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Two-Color Laser High-Harmonic Generation in Cavitated Plasma Wakefields

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Abstract. A method is proposed for producing coherent x-rays via high-harmonic generation using a laser interacting with highly-stripped ions in cavitated plasma wakefields. Two laser pulses of different colors are employed: a long-wavelength pulse for cavitation and a short-wavelength pulse for harmonic generation. This method enables efficient laser harmonic generation in the sub-nm wavelength regime.

INTRODUCTION

Coherent soft x-ray and vacuum ultraviolet light sources are of interest for many applications, and high-harmonic generation (HHG) is a compact method for producing ultrafast, coherent light in this spectral region (for a review, see Ref. [1]). In HHG, an ultrashort laser is focused into a gas, generating multiple harmonics of the laser frequency. Physically, harmonics are generated by bound atomic electrons that tunnel through the effective potential barrier formed by the atom and laser field, oscillate semi-classically in the laser field gaining momentum, and recombine with the parent atom, emitting high-energy photons (odd harmonics of the driving laser frequency).

The highest possible photon energy produced by the nonlinear HHG process $\hbar\omega_{\max}$ is predicted by the HHG cutoff rule [2]

$$\hbar\omega_{\max} \simeq U_i + 3.17\left(m_e c^2 a^2/4\right), \tag{1}$$

where U_i is the ionization potential of the gas and $U_p = m_e c^2 a^2/4$ is the laser ponderomotive potential, with $\omega_{\max}$ the maximum frequency of the emitted photon, $\hbar = h/2\pi$ the reduced Plank constant, m_e the electron mass, and c the speed of light in vacuum. Here a is the normalized quiver momentum of the electron in the laser field, i.e., $a^2 \simeq 7.32 \times 10^{19}\lambda^2[\mu\text{m}]I_L[\text{W/cm}^2]$ for a linear polarized laser pulse, with $\lambda = 2\pi c/\omega$ the laser wavelength and I_L the laser intensity. As indicated by Equation 1, higher photon energy is achieved by increasing the ionization potential and/or increasing the laser intensity. For this reason noble gases (e.g., He with $U_i = 24.6$ eV or Ne with $U_i = 21.6$ eV) are often employed for HHG.

Increasing the laser intensity, however, results in additional ionization and plasma production. The presence of free plasma electrons can inhibit HHG via refraction, thereby reducing the laser intensity. Plasma-induced refraction and defocusing can be prevented by relying on some form of laser pulse guiding, e.g., by propagation of the laser pulse in a gas-filled hollow waveguide [3]. The plasma electrons also produce a phase velocity mismatch between the driving laser and the harmonics, resulting in a reduced interaction length. For a broad laser pulse propagating in an underdense plasma, the phase velocity, normalized to c, is $\beta_p = 1 + \omega_p^2/2\omega^2$, where ω is the light frequency and $\omega_p = k_p c = (4\pi n_e e^2/m_e)^{1/2}$ is the plasma frequency with n_e the plasma electron density. The phase velocity mismatch between the fundamental frequency ω_1 and the N_h-th harmonic is given by $\Delta\beta_p = (\omega_p^2/2\omega_1^2)(1 - 1/N_h^2) \approx (\omega_p^2/2\omega_1^2)$ for high-harmonic numbers $N_h \gg 1$. The resulting phase slippage is proportional to the plasma density $\Delta\beta_p \propto n_e$. This slippage effect can severely limit the conversion efficiency between the laser fundamental and the harmonics, since the effective interaction length is limited to the coherence length L_{coh} between the fundamental and the harmonic (i.e., the propagation length required for the phase mismatch to reach π):

$$L_{\text{coh}} = \frac{2\pi c\omega_1}{\omega_p^2 N_h}. \tag{2}$$

The number of photons will increase quadratically with the coherence length L_{coh}. For example, if we consider 10% ionization of a 10^{18} cm^3 gas with $\lambda_1 = 0.8\,\mu$m light, 0.5 keV harmonics have a coherence length of only $L_{\text{coh}} = 43\,\mu$m. Overcoming this plasma-induced phase slippage has been a challenge, limiting the extent of coherent x-ray sources based on HHG.

