# Pulse-shape effects in ionization of atomic hydrogen by short-pulse XUV intense laser radiation: A sensitivity study

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Phys. Rev. A **91**, 053404 — Published 8 May 2015

DOI: 10.1103/PhysRevA.91.053404

# Pulse-Shape Effects in Ionization of Atomic Hydrogen by Short-Pulse XUV Intense Laser Radiation: A Sensitivity Study

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(Dated: April 21, 2015)

The displacement effect studied in a recent paper (Ivanov *et al.*, Phys. Rev. A **90** (2014) 043401) in atomic ionization by a short XUV pulse is investigated in more detail. It is shown that achieving a significant displacement critically depends on the assumption of a plateau in the envelope function of the electric field, and that the ramp-on is fine-tuned in such a way that a drift velocity generated during the ramp-on phase can increase this displacement further. Seemingly minor variations in the electric fields defined in slightly different ways cause significant changes in the final results, in particular regarding the angular-momentum distribution of the ejected electron. In light of such a strong sensitivity seen in the predictions made with idealized pulse shapes and the likely difficulties of preparing such pulses experimentally, an experimental realization of the displacement effect will likely be a major challenge.

PACS numbers: 32.80.Rm, 32.80.Fb, 42.50.Hz, 32.90.+a

## I. INTRODUCTION

In a recent paper [1], strong-field ionization of atomic hydrogen as well as lithium driven by a short extreme ultraviolet (XUV) pulse was studied. The key results of the paper were some peculiar effects in the angular-momentum distribution of the ejected electron, provided the pulse alone caused a significant non-zero displacement of the electron without it leaving the laser focus. The displacement is defined as the time integral of the pulse vector potential taken over the pulse duration, i.e., essentially the second integral over time of the electric field. These effects should be visible in the photoelectron angular distribution (PAD).

As noted already in the above paper, an important issue concerns the occurrence of pulses with a nonzero displacement experimentally. While half-cycle and single-cycle pulses with non-zero displacement have been observed [2, 3], and in some cases such pulses are predicted to even deliver nonzero momentum to a free electron [4], further attempts to identify the origin of the effect seemed a worthwhile effort. We emphasize that several theoretical papers (see, for example, [5, 6]) strongly favor pulses with a vanishing displacement. However, the condition is not of practical importance for very short pulses, for which the displacement would be small relative to the focusing region [7].

In the present paper, we investigate in detail the origin of the displacement, as it applies to the waveform that we (and many other authors in this field) have been using. We begin by discussing the advantages of choosing a particular (sine-squared) envelope function for the ramp-on and ramp-off parts of the pulse, compared to other common choices such as Gaussian or linear ramp-on/off. We then investigate the potential role of a plateau, i.e., a constant value of the envelope function, between the ramp-on and ramp-off phases.

Most importantly, we will discuss the differences resulting from either setting the electric field or the vector potential of the pulse first via a particular envelope function and then calculating the respective second one from the field defined first, i.e., by either integrating the electric field over time or by differentiating the vector potential with respect to time. The resulting electric fields will be (slightly) *different* in the two scenarios. Obtaining different results, therefore, does not violate the requirement of gauge-invariance, i.e., the need to predict the same observable quantities in the length and velocity gauges of the dipole operator. For this gauge invariance to hold, only the two fields generated directly from each other must yield the same answer. This is, indeed, the case for all calculations presented in this work.

Unless indicated otherwise, atomic units will be used throughout this paper.

## II. PULSE SHAPES

The fields studied in the present paper are of the general form

$$\boldsymbol{S}(t) = f(t)\, S_0 \sin(\omega t + \phi)\hat{\boldsymbol{z}}, \tag{1}$$

where $f(t)$ is the envelope function, $\omega$ is the central angular frequency, $\phi$ is the carrier-envelope phase (CEP), $\boldsymbol{S}(t)$ is either the electric field $\boldsymbol{E}(t)$ or the vector potential $\boldsymbol{A}(t)$, and $S_0$ is the corresponding amplitude. We assume linearly polarized transversal fields with a component only along the $\hat{\boldsymbol{z}}$ direction, as well as the dipole approximation, i.e., there is no spatial dependence in the field that needs to be accounted for.

