Multiple attosecond pulse generation in relativistically laser-driven overdense plasmas

X Lavocat-Dubuis\textsuperscript{1,2}, F Vidal\textsuperscript{2}, J-P Matte, J-C Kieffer and T Ozaki

INRS—Centre Énergie, Matériaux et Télécommunications,
1650 boulevard Lionel Boulet, Varennes, QC, J3X 1S2, Canada
E-mail: xavier.lavocat-dubuis@polymtl.ca and vidal@emt.inrs.ca

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Abstract. Using particle-in-cell simulations, we investigate the mechanisms that lead to attosecond pulses when an obliquely incident laser pulse interacts with an overdense plasma. We show that several attosecond pulses can be emitted per laser cycle as a result of the ejection of electron bunches associated with return currents within the plasma. The electron dynamics are investigated in phase space and with the help of the similarity parameter $S = n_e/a_0 n_c$.

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\textsuperscript{1} Present address: École Polytechnique de Montréal, Département de Mathématiques et Génie Industriel, CP 6079, succ. Centre-ville, Montréal, QC, H3C 3A7, Canada.
\textsuperscript{2} Authors to whom any correspondence should be addressed.
1. Introduction

In recent decades, the understanding of matter has improved significantly. From static pictures of atomic arrangement by x-ray diffraction using synchrotron radiation to dynamical studies of atomic rearrangement on the picosecond to femtosecond time scale [1, 2], the frontier has been pushed towards the study of attosecond time scale phenomena, such as the dynamics of bound electrons [3, 4]. Due to their short wavelength, the attosecond pulses used to study these fast dynamic processes are, in practice, associated with the high frequency end of the radiation spectrum produced by the interaction of a powerful femtosecond laser (pump) with gases [4–6]. In principle, attosecond pulses can also be generated from overdense plasmas [7]. Indeed, attosecond pulses arise from the constructive interferences of many high harmonics of the pump laser locked in phase. Whereas in gases the pump laser intensity must not be too high because of phase mismatch effects between the pump laser and the harmonics associated with the production of a high density of electrons [8, 9], in overdense plasmas there is no such limitation and therefore the harmonics intensity can be higher. Early numerical simulations indicated that at relativistic intensities the number of harmonic orders generated from overdense plasmas and their conversion efficiency increase dramatically [10].

The generation of high harmonics and attosecond pulses from overdense plasmas has been investigated experimentally, numerically using particle-in-cell (PIC) simulations and theoretically using the relativistic oscillating mirror (ROM) model in which the plasma surface is assumed to move as a whole under the influence of a laser field. See the recent extensive reviews of this subject [11, 12] for more details. It is now well established that up to the relativistic threshold of laser intensity, the main mechanism of high harmonics generation is coherent wake emission, whereas in the relativistic intensity regime, it is rather the relativistic Doppler effect induced by the relativistic motion of the electrons at the surface of the target. PIC simulations have revealed that in the relativistic intensity regime, high harmonics and attosecond pulses are associated with the emission of periodic high-energy electron bunches [12] coming from the plasma surface separated in time by the laser period (oblique incidence) or half the laser period (normal incidence) due to vacuum [13] and/or $J \times B$ heating [14].

Although only PIC simulations can reveal the full complexity of the electron dynamics, the ROM has proved to be extremely useful in establishing general scaling laws. For instance, Gordienko et al [15] showed in 2004 that, in the strongly relativistic limit, the harmonic spectrum intensity follows the universal law $I_m \propto m^{-p}$, where $m$ is the harmonic order and the exponent $p = 2.5–3$ depends on the laser pulse parameters. The universality of this power law stems from the fact that in ultrarelativistic interactions, there is no need to know the exact plasma surface motion. One year later, Gordienko and Pukhov [16] developed a similarity theory for ultrarelativistic laser–plasma interaction and found that the electron dynamics depend on the parameter $S = n_e/a_0 n_c$, the similarity parameter, rather than on $a_0$ and $n_e$ separately. Here, $a_0 = eE_0/m_ee\omega_0$ is the normalized vector potential, $E_0$ is the amplitude of the laser electric field, $n_e$ is the electron plasma density, $n_c = m_e\epsilon_0\omega_0^2/e^2$ is the critical density, $m_e$ is the electron rest mass, $\omega_0$ is the laser angular frequency and $e$ is the electron charge.

