Generation of ultrashort light pulses by a rapidly ionizing thin foil

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A thin and dense plasma layer is created when a sufficiently strong 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 rise time 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 that shows how the harmonics are created is introduced. The proposed scheme might be a promising way towards the generation of attosecond pulses. [S1063-651X(98)04208-1]

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I. INTRODUCTION

In recent years several mechanisms generating harmonics of electromagnetic radiation have been discovered. Among these harmonics from gases [1], harmonics from a laser pulse propagating through underdense plasma [2], and harmonics production from the plasma-vacuum boundary when a laser pulse impinges on a solid target [3] 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 extreme ultraviolet ‘‘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 that are produced by a laser pulse focused into a jet of rare gases [4]. Another method makes use of the fact that the efficiency of gas-harmonic generation is sensitive to the ellipticity of the incident laser light [5].

The method to generate an ultrashort *low order* harmonic laser pulse as proposed in this paper is based on the time-dependent electron density of the target material in a laser pulse–solid interaction. The mechanism is thus entirely different from those mentioned above, which are based on phase matching of nonlinear single atom responses [4], relativistic (and thus nonlinear) electron trajectories under the influence of the surrounding plasma [2], or the oscillating vacuum-target interface owing to the $\boldsymbol{v}\times\boldsymbol{B}$ nonlinearity in the Lorentz force. However, the key idea of the method proposed in this paper is similar to the one in Ref. [5], namely, to control the time duration of effective harmonics production by the incident pulse itself. In Ref. [5] it is the ellipticity that is the relevant parameter governing harmonic creation, while in the present paper it is the ionization rate that determines the rise in the free electron density of the target. The harmonic pulse duration is of the order of this rise time and thus can be tuned appropriately by varying the intensity of the incident pulse.

It turns out that although our method is capable of generating very short pulses of third and fifth harmonic light (about two fundamental laser cycles in length), the mechanism is not appropriate for generating particularly *high-order* harmonics efficiently. In a recently published conference proceeding [6] similar studies of ‘‘ionization harmonics’’ are presented, but the authors did not focus on the generation of ultrashort pulses there.

In Sec. II we introduce our simple one-dimensional analytical model to study how the laser pulse propagation is influenced by the ionizing thin foil. In Sec. III we discuss our particle-in-cell simulation results. Finally, we conclude in Sec. IV.

II. ONE-DIMENSIONAL MODELING

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 the 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

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

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 Eq. (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

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

In order to model thin foils we now assume a δ-like current in space [7]. 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 the 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 ℓ (as long as there is no strong electron density gradient across the foil). Inserting the current $j(x,t)$ into Eq. (2) and performing the spatial integration lead to

$$E_r(x,t)=-\frac{\ell}{2c\varepsilon_0}\int dt'\ \Theta(c(t-t')-|x|)\frac{\partial}{\partial t'}j_h(0,t'), \tag{3}$$

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

$$E(x,t)=E_0(x,t)-\frac{\ell}{2c\varepsilon_0}j_h(0,t_{\rm ret}) \tag{4}$$

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

$$j_h(0,t_{\rm ret})=\frac{e^2}{m}n(0,t_{\rm ret})\int_0^{t_{\rm ret}}dt'E(0,t'), \tag{5}$$

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. In addition, 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 [8]. All pulse intensities considered in this article do not cause relativistic electron motion.

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

$$E(x,t)=E_0(x,t)-\xi\left[1-\exp\left(-\int_0^{t_{\rm ret}}dt'\Gamma[E(0,t')]\right)\right]\times\int_0^{t_{\rm ret}}dt'E(0,t'), \tag{6}$$

where

$$\xi=\frac{e^2n_0\ell}{2c\varepsilon_0 m}=\pi\left(\frac{\omega_p}{\omega_1}\right)^2\frac{\ell}{\lambda_1}\omega_1. \tag{7}$$

ω_p is the plasma frequency of the fully ionized target, $\omega_p^2=e^2n_0/\varepsilon_0 m$, and ω_1 and λ_1 are the incident electromagnetic wave's frequency and length, respectively. The dimensionless parameter ξ/ω_1 determines how strongly 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 preionized or ionization is not completed already during the very early part of the pulse, Eq. (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 Eq. (6) in first order in ξ/ω_1 [i.e., we assume a thin foil and iterate Eq. (6) once]. At the position of the foil then

$$E(0,t)=E_0(0,t)-\xi\left[1-\exp\left(-\int_0^t dt'\Gamma[E_0(0,t')]\right)\right]\times\int_0^t dt'E_0(0,t') \tag{8}$$

holds. Here the difficulty is to calculate $\exp\{-\int_0^t dt'\Gamma[E_0(0,t')]\}$. The ionization rate Γ depends on the *absolute value* of the electric field, i.e., the rate has two maxima per fundamental laser cycle. Supposing 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 $\hat{\Gamma}$,

$$\Gamma[|E_0(t)|]=\hat{\Gamma}\left\{\frac{1}{2}a_0+\sum_{n=1}^{\infty}(a_{2n}\cos 2n\omega_1 t+b_{2n}\sin 2n\omega_1 t)\right\}. \tag{9}$$