One approach to achieving higher HHG photon energies is to use using longer wavelength laser pules to increase the laser intensity (ponderomotive potential) while avoiding plasma production [4]. This method is effective since $U_p \propto a^2 \propto (E_L\lambda_1)^2$, where E_L is the laser electric field, and the ionization rate is determined by the laser electric field. Using a long-wavelength pulse allows a large ponderomotive potential (for higher energy HHG) and a small laser electric field (for reduced plasma production). HHG using a long-wavelength pulse is challenging since the slippage is more severe, i.e., making phase matching more difficult, and owing to reduced HHG efficiency [5]. Since the return time of the electron to the parent atom following ionization is proportional to the laser wavelength $t_{\text{return}} \sim \lambda_1$, i.e., of order a single laser oscillation, longer HHG drive laser wavelength results in increased continuum electron wave-packet spreading, and reduced probability of electron-atom recollision and high harmonic photon emission. The efficiency of HHG has been shown to scale as $\propto \lambda_1^{-4}$ [6].

An alternative approach is to use a high-intensity pulse together with highly-stripped ions containing bound electrons with a large ionization potential. Since the value of U_i for an ion in a high-charge state can be more than an order of magnitude beyond that of the neutral atom, the cutoff energy, in principle, can be extended by an order of magnitude by using high-charge state ions. The use of ions for HHG has been limited by the high density of free plasma electrons that result from preparing the ions in high-charge states.

In Ref. [7] it was proposed to prepare high-charge stage ions by removing free-electrons using the space-charge force of a dense electron beam (or intense laser pulse). In this paper, we propose to use two laser pulses of different colors: a long-wavelength pulse to prepare ions with a bound electron in a high-charge state, and free of plasma electrons, and a short-wavelength pulse for harmonic generation with high efficiency.

This concept is illustrated in Figure 1. Here we consider a long-wavelength pulse propagating in a high-Z gas. The long-wavelength pulse generates some level ionization (preparing the ions in a state with the remaining bound electrons having a large ionization potential), expelling the ionized electrons via the large ponderomotive force, forming an electron free cavity, or bubble. A short-wavelength pulse is delayed with respect to the long-wavelength pulse and co-propagates in the co-moving ion cavity. The short-wavelength pulse has a large electric field and can generate higher laser harmonics, cf. Equation 1, using the remaining bound electrons, with high efficiency owing to its short wavelength.

ION CAVITY FORMATION

Tightly focused laser pulses of sufficient intensity can expel plasma electrons, leaving behind a nearly uniform ion cavity [8–10]. Here we consider an intense laser pulse propagating through a high-Z gas such that the laser ionizes the gas to some charge state, but not completely stripped of bound electrons, and the transverse laser intensity gradient ponderomotively expels the ionized electrons from the region of high laser intensity. The laser intensity required for electron cavitation (also referred to as bubble formation) can be estimated by considering a transverse laser ponderomotive force sufficiently strong to balance the space-charge force created by the ion cavity $\nabla\phi \sim \nabla\gamma_\perp$, where ϕ is the space-charge potential of the ion cavity normalized to m_ec^2/e and $\gamma_\perp^2 = 1 + a_0^2$ is the Lorentz factor of the transverse quiver motion of an electron in a circularly polarized laser pulse. The density perturbation is, via the Poisson equation, of $\delta n/n_0 = n/n_0 - 1 = k_p^{-2}\nabla^2\phi \sim k_p^{-2}\nabla_\perp^2\phi$. Hence cavitation ($\delta n \sim n_0$ to a radius greater than the laser spot size) requires laser intensities satisfying [10],

$$a_0^2/(1 + a_0^2)^{1/2} > \left(k_p w_0\right)^2/4, \tag{3}$$

assuming circular polarization (for linear polarization $a_0^2 \to a_0^2/2$).