Then, in 2006, Baeva et al [17] applied the similarity theory to the harmonic generation from overdense plasmas for normal incidence and immobile ions and found that the power-law spectrum is best fitted with the universal exponent $p = 8/3$. This universal scaling law was obtained by assuming the so-called ‘relativistic $\gamma$ spikes’, which correspond to abrupt changes...
in the $\gamma$ factor during the plasma surface motion. They also found that the harmonic cutoff

$$m_{\text{cr}} \propto \gamma_{\text{max}}^3$$

with $\gamma_{\text{max}}$ being the maximum $\gamma$ factor of the plasma surface, instead of being
proportional to $\gamma_{\text{max}}^2$, as in the case of the uniform moving mirror model [15]. This universal
power law for $I_m$ was verified experimentally by Dromey et al [18] in 2007, with an exponent
$2.5 \leq p \leq 3$ when the laser pulse is $P$ polarized. Also, in 2008, Boyd and Ondarza-Rovira [19] found,
from one-dimensional (1D) PIC simulations, that the universal power law is best fitted
with $5/3 \leq p \leq 10/3$ and that the spectrum may reflect the details of the interaction.

Since attosecond pulses are associated with the high end of the high harmonic spectra,
where the similarity law holds best, one can expect that some similarity laws also exist in the
underlying physical processes, especially those related to electron motion. In this paper, we
study, by means of PIC simulations, the high-order harmonics and attosecond pulses generated
from overdense plasmas in the relativistic limit, when the laser pulse is obliquely incident and
$P$ polarized. We first examine the harmonic spectra generated when the similarity parameter $S$
is kept constant. From these spectra, we then show that attosecond pulses come out by filtering
out the low-harmonic orders. We show, in particular, that in the ultrarelativistic regime, more
than one attosecond pulse is usually generated per laser cycle and that these pulses are directly
connected with several relativistic electron bunches coming from the plasma and associated with
return currents. We then show how the similarity theory with the parameter $S$ can be used to
understand the electron dynamics and how it manifests itself in the phase space of the electron
motion.

2. High-order harmonic decay law

When a few femtosecond laser pulse of relativistic intensity ($a_0 \gg 1$) interacts with a solid
target, the ionization of the target surface takes place in less than one laser cycle [20], therefore
creating a strongly overdense plasma, which reflects the laser light. But, as the plasma surface
oscillates at relativistic velocities, retardation effects lead to strong distortions of the reflected
pulse and thus to many harmonics of the laser frequency [21, 22]. Many attempts have been
made to model the plasma surface motion [23, 24] in order to interpret the experimental and
simulation results. According to the similarity theory [16], the power law of the harmonic
efficiency is universal. However, for $P$-polarized laser pulses, both experiments and simulations
found instead a range of exponents $p$ that fit the power-law spectrum [18, 19].

Since attosecond pulses are closely related to harmonic spectra, we first characterized the
harmonic spectra produced in various conditions. For the investigation presented in this paper,
we used the relativistic 1D PIC code BOPS [25], which can simulate oblique incidence by the
Bourdier scheme [26], which consists in performing a Lorentz transformation to the reference
frame in which the incidence is normal. This code assumes a pre-ionized medium with constant
electron number. We made several simulation runs with plasma densities and normalized laser
amplitudes in the ranges $n_e = 100 \sim 400n_c$ and $a_0 = 25 \sim 100$, respectively. The laser pulse
was Gaussian in time, of duration $\tau_p = 10\,\text{fs}$ at full-width at half-maximum (FWHM), and
wavelength $\lambda_0 = 0.8\,\mu\text{m}$. The time step used in the simulations was sufficiently small to resolve
many harmonics of the laser frequency and the mesh spacing was about the initial Debye length
(the initial temperature was 0.625 keV). The plasma ($10c/\omega_0$ thick) was in the middle of the
simulation box ($80c/\omega_0$ long) and had a step-like density profile at both the front and the rear
side. The aluminum ions were assumed to be an immobile neutralizing background. We did not
take the effect of the laser prepulse into account in our simulations (which generally reduces
the harmonic generation efficiency [27]) and we assumed that the main pulse interacts with a step-like high-density plasma profile. We performed complementary simulations with 6-fold thicker targets (not shown), and these gave the same results as the $10c/\omega_0$-thick target used in this paper. Thus, we may conclude that there is no artificial effect related to the choice of the target thickness.