A popular shape of the envelope function is the generalized form:

$$f(t) = \begin{cases} \sin^2\left(\frac{\pi t}{2n_1 T}\right), & 0 \le t \le n_1 T; \\ 1, & n_1 T \le t \le (n_1+n_2)\,T; \\ 1-\sin^2\left(\frac{\pi t}{2n_3 T}\right), & (n_1+n_2)\,T \le t \le t_f; \\ 0, & \text{otherwise}. \end{cases} \tag{2}$$

Here $T = 2\pi/\omega$ is the period for one cycle of the central frequency, while $n_1$, $n_2$, and $n_3$ denote the number of cycles in the ramp-on, plateau, and ramp-off phases of the pulse, respectively. We start the $n_1$-$n_2$-$n_3$ pulse at the initial time $t_i = 0$, and the field is zero again at the final time $t_f = (n_1 + n_2 + n_3)\,T$. As will be shown below, $n_1$, $n_2$, and $n_3$ do not necessarily have to be integer numbers, although some restrictions apply in order to ensure a physical pulse that can propagate according to Maxwell's classical equations for electromagnetic waves in vacuum. For the purpose of the present work, we will restrict ourselves to cases with $n_1 = n_3$, but we will allow $n_2 \neq 0$.

We begin by recalling the relationship between the electric field $\boldsymbol{E}(t)$ and the vector potential $\boldsymbol{A}(t)$. The former is used in the length gauge of the electric dipole operator while the latter is employed in the velocity gauge. The two fields are related by

$$\boldsymbol{E}(t) = -\frac{1}{c}\,\frac{d}{dt}\boldsymbol{A}(t) \tag{3}$$

and

$$\boldsymbol{A}(t) = -c\int_0^t \boldsymbol{E}(t')dt', \tag{4}$$

where $c$ is the speed of light in vacuum. For infinitely long sinusoidal "pulses", i.e., those used in standard treatments of weak-field photoionization processes, one can still start by setting $\boldsymbol{A}(t)$ as schematically outlined in Eq. (1) with the envelope of Eq. (2) and then obtaining $\boldsymbol{E}(t)$ from Eq. (3) before performing the calculation in the length gauge. The only differences would be a factor $1/\omega$ and an extra phase of $\pi/2$ (90°) in $\boldsymbol{A}(t)$ relative to $\boldsymbol{E}(t)$. With those adjustments made, the predictions should be identical, i.e., gauge-invariant for cases such as atomic hydrogen, where the orbitals are eigenfunctions of the corresponding field-free hamiltonian. For pulses of finite length, the results will still be very similar, provided the effect of the additional time derivative of the envelope function during the ramp-on/off phases is effectively negligible.

Another important aspect of choosing suitable pulses concerns the boundary conditions at the beginning and the end of the pulse. In any realistic scenario, the electric field should vanish before and after the pulse, unless one wants to introduce some overlaying direct current (DC) field to further stir the ejected electrons. The immediate consequence of a vanishing electric field is the fact that a pure Gaussian ramp-on/off is not realistic. At the very least, one would require some smooth correction on the edges of the pulse envelope, or be prepared to treat such a long pulse that this condition is sufficiently well fulfilled to avoid any numerical artifacts by jumping from zero to non-zero values and back.

While the above problem can easily be avoided by employing sine-squared or even linear ramp-on/off functions, another condition concerns the time integral of the electric field, which is proportional to the vector potential when the pulse is over. For a pulse to fulfill Maxwell's classical equations for electromagnetic waves, this integral needs to vanish as well. According to Madsen [5], in theoretical calculations this condition should be taken "literally". As a consequence, we cannot choose arbitrary values for $n_1$, $n_2$, $n_3$, and $\phi$ if the electric field is defined as outlined above. It is straightforward to construct unphysical cases where the integral would not vanish.

The latter problem, in turn, can be avoided by starting with the vector potential $\boldsymbol{A}(t)$ instead of the electric field $\boldsymbol{E}(t)$. In that case, however, care has to be taken that the latter approaches its zero value smoothly at $t_i$ and $t_f$. This is not necessarily the case, for example, when a linear ramp-on/off envelope is chosen for $\boldsymbol{A}(t)$, with the details depending on the CEP.

Finally, we consider the displacement, i.e., the first time integral of the vector potential or the second time integral of the electric field. As shown below, large nonzero displacements can be constructed by defining the electric field with a long plateau in the envelope function. On the other hand, if the vector potential is set via Eqs. (1,2), even the plateau will not generate a significant displacement.

Figure 1 exhibits a few examples of the situation described so far. Specifically, we assume a few-cycle linearly polarized laser field of peak intensity $10^{14}$ W/cm$^2$ (i.e., maximum field amplitude 0.05338 a.u.) with a central frequency of 0.70 a.u. Except for the length of the plateau, which was reduced from 36 optical cycles (o.c.) to 5.5 o.c. or 6.0 o.c., such pulses were discussed in the earlier work [1]. The principal reason for reducing the length of the plateau in the present illustration is to limit the displacement shown in some of the panels and hence improve the visibility.