Since the rate Γ 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 Eq. (9) vanish. This finally leads to

$$E(t)=\hat{E}_0\cos\omega_1 t-\frac{\xi}{\omega_1}\left\{1-\exp(-\alpha_0 t)\times\prod_{n=1}^{\infty}\sum_{m=-\infty}^{\infty}(-i)^m\exp(-i2nm\omega_1 t)I_m(\alpha_n)\right\}\times\hat{E}_0\sin\omega_1 t. \tag{10}$$

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 α_0,α_n are slowly time dependent due to their dependence on $\hat{\Gamma}$. From Eq. (10) we can deduce that by ionization in first order of ξ/ω_1 only *odd harmonics* will be produced: In Eq. (10) the term in curly brackets is composed of even harmonics, but multiplied by $\sin\omega_1 t$, odd harmonics are created.

Harmonic production is enhanced when ξ/ω_1 is increased. Therefore, one expects that 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

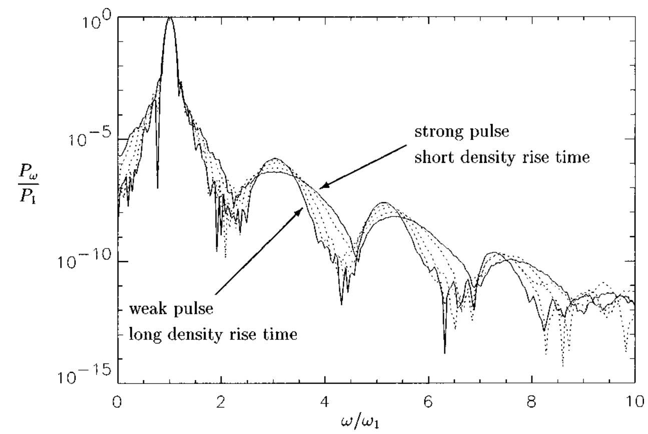

FIG. 1. Spectra of the transmitted light for five different peak field strengths $\hat{E}_0$, corresponding to intensities $I=4.0\times10^{14}$ (solid), 4.8, 6.5×10^{14}, 1.1×10^{15} (dotted), and 1.6×10^{15} W/cm^2 (solid again). All other parameters were held constant: wavelength $\lambda_1=815$ nm, foil thickness $\ell=\lambda_1/10$, $T=30$ fs incident $\sin^2$-shaped laser pulse, and density $n_0=n_c=1.68\times10^{21}$ cm^{-3}. The higher the field strength the broader the harmonics peaks in the spectrum.

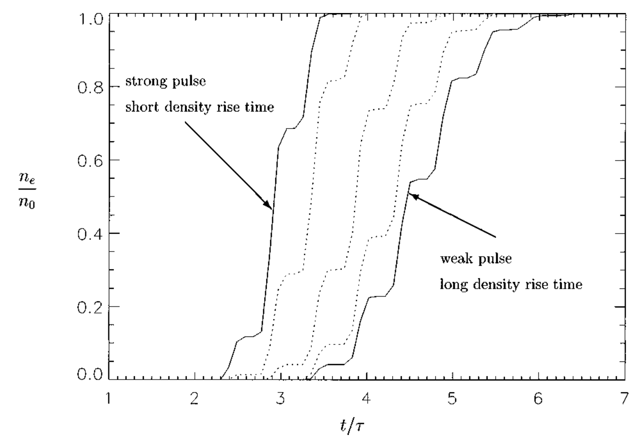

FIG. 2. Normalized electron density vs time for the five field strengths of Fig. 1. A rise time covering three fundamental periods for the weakest pulse and only one cycle for the strongest pulse can be inferred (each stair in the density corresponds to one half cycle).

treatment with respect to ξ/ω_1 preceded this. Furthermore, increasing the density n_0 or the thickness ℓ simply makes the foil less transparent.

