Generation of Ultra-short Light Pulses
by a Rapidly Ionizing Thin Foil

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Abstract

A thin and dense plasma layer is created when an intense laser pulse impinges on a solid target. The nonlinearity introduced by the time-dependent electron density leads to the generation of harmonics. The pulse duration of the harmonic radiation is related to the risetime of the electron density and thus can be affected by the shape of the incident pulse and its peak field strength. Results are presented from numerical particle-in-cell-simulations of an intense laser pulse interacting with a thin foil target. An analytical model which shows how the harmonics are created is introduced. The proposed scheme might be a promising way towards the generation of attosecond pulses.

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In recent years several mechanisms generating harmonics of electromagnetic radiation have been discovered. Among these high-order harmonics from gases [1] and harmonics production from the plasma-vacuum boundary when a laser pulse impinges on a solid target [2] are the most prominent ones. Especially the high-order gas-harmonics, exhibiting a “plateau” instead of a rapid decrease with the harmonic order, seem to be a promising source for xuv “water-window”-radiation. Apart from the effort to make progress towards shorter wavelengths, another goal is to achieve shorter pulse durations because the temporal resolution in pump-probe experiments clearly depends on the pulse length. One scheme proposed to generate attosecond pulses is based on phase-matching pulse trains which are produced by a laser pulse focused into a jet of rare gases [3]. Another method makes use of the fact that the efficiency of gas-harmonics generation is sensitive to the ellipticity of the incident laser light [4].

The method to generate an ultra-short low order harmonic laser pulse as proposed in this Letter is based on a completely different mechanism. We would like to stress in advance that the method is not appropriate for generating particularly high order harmonics efficiently.

We assume a linearly polarized laser pulse impinging perpendicularly on a thin foil target. In the following analytical and numerical calculations the whole setup is treated one dimensionally in space, i.e., the laser pulse propagates along $x$ and the electric field is in $y$-direction. The foil will be ionized by the pulse. To calculate the pulse propagation through a medium with varying free electron density one has to solve the inhomogeneous wave equation

$$\frac{\partial}{\partial x^2} E(x, t) - \frac{1}{c^2} \frac{\partial}{\partial t^2} E(x, t) = \frac{1}{\varepsilon_0 c^2} \frac{\partial}{\partial t} j(x, t).$$

(1)

The Green’s function of this equation is $G(x, x', t, t') = -c \Theta[c(t - t') - |x - x'|]/2$ where $\Theta(y)$ is the step-function, i.e., $\Theta(y) = 1$ for $y > 0$ and 0 otherwise. The solution of (1) can be written as the sum of the incident field $E_0(x, t)$ and the radiation field produced by the current $j(x, t)$, i.e., $E(x, t) = E_0(x, t) + E_r(x, t)$, with

$$E_r(x, t) = -\frac{1}{2c \varepsilon_0} \int dt' \int dx' \Theta[c(t - t') - |x - x'|] \frac{\partial}{\partial t'} j(x', t').$$

(2)
In order to model thin foils we now assume a delta-like current in space [5]. If the thin foil is located at $x = 0$ the current is $j(x, t) = [-en_e(x, t)v_e(x, t) + Zen_i(x, t)v_i(x, t)]\ell \delta(x)$, with $n_{e,i}$ and $v_{e,i}$ the electron and ion density, and velocity, respectively, and $Z$ the ion’s charge state. Integrating $j(x, t)$ over $x$ one finds that the current per unit area equals that of a “real” physical thin foil of thickness $\ell$ (as long as there is no strong electron density gradient across the foil). Inserting the current $j(x, t)$ into (2) and performing the spatial integration lead to

$$E_r(x, t) = -\frac{\ell}{2c\varepsilon_0} \int dt' \Theta[c(t - t') - |x|] \frac{\partial}{\partial t'} j_h(0, t')$$

(3)

where $j_h(x, t) = -en_e(x, t)v_e(x, t) + Zen_i(x, t)v_i(x, t)$. If we assume that the pulse hits the target at $t = 0$ we finally get

$$E(x, t) = E_0(x, t) - \frac{\ell}{2c\varepsilon_0} j_h(0, t_{\text{ret}})$$

(4)

for the electric field ($t_{\text{ret}} = t - |x|/c$ is the retarded time). The current $j_h(0, t_{\text{ret}})$ itself depends on the electric field. Neglecting the ionic contribution to the current, we have

$$j_h(0, t_{\text{ret}}) = \frac{e^2}{m} n(0, t_{\text{ret}}) \int_0^{t_{\text{ret}}} dt' E(0, t')$$

(5)

where $n = n_e$.