In figure 1, we show spectra obtained at incidence angle $\theta = 45^\circ$ and for plasma densities $n_e = 100n_c$, $200n_c$, and $400n_c$ while keeping $S = 3$. All of the spectra are best fitted with an average exponent $p \simeq 3$ for the high-order harmonics, in agreement with previous works [15, 17, 19]. We obtained the same average exponent for plasmas with $n_e = 100n_c$ and $S = 1–4$. We could note modulations in the harmonic spectra, as observed experimentally [24]. Indeed, according to [28], when $a_0 \geq 1$, the laser electric and magnetic forces are able to drive electron bunches out of the plasma surface. (An intuitive and practical definition of the concept of ‘plasma surface’ used in what follows can be the locus of 50% of the initial plasma density.) These electron bunches are then pulled back into the plasma where they can excite plasma oscillations, which in turn give the modulations in the harmonic spectra. However, in [15, 17] it was shown how the interferences produced by different ‘relativistic $\gamma$ spikes’ during a laser period can lead to spectrum modulations as well when $a_0 \gg 1$.

To assess the range of validity of these findings, we also made runs with mobile aluminum ions and very little difference was seen for $\tau_p = 10$ fs. However, since most of the high-power (10–100 TW) laser systems currently in operation deliver pulses with a duration of $\tau_p \simeq 50$ fs, we thus used as well a laser pulse of duration $\tau_p = 55$ fs with mobile ions in our simulations. For instance, using a plasma density of $n_e = 400n_c$ and $a_0 = 100$ ($S = 4$), we observed that the harmonic efficiency is drastically decreased (almost no harmonic above 100) and that the spectrum modulations are strongly attenuated. We observed that the plasma expansion is about $2c/\omega_0$ at a time of $0.5\tau_p$ past the peak of the pulse and increases rapidly after that point. We believe that, due to plasma expansion, the electron bunches are less effective in exciting plasma

![Figure 1.](http://www.njp.org/)
waves when they return to the plasma [28]. Because the electron temperature, which determines the sound velocity in the plasma and thus the plasma expansion velocity, scales as $a_0$, even for short laser pulse durations, plasma expansion could impede the harmonic generation when $a_0 \gg 1$.

3. Attosecond pulse generation

Mathematically, the interference of many monochromatic light waves gives rise to a short pulse and the more Fourier components there are in the signal with comparable amplitudes and locked in phase, the shorter the pulse. The slow power-law decay of the harmonic spectrum and the fact that the harmonics emitted from the solid surface are phase-locked provide favorable conditions for the generation of attosecond pulses that may be suitable for attosecond time scale experiments [29]. However, to achieve attosecond temporal resolution, it is essential to generate single, isolated attosecond pulses [30]–[32]. The emission of attosecond pulses is due to the fact that the acceleration of the plasma surface varies on a time scale much shorter than the laser period.

Figure 2 shows simulation results for the reflected magnetic field $B_z$ (the direction $z$ being normal to the plane of incidence) as a function of time for $S = 4$ and $S = 2$, with $n_e = 100n_c$ and $\tau_p = 10$ fs. We filtered out the first 50 harmonic orders and Fourier transformed the remaining spectrum. We obtained pulses of $\sim 40$ as (0.015 laser cycle), produced at each laser cycle. Experimentally, by polarization gating (rapid switch between circular and linear polarizations), it is possible to select a single attosecond pulse within a pulse train [29, 31]. However, in this strongly relativistic regime, we can see that the process of high harmonic generation from solid surfaces gives rise to not only one but two attosecond pulses per laser cycle, separated in time by $\sim 200$ as when $S = 4$ and $\sim 400$ as when $S = 2$. (The reason for this difference will be discussed later in section 4.) The amplitude of the second attosecond pulse is up to one third of that of the first one, as can be seen in the insets of figure 2. The polarization gating technique ($\sim 5$ fs gate) would thus be hardly able to separate these attosecond pulses.
Figure 3. Attosecond pulse generation for a single-cycle cosine wave with the absolute phase $\phi = 0$. The parameters are $n_e = 100n_c$, $S = 4$ and $\theta = 45^\circ$. (a) Incident (---) and reflected (·····) pulses and the attosecond pulses (——) after filtering. (b) The two attosecond pulses, separated by $\sim 200$ as, can be fitted with Gaussian envelopes of $\sim 40$ as at FWHM.