The left column of Fig. 1 shows cases in which the electric field $\boldsymbol{E}$ is defined through Eqs. (1,2), while the right column exhibits the corresponding cases for setting $\boldsymbol{A}$ this way. On first sight the electric fields and the vector potentials in each row might be mistaken as being the same. They are, indeed, very similar, but they differ due to the way either the electric field or the vector potential is obtained from its respective counterpart during the ramp-on/off phases.

We start our discussion with the left column of Fig. 1, i.e., cases in which we define the electric field $\boldsymbol{E}$ via Eqs. (1,2). Each panel exhibits the $z$-component of $\boldsymbol{E}$ and that of the vector potential $\boldsymbol{A}$ (obtained from the time integral of $\boldsymbol{E}$), as well as the classical displacement

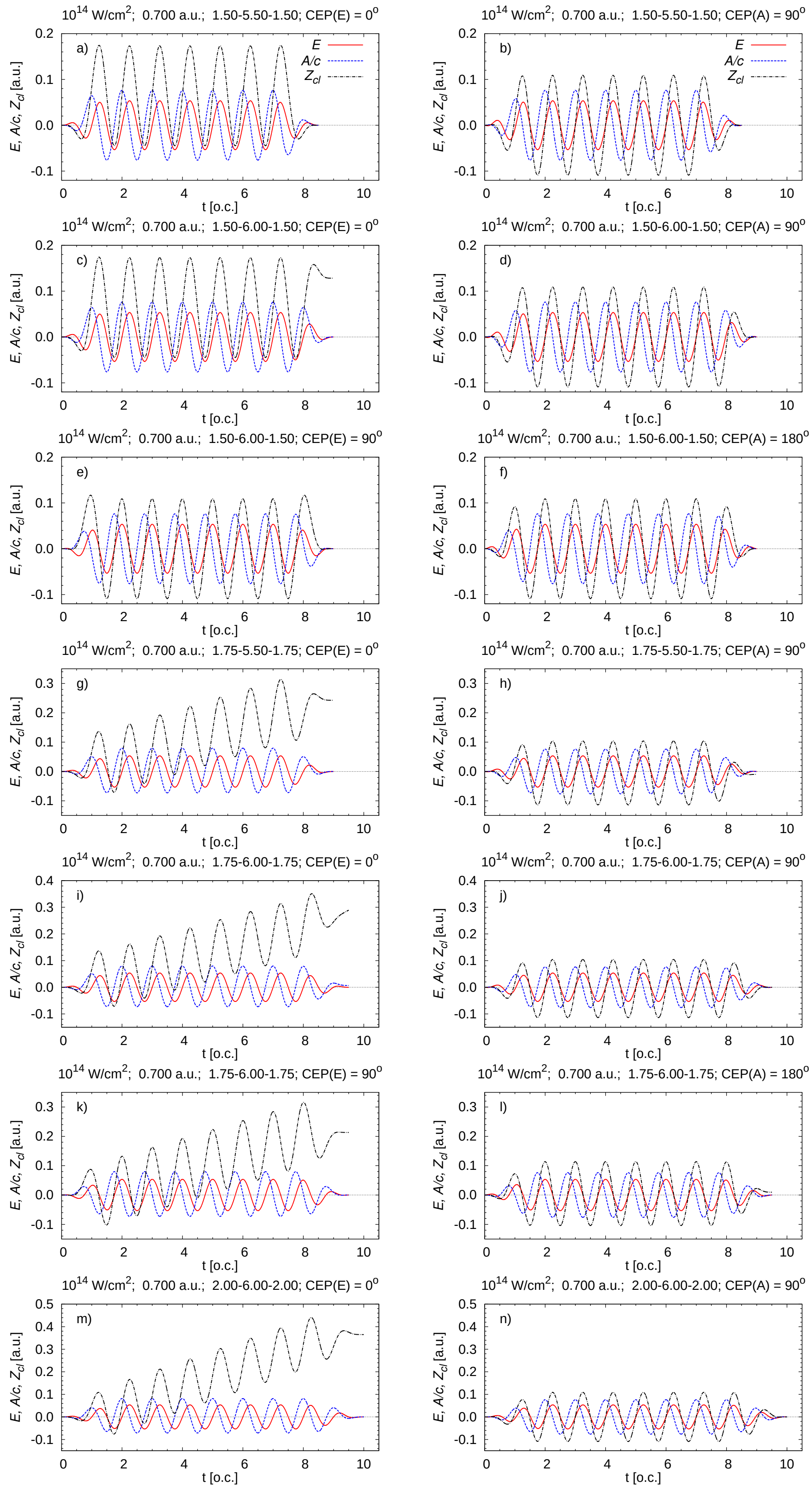


FIG. 1: $z$-components of the electric field $\boldsymbol{E}$ and the vector potential $\boldsymbol{A}/c$, and the displacement $Z_{cl}$ for a linearly polarized laser field of peak intensity $10^{14}$ W/cm$^2$ with a central frequency of 0.70 a.u. The header of each panel indicates the number of ramp-on–plateau–ramp-off cycles. In the left panels, the electric field is set as outlined in the text for the CEP indicated, while the vector potential and the displacement are obtained by integrating the field once or twice, respectively. In the right panels, we start with the vector potential instead. The field and the displacement are then obtained by differentiating with respect to or integrating over time (once).

(i.e., the time integral of the linear momentum associated with the $\boldsymbol{A}$-field). This displacement is defined as

$$Z_{cl}(t) = \int_0^t A(t')\, dt'. \tag{5}$$

The header of each panel indicates the number of ramp-on–plateau–ramp-off cycles, as well as the CEP. Starting with panels a), c), and e), which are all for 1.5 o.c. for the ramp-on/off parts, we see that changing the length of the plateau from 5.5 o.c. in panel a) to 6.0 o.c. in panel c) creates a non-zero displacement. That displacement, however, vanishes again if the CEP is changed from $0^\circ$ to $90^\circ$ in panel e).

Moving on to panels g), i), and k), each of which are for 1.75 o.c. for the ramp-on/off parts, we see a non-zero displacement for all three cases. Note, however, that panel i) represents a pulse with non-zero value of the vector potential at the end. This is difficult to see in the curve for $\boldsymbol{A}/c$ itself, but it can be inferred from the clearly nonzero derivative of the displacement $Z_{cl}$ when the pulse is over. Since we want to limit our discussion to pulses that do not deliver a finite linear momentum to the electron, this case should be excluded. We only present it here in order to show that care has to be taken when the electric field is set. Panels g) and k), on the other hand, do yield a vanishing vector potential at the end of the pulse, but circumventing the problem of generating an unphysical pulse requires particular choices of the CEP and/or the length of the plateau.