III. NUMERICAL RESULTS

A particle-in-cell (PIC) code [one-dimensional in space and two velocity components 1D2V] was used to simulate laser pulse–solid interaction. In order to incorporate ionization the rate equations governing the ionization state of the target were solved during each ''PIC cycle'' [9]. For simplicity only one ionization state with the ionization energy of hydrogen (13.6 eV) was assumed. Landau's tunneling rate [10] was used, which is a reasonable choice for the field strengths and frequencies under consideration. The short rise time of the electron density forces a tiny time step. Usually one fundamental laser cycle (wavelength) was sampled by 1000 temporal (spatial) grid points. 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 five different peak field strengths $\hat{E}_0$, corresponding to intensities $I=4.0$, 4.8, 6.5×10^{14}, 1.1, and 1.6×10^{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., $n_0=n_c=1.68\times10^{21}$ cm^{-3}.

The higher the field strength, the broader the harmonic peaks in the spectrum. The pulse length of the harmonics radiation is closely related to the rise time of the electron density in the foil since as soon as the density remains constant harmonic production will stop. In Fig. 2 the normalized electron density is plotted vs time for the five field strengths of Fig. 1. A rise time covering three 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 rise time is very sensitive to field strength and pulse shape. Increasing the field strength leads to a decreasing density rise time and hence to a shorter harmonics pulse length. However, the conversion efficiency decreases when the field strength of the incident pulse is increased since the nonlinearity switches off too soon. Furthermore, the ''harmonic'' peaks are shifted and asymmetrically broadened if complete ionization occurs within only one fundamental cycle (or even less). In the limit of a steplike behavior of the electron density the spectrum resembles the Fourier transform of the Θ function with no ω_1-harmonic 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 nth harmonic. For the five cases of Fig. 1 one finds for the pulse length of the third harmonic $T_3=3.3$, 3.0, 2.3, 2.0, and 1.9 times the fundamental period $\tau=2\pi/\omega_1$. A lower limit for T_3 certainly is τ itself because a shorter rise time of the electron density leads to a vanishing ω_1 structure in the spectrum. The power in the third harmonic is about 10^{-6} of the fundamental. This conversion efficiency is similar to the one in Ref. [2], while all other methods mentioned in the Introduction are superior as far as power transferred to (short pulse) harmonic radiation is concerned.

One may object that the incident pulse intensity we took in our numerical simulations was already small (at least for ''up-to-date'' short pulse laser systems) so that the third-harmonic pulse with only a millionth of its intensity is not acceptable at all. However, the incident pulse might be a stronger but defocused pulse so that the third-harmonic output, when focused, becomes considerable. In addition, 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 ξ/ω_1 and $\hat{\Gamma}/\omega_1$ one can estimate the ''experimental parameters'' ℓ, 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 2τ. If the incident light has already a rather short wavelength (e.g., if light, produced by one of the high-harmonic mechanisms described above, is used), then the 2τ limit can be shifted towards the attosecond domain. However, in that case it would be certainly a challenging task to find the optimal parameters ℓ and n_0 for a manufacturable thin foil. In recent experiments with thin foil targets [11,12] the thickness ℓ was about 70–100 nm and the electron density n_0 (when fully ionized) was of the order of 5×10^{23} cm^{-3}, i.e., denser by a factor 100 than assumed in our numerical simulations presented so far.

One may wonder if it is really essential to take a *thin* foil. When a sufficiently strong laser pulse impinges on a thick overdense target only a thin plasma layer is created anyway and our analytical treatment in Sec. II should therefore apply as well. We thus expect also from thick targets short harmonic radiation, at least in reflection.

In Fig. 3(a) spectra calculated from time rows of the transverse electric field $E(x,t)$ in front (reflected light), in the middle, and at the rear edge (transmitted light) of a thicker target are presented. The target was one wavelength thick, $\ell = \lambda$, and ten times overcritical, $n_0 = 10 n_c = 1.68 \times 10^{22}$ cm^{-3}. The incident pulse had an intensity $I = 1.6 \times 10^{15}$ $\mathrm{W/cm}^2$. The other parameters were $\lambda_1 = 815$ nm, $T = 30$ fs, and $\sin^2$ in shape again. The spectra have a rather complicated structure owing to the complex spatial and temporal dependence of the free electron density. This can be seen in Fig. 3(b), where the target density is shown as a contour plot vs time and space. During the pulse the target gets fully ionized only in a thin layer at the front. The electron density quickly rolls off and at the rear side not even the critical density is reached. In the spectra we clearly observe the well-known effect that higher frequency radiation penetrates easier the target than light of the fundamental frequency does since for third-harmonic light the target density is only slightly overcritical, while it is even undercritical for the fifth harmonic. This effect was utilized as an experimental method to determine the plasma density [11]. We observe also frequency shifts since the peaks are not located any longer at the odd harmonics' positions. Obviously, thick targets are not as easily accessible by means of simple physical reasoning as it is the case for thin foils where the δ-current model described in Sec. II works well.