Here it has been assumed that all newly created electrons are born with the appropriate fluid element velocity and that collisional as well as relativistic effects are negligible. Besides, we neglect in our analytical treatment energy subtraction from the pulse due to the finite ionization energy of the target material. How this energy loss as well as momentum transfer due to the velocity distribution of the ionization produced electrons can be incorporated in a fluid description is studied in [6]. All pulse intensities considered in this Letter do not cause relativistic electron motion.

Supposing an ionization rate $\Gamma$ applicable for pulse intensities under consideration has been chosen, the electron density $n$ in the foil is given by $n(0, t_{\text{ret}}) = n_0 \left[1 - \exp \left(-\int_0^{t_{\text{ret}}} dt' \Gamma[E(0, t')]\right)\right]$. When the target is fully ionized the electron density is $n_0$. We finally end up with the following integral equation for the electric field $E(x, t)$,
\[ E(x, t) = E_0(x, t) - \xi \left[ 1 - \exp \left( - \int_0^{t_{\text{ret}}} dt' \Gamma[E(0, t')] \right) \right] \int_0^{t_{\text{ret}}} dt' \ E(0, t'), \] 

(6)

where

\[ \xi = \frac{e^2 n_0 \ell}{2 e \varepsilon_0 m} = \pi \left( \frac{\omega_p}{\omega_1} \right)^2 \frac{\ell}{\lambda_1} \omega_1. \] 

(7)

\(\omega_p\) is the plasma frequency of the fully ionized target, \(\omega_p^2 = e^2 n_0 / \varepsilon_0 m\), and \(\omega_1\) and \(\lambda_1\) are the incident EM wave’s frequency and length, respectively. The dimensionless parameter \(\xi/\omega_1\) determines how strong the propagation of the incident pulse is affected by the foil. For \(\xi/\omega_1 \ll 1\) the foil is optically “thin”.

If the foil is not pre-ionized or ionization is not completed already during the very early part of the pulse, (6) remains nonlinear due to the electron-density shape-factor which depends on the electric field through the rate \(\Gamma[E]\). Therefore one expects harmonics in the transmitted and reflected light.

In what follows we will restrict ourselves to study (6) in first order in \(\xi/\omega_1\) (i.e., we assume a “thin” foil and iterate (6) once). At the position of the foil then

\[ E(0, t) = E_0(0, t) - \xi \left[ 1 - \exp \left( - \int_0^t dt' \Gamma[E_0(0, t')] \right) \right] \int_0^t dt' \ E_0(0, t') \] 

(8)

holds. Here, the difficulty is to calculate \(\exp(- \int_0^t dt' \Gamma[E_0(0, t')])\). The ionization rate \(\Gamma\) depends on the absolute value of the electric field, i.e., the rate has two maxima per fundamental laser cycle. Supposed that the pulse envelope \(\hat{E}_0\) is sufficiently adiabatic the rate may be expanded in a Fourier-series with even multiples of the fundamental frequency only, and a slowly time-dependent envelope \(\hat{\Gamma}\),

\[ \Gamma[|E_0(t)|] = \hat{\Gamma} \left\{ \frac{1}{2} a_0 + \sum_{n=1}^{\infty} (a_{2n} \cos 2n\omega_1 t + b_{2n} \sin 2n\omega_1 t) \right\}. \] 

(9)

Since the rate \(\Gamma\) is a complicated functional of the field, in general all terms in the expansion (9) are present. However, if we assume the incident pulse (divided by its amplitude) to be an even function in time, \(E_0(t) \sim \cos \omega_1 t\), all coefficients \(b_{2n}\) in (9) vanish. This finally leads to
\[ E(t) = \hat{E}_0 \cos \omega_1 t + \frac{-\xi}{\omega_1} \left\{ 1 - \exp(-\alpha_0 t) \prod_{n=1}^{\infty} \sum_{m=-\infty}^{\infty} (-i)^m \exp(-i2nm\omega_1 t) I_m(\alpha_n) \right\} \times \hat{E}_0 \sin \omega_1 t. \]  

Here, \( \alpha_0 = \hat{\Gamma}a_0/2 \), \( \alpha_n = \hat{\Gamma}a_{2n}/2n\omega_1 \), and \( I_m \) is the modified Bessel-function. Note that \( \alpha_0, \alpha_n \) are slowly time-dependent due to their dependence on \( \hat{\Gamma} \).