Finally, panel m) is a very similar case to the 2-36-2 pulse presented previously in Fig. 5 of [1]. In this case, there is apparently a non-zero linear momentum, or a drift velocity, delivered to the electron during the ramp-on phase. Furthermore, it is important that the displacement generated during this phase is in the same direction as the drift velocity. This somewhat coincidental outcome then allows the plateau to make the linear momentum oscillate around this value, with the net effect of increasing the displacement further, basically proportional to the length of the plateau. During the ramp-off phase, the linear momentum is brought back to zero, thereby yielding both a vanishing electric field $\boldsymbol{E}$ and vector potential $\boldsymbol{A}$. Hence this pulse fulfills the minimum requirements from Maxwell's equations.

Let us now consider the changes that occur in the fields and the displacement when the vector potential $\boldsymbol{A}$ is set via Eqs. (1,2) instead of the electric field $\boldsymbol{E}$. The corresponding cases are shown in the panels in the right column of Fig. 1. Close inspection of the figures, as well as the underlying equations, shows immediately that any choice for the ramp-on/off periods and the plateau, as well as the carrier envelope phase, *guarantees* both $\boldsymbol{E}$ and $\boldsymbol{A}$ to vanish before and after the pulse, and that they do so in a smooth way if the ramp-on/off parts are set via a sine-squared envelope function. We emphasize again that this cannot be guaranteed for a linear ramp-on/off, nor for a pure Gaussian. For a corrected Gaussian envelope that causes the fields to completely vanish before and after, one would have to ensure that the correction is done in a way that ensures a smooth behavior of the electric field, i.e., the time derivative of the vector potential. The importance of using pulses without discontinuities, in particular without discontinuities of the derivative at the edges of the pulse, was also emphasized in [7].

Furthermore, it is virtually impossible to generate a significant nonzero displacement at the end of the pulse when the vector potential is set. In principle, the ramp-on phase can produce some displacement, but in this case the plateau will only cause oscillations around this value rather than a significant further deviation from it.

We stress again that the panels on the left and the right of Fig. 1 describe different physical situations. The fields are slightly different, but on each side they are individually connected via Eqs. (3) and (4), respectively.