To generate a single attosecond pulse, it was proposed to use a few laser cycle pulse [30]. However, as shown in figure 3, even for a single-cycle cosine pulse (absolute phase $\phi = 0$) at $\lambda_0 = 0.8 \mu m$, we observed two main attosecond pulses of $\sim 40$ as in one laser cycle, separated by $\sim 200$ as and even a third one; however, much weaker than the first two (we can almost see them in figure 2(b) as well). This result remains the same using a sine waveform.

In every case, it appears that the first attosecond pulse in one laser cycle is stronger than the others because it is connected with the maximum ejection velocity of the electron bunch (which then induces a strong relativistic Doppler shift), as we will see next. Attosecond pulses are seen on both the front and the rear side of the plasma, but in the latter case, their amplitude was much weaker. In the case of $S = 4$ at normal incidence, we observed weaker attosecond pulses of duration $\sim 40$ as produced at twice the laser frequency, but did not observe secondary attosecond pulses. Note that in [30], the authors showed similar secondary pulses for similar parameters ($a_0 = 20$, $n_e = 80n_c$, $S = 4$) but did not explain this feature.

The emission of attosecond pulses is directly connected with the dynamics of the plasma surface electrons. Figures 4(a), (c) and (e) present the electron phase space ($x$, $p_x$) and figures 4(b), (d) and (f) show the electromagnetic fields in the simulation reference frame (primed quantities) in which the incident beam is normal to the plasma surface, at time $t = 17.3$ fs (cf figures 4(a) and (b)), $t = 17.8$ fs (cf figures 4(c) and (d)) and $t = 18$ fs (cf figures 4(e) and (f)) (time in the laboratory frame). The plasma surface is initially located at $x = 5$, and $t = 0$ is defined as $1.5\tau_p$ before the peak of the laser pulse. The advantage of the simulation reference frame is that the field $E'_x$ is essentially due to the space charge field and involves no contribution from the laser field, which simplifies the discussion somewhat.

When the laser wave impinges on the plasma, it pushes the electrons over a (normalized) distance $k_0x \approx 0.5$, where $k_0 = 2\pi/\lambda_L$ is the laser wavenumber (i.e. from 5 to 5.5) (cf figure 4(a)) and the electron density increases to a value many times the initial one. This creates a strong positive space charge field $E'_x$, which tends to accelerate the electrons towards
Figure 4. Electron phase space and electromagnetic field structure along the $x$-axis at $t = 17.3$ fs (a, b), at $t = 17.8$ fs (c, d) and at $t = 18$ fs (e, f) in the case of $S = 4$, $n_e = 100n_c$ and $\theta = 45^\circ$. The plasma surface is initially located at $x = 5$. Space is normalized to the laser wave vector $k$ and momentum to $mc$. The primes refer to quantities in the simulation frame with $E_x'$ (---), $B_z'$ (-----) and $E_y'$ (· · · · · ·).

the vacuum, but the magnetic field $B_z'$ (cf figure 4(b)) is strong enough to prevent the electrons, which have a relativistic velocity along the $y$-axis, from escaping and, most of the time, the electrons are confined inside the plasma (cf figure 4(a)) and end up circling in the incidence.
plane \((x, y)\). The \(\mathbf{E} \times \mathbf{B}\) electron drift induces a strong negative current along \(y\) (not shown), which results in a self-generated magnetic field about four times higher than the incident field \((a_0 = 25)\), which efficiently traps the electrons. The self-generated magnetic field is therefore an important feature of the electron dynamics in this relativistic regime because it traps the electrons inside the plasma, while the electric field \(E'_y\) is negligible (cf figure 4(b)). Then, \(E'_y\) grows and it may happen that for a short time \(E'_x\) is strong enough to cancel the magnetic field effect (cf figure 4(d)). \(E'_x\) being positive, the electric current thus generated tends to counteract the one giving rise to \(B'_z\) (cf figures 4(b) and (d)). At this very moment, the trapping structure is broken and the space charge field \(E'_x\) can accelerate the electrons towards the vacuum, therefore creating an electron bunch (cf figure 4(c)), while \(B'_z\) is reversing (cf figures 4(d)–(f)). (We note that the electron drift velocity can also contribute to the ejection of electrons before the magnetic field reverses.) This dense electron bunch compresses the reflected laser wave to produce an attosecond pulse. Indeed, in figure 4(f), we see that \(E'_x\) can no longer penetrate the plasma. Yet, due to the positive charge left by the ejected electrons, the plasma generates a strong return current \(\sim 200\) as later (cf figures 4(e) and (f)), thus giving rise to a second attosecond electron bunch. This return current is associated with the acceleration of slower electrons close to the surface. The effect of the first electron jet, apart from the return current, is to create a strong magnetic field, which will deflect the second electron jet. We note that the generation of strong magnetic fields may explain the emission of electron bunches out of the specular direction [33].