Our experiences from several PIC runs with different target densities and thicknesses can be summarized as follows: As soon as there is a density gradient across the foil the spectra of the reflected and transmitted light get distorted (compared to the thin foil spectra in Fig. 1). To avoid this density gradient the optimal ℓn product is about $0.1 \lambda_1 n_c$, which is, with present day technology, not easy to achieve. Using a higher incident pulse intensity in order to ionize the target more homogeneously reduces the density rise time and thus the conversion efficiency.

Apart from these ''technical problems'' one may argue that during the plasma formation process electron-ion collisions might be important, especially for the relatively low field strengths $\approx 10^{15}$ $\mathrm{W/cm}^2$. 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 ν_{ei} into our 1D2V PIC code.

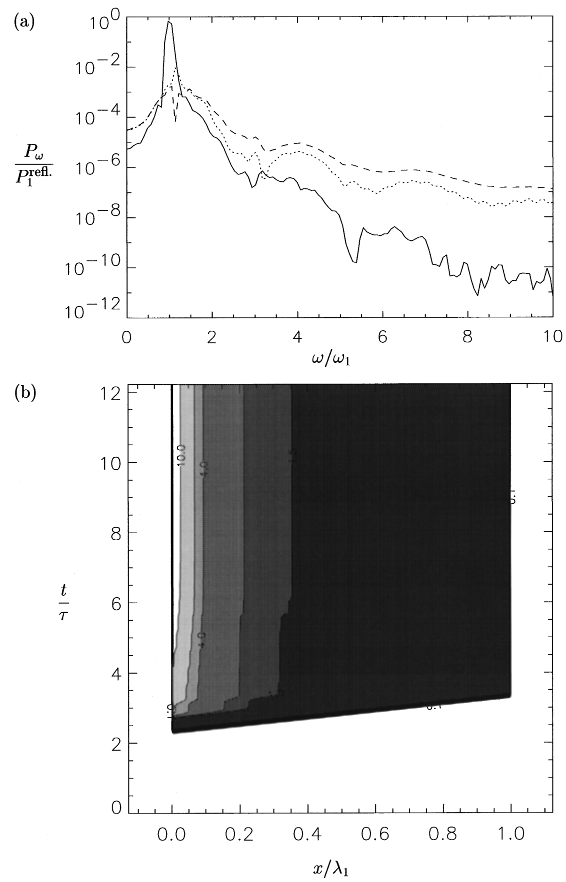

FIG. 3. (a) Spectra taken in front (solid), in the middle (dotted), and at the rear side (dashed) of the $\ell = \lambda_1$ foil, normalized to the fundamental peak of the reflected light. The incident pulse had an intensity $I = 1.6 \times 10^{15}$ $\mathrm{W/cm}^2$. The other parameters were $\lambda_1 = 815$ nm, $T = 30$ fs, and $\sin^2$ in shape. For the fundamental frequency, the plasma layer at the front end is 10 times overcritical while higher frequency light can penetrate more easily. Compared to the clear harmonics structure in Fig. 1 the spectra are rather distorted. The ionization harmonics peaks appear upshifted. (b) Target electron density vs space (scaled in fundamental wavelength) and time (in fundamental cycles). At the front end the target is (for the incident light) 10 times overcritical while at the rear side the target is not fully ionized ($n = n_0/100 = n_c/10$).

This leads to dissipation of energy due to friction of the oscillating charge sheets (note that in a 1D PIC code each ''computerparticle'' represents an actual charge sheet [9]). The dissipated energy is used to determine a ''sheet temperature,'' which in turn enters into ν_{ei}. We found collisions causing mainly distortions at high frequencies but the third and fifth 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 [8]). 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 a thin foil may be used to *measure* the ionization time of the foil instead of presupposing an ionization rate. This would offer an opportunity to check the validity of ionization models experimentally.

IV. CONCLUSION

In summary, we have studied the spectrum of a perpendicularly incident laser pulse when transmitted through a rapidly ionizing foil. A simple analytical method was utilized to show what the underlying mechanism for the generation of short pulse odd harmonic radiation is. 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 rise time of the electron density in the target 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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