## III. NUMERICAL RESULTS

Most of the calculations for the present work were performed in the length gauge of the electric dipole operator with the Crank-Nicolson [8] time propagation scheme. We used an updated version of a computer program originally developed for coherent control of atomic hydrogen [9]. As reported in our previous work [1], the numerical results were checked using the velocity gauge in connection with a matrix iteration [10, 11] propagator. The code is very stable and has been employed extensively in recent benchmark calculations for both atomic hydrogen [12–15] and lithium [16, 17] targets. Within the thickness of the lines or the size of the symbols, our results are independent of both the gauge and the propagation scheme.

Example results for some of the pulses introduced above, as well as some longer plateaus, are shown in Figures 2-4. We start with a 1.5-6.0-1.5 pulse, corresponding to panels c)-f) of Fig. 1. Once again, we emphasize that the pulses obtained with setting $\boldsymbol{E}$ (left column) or $\boldsymbol{A}$ (right column) according to Eqs. (1,2) look extremely similar, but they describe different physical situations. Defining the electric field this way causes a non-zero displacement for $\phi = 0^\circ$, but that displacement does not grow significantly further after the ramp-on, due to the special choice of 1.5 o.c. for the duration of the ramp-on. We see already that the angular-momentum distribution of the ejected electron is different depending on which field we start with in Eq. (1). The case that produces a non-zero displacement has a slightly enhanced tendency of generating "unusual" angular momenta, i.e., those that differ from $l = 1$ expected from the standard weak-field one-photon ionization process.

Moving on to Fig. 3, where only the length of the plateau is extended, we see a very similar pattern. The pulse with the electric field being defined via Eqs. (1,2) again creates a small non-zero displacement for $\phi = 0^\circ$, but other than enhancing the overall ionization probabil-