In the laboratory frame, which moves at velocity \(-c\sin(\theta)\) with respect to the simulation (‘boosted’) frame, the quantities shown in figure 4 transform as \(x = x'\), \(P_x = P'_x\) and

\[
E_x = (E'_x - c\sin(\theta)B'_z) / \cos(\theta),
\]

\[
E_y = E'_y,
\]

\[
B_z = (B'_z - \sin(\theta)E'_x/c) / \cos(\theta),
\]

where \(c\) is the speed of light and \(\theta\) is the incidence angle. Therefore, \(E_x\), \(E_y\) and \(B_z\) have the same sign and same shape as \(E'_x\), \(E'_y\) and \(B'_z\), respectively, and the discussion made for the primed quantities (simulation frame) can be transposed without changes for the unprimed quantities (laboratory frame).

To gain more insights into how the emission of electron bunches is synchronized with the emission of attosecond pulses, we look at the laser electric field \(E_y = E'_y\) at the plasma surface as a function of time. Figure 5 shows the electric field along the \(y\)-axis (tangential to the plasma surface) and the electron charge density, both at the plasma surface \((x = 5)\) in the case of \(S = 4\), \(n_e = 100n_c\) and \(\theta = 45^\circ\). We can see that the temporal shape of \(E_y\) is strongly influenced by the presence of very-short-duration electron bunches \(\sim 50\) as that sweep across the initial plasma surface. We could note the simultaneity between the sharp edge of the electric field, which is associated with the generation of very short electromagnetic pulses, and the ejection of electrons. The electron density \(n_b\) in the bunches is very high, almost three times the initial plasma density, i.e. \(n_b \approx 300n_c\) in the present case. Accordingly, we note that \(\sim 200\) as after the first electron jet, there is a second one, which is somewhat less dense and less energetic. Indeed, the space charge field \(E'_x\), which accelerates the electrons towards the vacuum, is weaker (cf figure 4(f)) when the second electron bunch is ejected out of the plasma surface. Consequently, the second attosecond pulse is weaker than the first one. The structure located between \(t = 18.5\) fs and \(t = 19\) fs is not an electron bunch; it is associated with the onset of the...
trapping of the plasma surface electrons before they are pushed in by the laser. Again, we can see that, most of the time, the electrons are located inside the plasma surface (almost 0 density) and only when they are pushed out into the vacuum will they compress the reflected laser wave, producing attosecond pulses.

4. Relativistic regime: similarity

In this section, we show how the ultrarelativistic similarity theory [16, 17], based on the parameter $S = n_e/a_0n_c$, manifests itself in attosecond pulse generation. According to the similarity theory, when $a_0 \gg 1$, the electron momentum $P$, the electric and magnetic fields scale as $a_0$ and the electron dynamics depends on $S$ instead of $a_0$ and $n_e$ separately. For instance, if we assume that all the electrons ejected from the plasma surface move as a whole, an electrostatic field will be created (given by the Poisson equation) $E_p = n_e e \Delta x / k \varepsilon_0$, where $\Delta x$ is the (normalized) excursion length. If we also assume that this field balances the normal component of the external field, which is about $2E_0 \sin \theta$, we obtain $\Delta x \simeq 2 \sin \theta / S$. The excursion length $\Delta x$ of the electrons thus depends only on $S$ instead of $a_0$ and $n_e$ separately. Furthermore, the (normalized) skin depth, which reads, in the relativistic case, $\delta = \sqrt{\gamma \omega / \omega_p}$, becomes $\delta \simeq 1 / \sqrt{S}$ when $a_0 \gg 1$. Again, the skin depth does not depend on the plasma frequency alone but on $a_0$ as well through $S$. These examples show that when $S$ is kept constant, the electron dynamics are similar.

