in the upward direction. The downward feature is much more pronounced for $\phi = \pi/2$ as can be seen in figure 3(f). This is due to the higher ionization probability for electrons, which are released at $t \approx T/4$ as shown by the blue arrow. The second upward peak at low energies is due to a *second-order soft recollision*. This process is indicated by the longer red arrow, which shows that the electron returns

Table 1. Peak energies E_k as obtained with equation (9). All energies are given in eV. For positive final momenta ($p''_z > 0$) the initial times are $t' = 0$ and $t' = -T/4$ for $\phi = 0$ and $\phi = \pi/2$, respectively. For negative final momenta ($p''_z < 0$) the initial times are $t' = -T/2$ and $t' = T/4$ for $\phi = 0$ and $\phi = \pi/2$, respectively.

Number of cycles	Order of LES	$\phi = 0$		$\phi = \pi/2$	
		$p''_z > 0$	$p''_z < 0$	$p''_z > 0$	$p''_z < 0$
$n = 1$	$k = 1$	1.06		0.97	
$n = 2$	$k = 1$	1.82	2.26	2.09	1.49
	$k = 2$	0.38		0.41	0.24

to the ion only after about 5/2 cycles (cf also the dashed violet line in figure 2). Our numerical calculations yield for the second-order peak energies of $E \approx 0.2$ eV. Such small energies make it challenging to observe them experimentally.

The position of the calculated LES peaks can be estimated by the condition for a soft recollision at time t_k at which $p_z(t_k, t') = 0$, cf figure 2. For infinite monochromatic fields, this leads to the simple expression in equation (1). For few-cycle pulses, however, the corresponding momenta are given by the integral

$$p_k \approx \frac{1}{t_k - t'} \int_{t'}^{t_k} \mathrm{d}\tau\, A(\tau), \tag{9}$$

which follows from equation (8*a*) by assuming $z' = 0$ and replacing $A(t') \to p_k$. With $A(\tau)$ from equation (4*b*) these integrals can be solved analytically but result in lengthy expressions. Instead of presenting them we list the corresponding values for the peak energies $E_k = {p_k}^2/2$ in table 1. For the initial time t', we have chosen multiples of $T/4$ near which the electric field has a maximum, see table 1 for the corresponding values. The recollision times are $t_k = t' + (k + 1/2)T$. Although figure 3 shows focus-averaged spectra, the values given in the table agree surprisingly well the location of the LES peaks in the figure. This agreement further supports our assignments of the LES structures to first- and second-order recollision as indicated by the arrows in the insets of figure 3.

5. Summary

We have investigated theoretically low-energy photo-electron spectra generated by few-cycle long-wavelength laser pulses using a classical model. A gradual increase of the pulse length supports the interpretation that the LES is due to soft recollisions with the ion. Such recollisions can occur at later instances after the tunnelling, rendering the LES a series of peaks. However, these peaks can only emerge if the pulse has a sufficient number of cycles. For a Gaussian half-cycle pulse, we predict that the LES vanishes. Apart from the pulse length the photo-electron spectra also depend on the CEP. An experimental realization of the proposed scenario should be feasible since CEP stabilization has been recently extended to the μm-wavelength regime (Schmidt *et al* 2011, Bergues *et al* 2011).

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