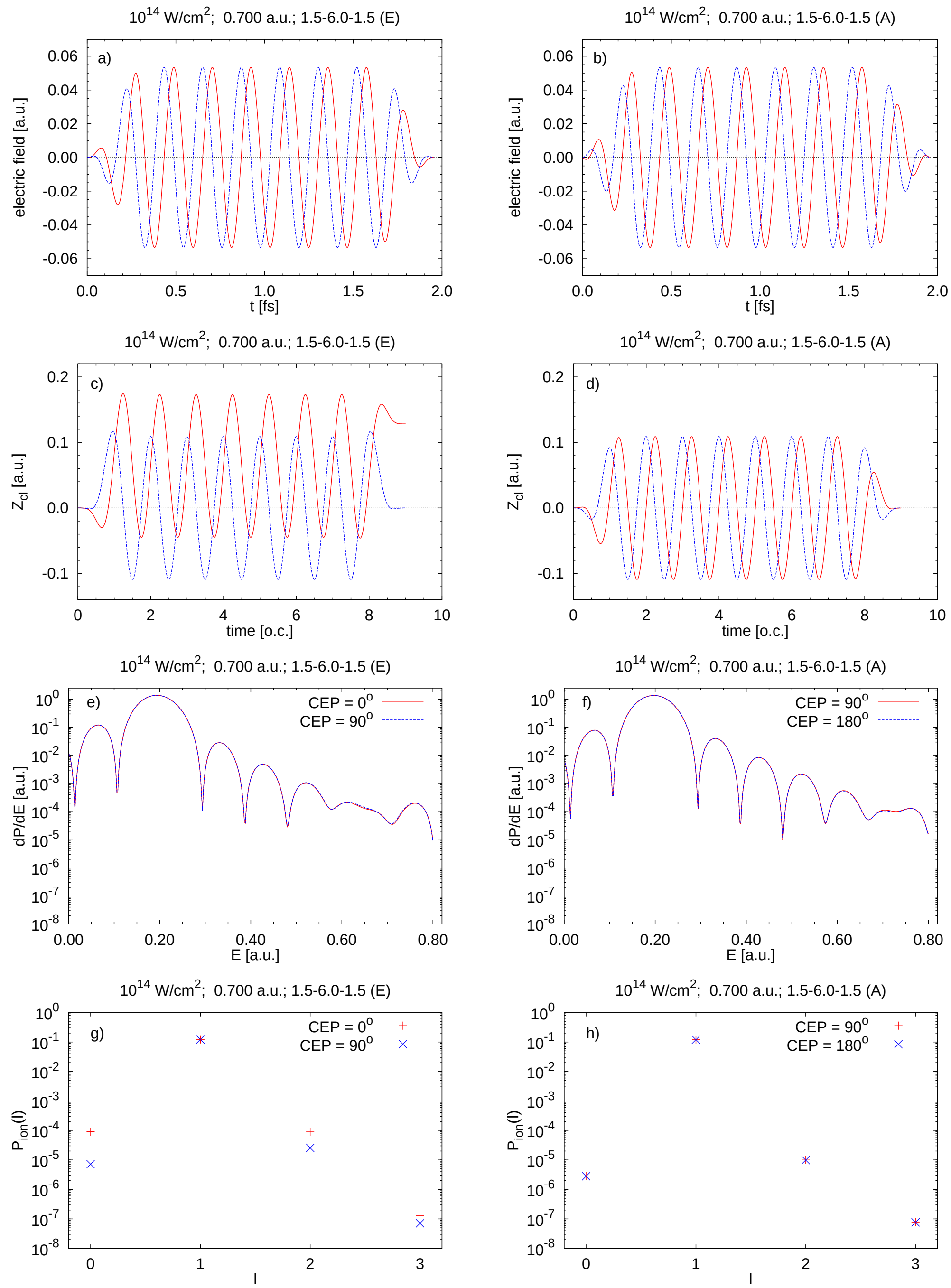


FIG. 2: (Color online): Electric field, classical displacement, energy-differential ionization probability, and $l$-decomposition for a 1.5-6.0-1.5 pulse with a central frequency of 0.7 a.u. and a peak intensity of $10^{14}$ W/cm$^2$. In the left (right) panels, the electric field (vector potential) is set according to Eqs. (1,2). For space reasons, the CEP used for panels a)-f) is indicated only in panels e) and f).

ity due to the increased length of the pulse, the plateau does not have any significant effect.

However, the plateau does play a major role if the ramp-on/off time is extended from 1.5 to 2.0 o.c. This is shown in Fig. 4. The $\phi = 0°$ pulse with $\boldsymbol{E}$ being set according to Eqs. (1,2) now causes a much larger displacement, which simply grows proportionately to the length of the plateau and the nonzero linear momentum generated during the ramp-on. Since the latter is proportional to the amplitude of the field, the displacement grows to about 3.5 a.u. for a pulse with quadrupled peak intensity (twice the amplitude) and a plateau of 36 o.c. (c.f. Fig. 5 of [1]). Already for the pulse shown in Fig. 4,