The best way to observe the similarity is to look at the electron phase space. Figure 6 represents the electron phase space at $t = 17$ fs for two plasmas having the same similarity parameter $S = 4$ but different densities, that is, $n_e = 100n_c$ and $n_e = 200n_c$. The electron trajectories are very similar in both cases. We can note the circling trajectories (trapping structure in the self-generated magnetic field) of the surface plasma electrons. For $S = 4$, we have $\delta \simeq 1 / \sqrt{S} = 0.5$. Thus, in both cases, the trapping structure is about a skin depth long, as expected since the applied field practically vanishes beyond this distance. The excursion length outside the plasma is $\sim \sqrt{2} / S = 0.35$, as can be seen in figure 4(e). For other values of $S$, we observed the same behavior. For example, in the case of figure 2, when $S = 2$, the secondary
pulse is delayed in time by approximately twice the value when $S = 4$, with 400 and 200 as, respectively. Indeed, the electrons of the second jet (return current) are pushed out of the plasma surface when the electrons of the first jet are going back to the plasma (cf figure 4(e)), covering a distance $\propto 1/S$, the excursion length, at nearly the velocity of light.

The similarity theory, with the similarity parameter $S$, thus gives a simple way to analyze the plasma behavior in the relativistic limit. Several short high-density electron bunches per laser cycle, strong magnetic field generation, as well as trapped structures are all common features in this limit.

In principle, all of the conclusions of the similarity theory based on the ‘relativistic $\gamma$ spikes’ model of the plasma surface motion [17] can be interpreted in terms of the motion of the electrons. In particular, in [17] the ‘$\gamma$ spikes’ were related to the nulls of the tangential momentum $P_y$ of the electrons. We have previously seen that, when this occurs, the current produced by the self-generated magnetic field is canceled by the laser electric field (cf figure 4(d)) and the longitudinal space charge field gives rise to the attosecond bunches. We can thus connect the ‘$\gamma$ spikes’ directly to the electron bunches.

In addition, the cutoff frequency of the harmonic spectra $m_{cr} \propto \gamma_{\text{max}}^3$, obtained in [17] can be simply understood in terms of relativistic radiating charges. Indeed, as shown in [34], the radiation emitted by relativistic electrons is the same as that emitted if they moved along an instantaneously circular path of radius $R$. A cutoff frequency is obtained at $\omega_{cr} \approx (c/R)\gamma^3$. Setting $c/R \approx \omega_0$, the laser angular frequency, one finds that $\omega_{cr} \approx \omega_0\gamma^3$, which means that the highest harmonic is of the order of $\gamma^3$. Thus, the spectrum of a strongly relativistic particle has the same cutoff dependence as for the harmonics generated from a plasma surface in the framework of the ‘relativistic $\gamma$ spikes’ model of Baeva et al [17].

5. Conclusion

High harmonics and attosecond pulses produced from a solid surface in the strongly relativistic regime ($a_0 \gg 1$) have been investigated by means of PIC simulations. We checked that the
power law of the harmonics intensity spectrum decay is best fitted with an average exponent $p = 3$ for both oblique and normal incidence. This result was found to hold for all of the similarity parameters ($S = 1−4$) and electron densities ($n_e = 100−400n_c$) investigated. This slow (although modulated) decay of the harmonic spectrum is a necessary condition for the generation of attosecond pulses. Indeed, by filtering out the first 50 harmonics, we obtained trains of $\sim 40$ as (0.015 laser cycle) pulses. The attosecond pulses are associated with the ejection of electron bunches from the plasma. We have seen that not only one but several electron bunches are usually produced per laser cycle in oblique incidence due to the plasma return currents. These return currents result from the acceleration of electrons by the positive charge left by the preceding electron bunch. The multiple attosecond pulses, which are separated by only a few hundreds of attoseconds, may limit the suitability of this scheme for investigating ultrashort time scale dynamics. The secondary attosecond pulse, which can have up to 30% of the amplitude of the first one, may indeed not be negligible in processes depending on the field amplitude instead of the intensity, such as electron acceleration and tunnel ionization. In addition, we have shown that because of the strong self-generated magnetic field, the electrons are trapped inside the plasma most of the time and that this feature is common to relativistically driven overdense plasmas. The similarity parameter $S$ proves to be a key parameter for understanding the electron dynamics as they are related, in particular, to the plasma skin depth $\sim 1/\sqrt{S}$ and the electron excursion length $\sim 1/S$.

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