From (10) we can deduce that by ionization in first order of \( \xi/\omega_1 \) only odd harmonics will be produced: In (10) the term in curly brackets is composed of even harmonics, but multiplied with \( \sin \omega_1 t \) odd harmonics are created.

Harmonics production is enhanced when \( \xi/\omega_1 \) is increased. Therefore increasing the density or the thickness of the foil acts in favor of the creation of harmonics. However, one has to bear in mind that a perturbative treatment with respect to \( \xi/\omega_1 \) preceded here. Furthermore, increasing density \( n_0 \) or thickness \( \ell \) simply makes the foil less transparent.

In order to get rid of the product of sums of Bessel-functions in (10) we now assume that the \( n = 1 \)-contribution in the Fourier-decomposition of the rate strongly dominates, i.e., \( \alpha_2, \alpha_3, \ldots \approx 0 \). Eq. (10) then simplifies to

\[ E(t) = \hat{E}_0 \cos \omega_1 t + \frac{-\xi}{\omega_1} \left\{ 1 - \exp(-\alpha_0 t) \sum_{m=-\infty}^{\infty} (-i)^m \exp(-i2m\omega_1 t) I_m(\alpha_1) \right\} \times \hat{E}_0 \sin \omega_1 t. \]

The argument of the Bessel functions, \( \alpha_1 = \hat{\Gamma}a_2/2\omega_1 \) is basically the ratio of ionization rate to fundamental frequency. Supposing that \( \alpha_1 \ll 1 \) which is the case if ionization lasts for several laser cycles, we can use the asymptotic expression \( I_n(z) \approx [(z/2)^n]/n! \) for the modified Bessel-function. In this approximation the relative peak height of subsequent harmonic orders is proportional to \( \alpha_1^2 \), i.e., increasing the field strength (which leads to an increased ionization rate) should result in less rapidly dropping harmonics in the spectrum. However, the efficiency of harmonics generation decreases with increasing field strength simply due to the fact that the nonlinearity in Eq. (10) switches off as soon as the foil
is fully ionized. Therefore there seems to be no way to achieve very high harmonics by ionization effects.

A 1D particle-in-cell (PIC) code was used to simulate laser pulse-thin foil interaction. In order to incorporate ionization the rate equations governing the ionization state of the target were solved during each “PIC-cycle” [7]. For simplicity only one ionization state with the ionization energy of hydrogen (13.6 eV) was assumed. Landau’s tunneling-rate [8] was used which is a reasonable choice for the field strengths and frequencies under consideration. The short risetime of the electron density forces a tiny time step. Usually one fundamental laser cycle (wavelength) was sampled by 1000 temporal (spatial) gridpoints. About $10^4$ computer particles, sampling the physical charge densities of the thin foil were found to be sufficient. The ions were mobile (although this is unimportant for the effect under consideration) and 1836 times heavier than the electrons (hydrogen).

In Fig. 1 numerically computed spectra of the transmitted light are shown for 5 different peak field strengths $\hat{E}_0$, corresponding to intensities $I = 4.0, 4.8, 6.5 \times 10^{14}$ and $1.1, 1.6 \times 10^{15}$ W/cm$^2$. All other parameters were held constant: wavelength $\lambda_1 = 815$ nm, foil thickness $\ell = \lambda_1/10$, incident $\sin^2$-shaped laser pulse of duration $T = 30$ fs, and the density was the critical one with respect to the fundamental frequency, i.e., $n_0 = n_c = 1.68 \times 10^{21}$ cm$^{-3}$.

The higher the field strength, the broader are the harmonics peaks in the spectrum. The pulse length of the harmonics radiation is closely related to the risetime of the electron density in the foil since as soon as the density remains constant harmonics production will stop. In Fig. 2 the normalized electron density is plotted vs. time for the 5 field strengths of Fig. 1. A risetime covering 3 fundamental periods for the weakest pulse and only one cycle for the strongest pulse can be inferred from the plot (each stair in the density corresponds to one half cycle). The density risetime is very sensitive to field strength and pulse shape. Increasing the field strength leads to a decreasing density risetime and hence to a shorter harmonics pulse length. However, we already mentioned above that the conversion efficiency decreases when the field strength of the incident pulse is increased since the nonlinearity
switches off too soon. Furthermore the “harmonics” peaks are shifted and asymmetrically broadened if complete ionization occurs within only one fundamental cycle (or even less). In the limit of a step-like behavior of the electron density the spectrum resembles the Fourier-transform of the Θ-function with no $\omega_1$-harmonics structure at all.