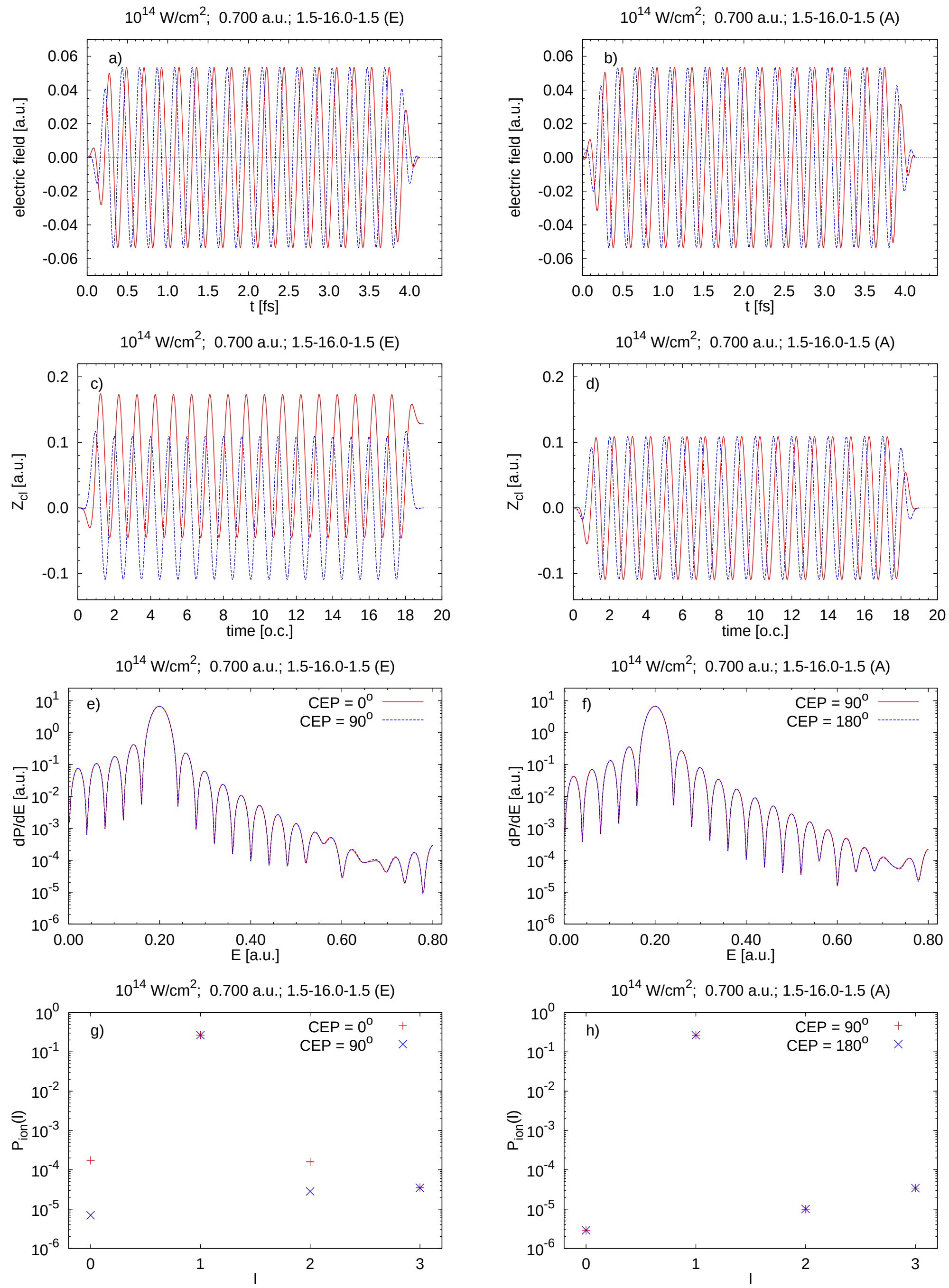


FIG. 3: (Color online): Same as Fig. 2 for a 1.5-16.0-1.5 pulse.

the likelihood of finding angular momenta other than the expected $l = 1$ is substantial, and it grows dramatically with increasing displacement (c.f. Fig. 3 of [1]).

## IV. CONSEQUENCES

The above findings are significant regarding both theoretical predictions in general and for detailed comparison with experimental measurements. It is important to keep in mind that virtually all theoretical calculations are performed with idealized pulse shapes, which are unlikely to be present in concrete experiments. Hence it is important to investigate the stability of theoretical predictions against minor changes of the input parameters. If there is a high sensitivity, the challenges to experimentally verify such predictions would likely be increased dramatically.