In Fig. 1 the pulse length of the harmonic radiation can be estimated by fitting the peaks in the spectrum to a Fourier transformed “test envelope” $\sim \sin^2 \pi t/T_n$. $T_n$ is the pulse duration of the $n$th harmonic. For the 5 cases of Fig. 1 one finds for the pulse length of the 3rd harmonic $T_3 = 3.3, 3.0, 2.3, 2.0, 1.9$ times the fundamental period $\tau = 2\pi/\omega_1$. A lower limit for $T_3$ certainly is $\tau$ itself because a shorter risetime of the electron density leads to a vanishing $\omega_1$-structure in the spectrum. The power in the 3rd harmonic is about $10^{-6}$ of the fundamental.

One may object that the incident pulse intensity was already small (at least for “up-to-date” short pulse laser systems) so that the 3rd-harmonic pulse with only a millionth of its intensity is not very useful. However, the incident pulse might be a stronger but defocused pulse so that the 3rd harmonic output, when focused, becomes considerable. Besides, using a shorter fundamental wavelength (and correspondingly a thinner and/or denser foil) would require a higher field strength to fully ionize the target within the same number of cycles. By examining the dimensionless parameters $\xi/\omega_1$ and $\hat{\Gamma}/\omega_1$ one can estimate the “experimental parameters” $\ell, n_0$ and $\hat{E}$ in order to meet the desired harmonic pulse duration $T_3$. The practical limit for $T_3$ found in the numerical simulations is about $2\tau$.

If the incident light has already a rather short wavelength (e.g., if light, produced by one of the high-harmonics mechanisms described above are used) then the $2\tau$-limit can be shifted towards the attosecond-domain. However, in that case it would be certainly a challenging task to find the optimal parameters $\ell$ and $n_0$ for a manufacturable thin foil.

One may argue that during the plasma formation process in the thin foil electron-ion-collisions might be important, especially for the relatively low field strengths about $\approx 10^{15}$ W/cm$^2$. Since it is during the plasma formation where the harmonics are produced there might be serious distortions in the spectrum of the transmitted light. In order to
take electron-ion-collisions into account we introduced a collision frequency $\nu_{ei}$ into our 1D PIC-code. This leads to dissipation of energy due to friction of the oscillating charge-sheets (note that in a 1D PIC-code each “computer-particle” represents an actual charge sheet [7]). The dissipated energy is used to determine a “sheet-temperature” which, in turn, enters into $\nu_{ei}$. We found collisions causing mainly distortions at high frequencies but the 3rd and 5th harmonic-peaks were almost unaltered. For stronger incident pulses the effect of collisions is even less.

With our PIC-code we also examined the effects of energy subtraction due to the ionization energy of the target material (according to the model in [6]). The most prominent effect, as far as harmonics generation is concerned, is that ionization gets slowed down slightly (which can be compensated by choosing a higher incident pulse intensity).

It is worth mentioning that the observed effect of harmonics production due to the rise of the electron density in the foil may be used to measure the ionization time of the foil instead of presupposing an ionization rate. This offers an opportunity for checking the validity of ionization models experimentally.

In summary, we have studied the spectrum of a perpendicularly incident laser pulse when transmitted through a rapidly ionizing thin foil, numerically as well as analytically. Low order odd harmonics were observed. The pulse duration of the harmonic radiation is only a few cycles with respect to the frequency of the incident laser light. The pulse length is governed by the risetime of the electron density in the foil and therefore it can be easily tuned through adjusting the peak field strength of the incident pulse. This might be a promising way towards the generation of attosecond pulses.

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FIGURES

FIG. 1. Spectra of the transmitted light for 5 different peak field strengths $E_0$, corresponding to intensities $I = 4.0, 4.8, 6.5 \times 10^{14}$ and $1.1, 1.6 \times 10^{15}$ W/cm$^2$. All other parameters were held constant: wavelength $\lambda_1 = 815$ nm, foil thickness $\ell = \lambda_1/10$, $T = 30$ fs incident sin$^2$-shaped laser pulse, and density $n_0 = n_c = 1.68 \times 10^{21}$ cm$^{-3}$. The higher the field strength the broader are the harmonics peaks in the spectrum.

FIG. 2. Normalized electron density vs. time for the 5 field strengths of Fig. 1. A risetime covering 3 fundamental periods for the weakest pulse and only one cycle for the strongest pulse can be inferred (each stair in the density corresponds to one half cycle).
Fig. 1: D. Bauer et al., “Generation of Ultra-short Light Pulses by...”
strong pulse
short risetime

weak pulse
long risetime

Fig. 2: D. Bauer et al., “Generation of Ultra-short Light Pulses by ...”