Even though setting either the electric field $\boldsymbol{E}$ or the

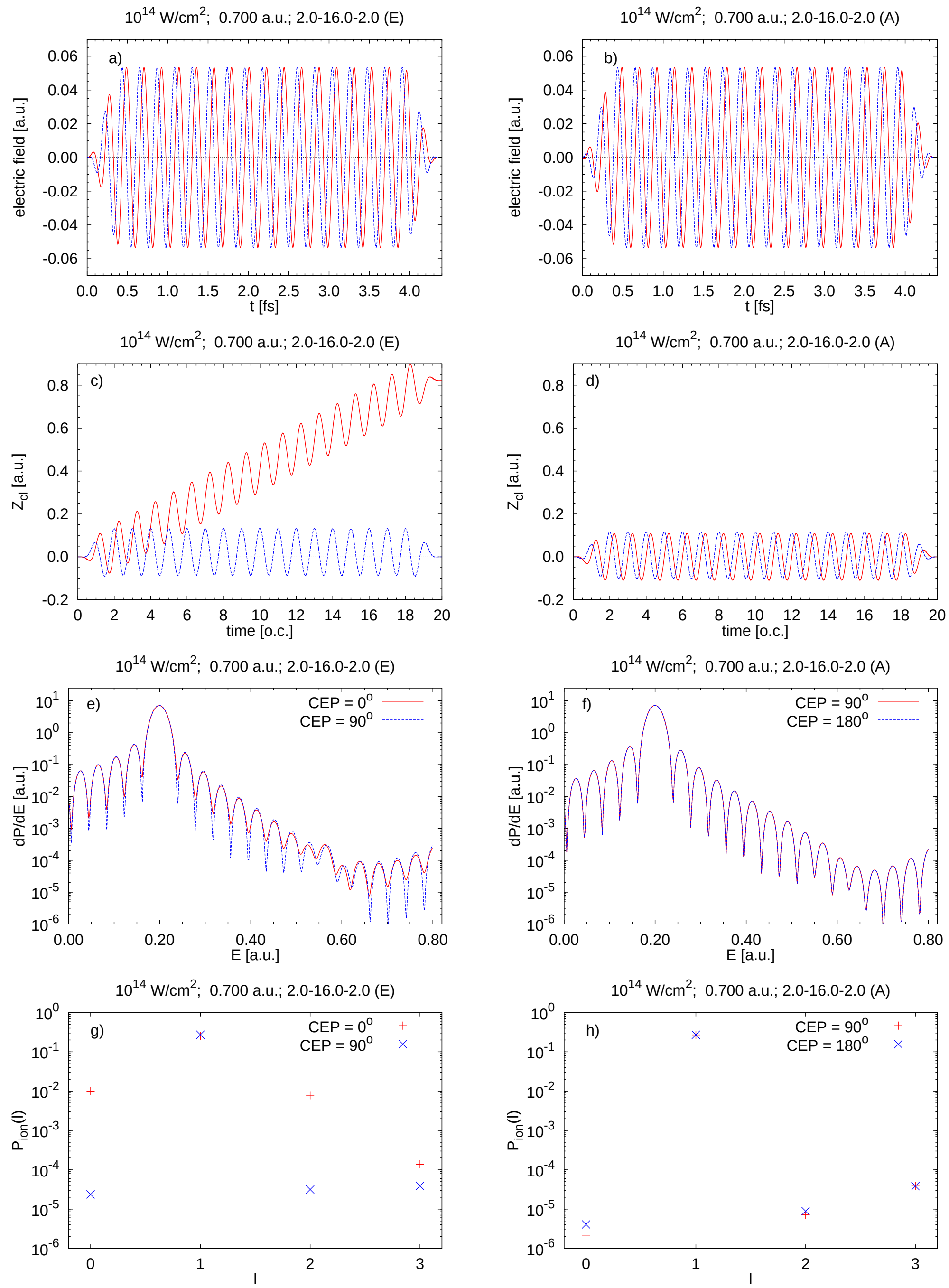


FIG. 4: (Color online): Same as Fig. 2 for a 2.0-16.0-2.0 pulse.

vector potential $\boldsymbol{A}$ according to Eqs. (1,2) and obtaining the other one by either differentiating with respect to or by integrating over time produces electric fields that are hardly distinguishable on first sight, the ramp-on/off parts cause some differences between the fields obtained in the two approaches. These differences, in turn, can lead to significant displacements under certain circumstances. They are then reflected in the angular momentum of the ejected electron and, ultimately, in its angular distribution as a directly (at least in principle) measurable observable. Examples of the latter are shown in Figs. 4 and 7 of [1].

Most importantly, the displacement is always small if the vector potential is set via Eqs. (1,2). For sufficiently smooth ramp-on/off parts of the envelope function (e.g., a sine-squared behavior), therefore, setting $\boldsymbol{A}$ this way will virtually guarantee "normal" results regarding the angular momentum distribution of the ejected electron.

Furthermore, those results are very stable, i.e., they are qualitatively independent of the carrier-envelope phase, the length of the ramp-on/off phases, and the length of any plateau that may be there. Specifically, ionization from an atomic S-state with radiation of sufficiently high central frequency to cause a "one-photon" transition will essentially produce a $p$-electron, even for very short pulses.

Defining the electric field according to Eqs. (1,2), on the other hand, may lead to significant displacements, which can theoretically be increased without limit by simply extending the plateau in the envelope function. Since the displacement is strongly dependent on the details of the ramp-on/off parts, the carrier envelope phase, and the length of the plateau, realizing such pulses will likely be a serious experimental challenge. As noted earlier, pulses with nonzero displacement and even nonzero linear momentum transfer have been generated and discussed before [2–4], albeit for much different pulse shapes than those discussed here and in our previous work [1].

## V. CONCLUDING REMARKS

We have analyzed in detail the effect of pulse shapes on theoretical predictions for atomic ionization by short XUV pulses. Given the sensitivity of some of these predictions to the details of the pulses, even when they fulfill the basic requirements of Maxwell's classical theory, we believe that defining $\boldsymbol{A}$ according to Eqs. (1,2) is generally more appropriate than using those equations for $\boldsymbol{E}$ in theoretical studies that typically employ some idealized pulse shapes. The perfect scenario, of course, would be an experimental determination of the pulse characteristics. This could be fed into a computer code on a time grid and would then allow for a direct comparison between experimental results and theoretical predictions for any observable of interest. Given the likely experimental challenges associated with such a procedure, however, it is not clear when/if this will become reality.

**Acknowledgments**

We thank Drs. Igor Ivanov and Anatoli Kheifets for helpful comments on the manuscript, and John Emmons for providing a job script that allowed us to systematically analyze a wide range of laser parameters. This work was supported, in part, by the United States National Science Foundation under grants No. PHY-1068140, PHY-1430245, and the XSEDE allocation PHY-090031 (KB and JV).

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