The expelled electrons will return to the evacuated channel via the space-charge force provided by the ions. The electron response time to the space-charge separation will be the nonlinear plasma frequency $\omega_p\gamma_\perp^{-1/2}$. Therefore

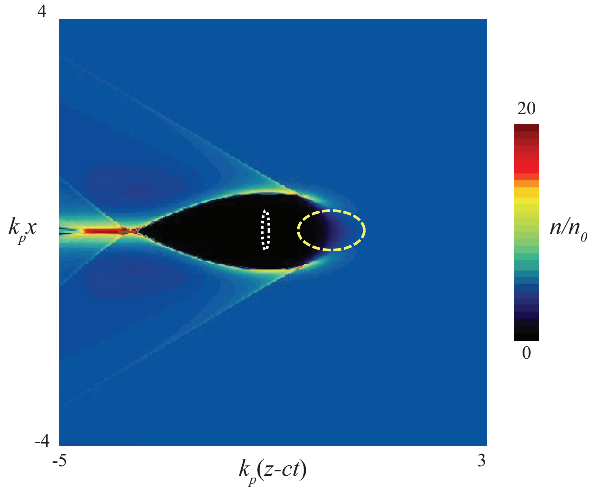

FIGURE 1. Simulation of ion cavity formation using a 10 μm CO_2 laser pulse ($a_0 = 1$, circularly-polarized, 75 μm spot size, and 500 fs RMS duration) propagating in a plasma density of $n_0 = 10^{15}$ cm^{-3}. Color scale indicates electron density, n/n_0. The dashed yellow curve is the envelope of the CO_2 laser pulse. The white dotted curve represents a delayed 0.4 μm ($a_1 = 0.05$, linearly-polarized, 75 μm spot size, 50 fs RMS duration), frequency-doubled Ti:Al_2O_3 pulse for laser harmonic generation.

the short-wavelength HHG-drive pulse should be separated from the long-wavelength pulse for cavity formation by $\tau_{\text{sep}} < \gamma_\perp^{1/2}\omega_p^{-1}$. Note that, since the long-wavelength pulse driving the cavity formation is propagating in plasma, the short-wavelength HHG drive pulse will slip forward with respect to the long-wavelength pulse. However, for typical parameters, this slippage distance is much larger than the coherence length and larger than the Rayleigh length Z_{R} of the focused pulses, $L_{\text{slip}} > Z_{\text{R}} \gg L_{\text{coh}}$.

The phase velocity of the HHG-drive laser will be determined by the laser spot size w_1 and wavenumber: $\beta_p \approx 1 + 2/(w_1^2 k_1^2)$, which will lead to phase slippage with respect to the harmonics (with phase velocity $\beta_p \approx 1$ for $N_h \gg 1$). Therefore, larger laser spots should be used to eliminate this geometrical effect. Ion cavities with large radial dimensions, facilitating large laser spot sizes, can be created in the nonlinear regime. Cavity formation in the nonlinear regime is also accessible using high-density relativistic electron beams, as discussed in Ref. [7].

HIGH-HARMONIC GENERATION

The spectrum of the emitted HHG radiation can be calculated from the dipole moment responsible for the polarization current and radiation emission [1]. The dipole moment $r(t) = \langle\psi|r|\psi\rangle$, where $|\psi\rangle$ is the time-dependent wavefunction for a single electron, can be expressed as [11]

$$r(t) \sim \sum_{t_i} e^{-iS/\hbar} + \text{c.c.}, \tag{4}$$

where the summation is over the ionization times t_i that return to the atom at t, and S is the action along the classical trajectory. For a weakly-relativistic laser field $a^2 \ll 1$, the action is given by the dipole approximation

$$S(t, t_i) = \int_{t_i}^{t} dt' \left\{U_i + m_e c^2 \left[q_1 - a(t')\right]^2/2\right\}, \tag{5}$$

where

$$q_1(t, t_i) = \frac{1}{(t - t_i)} \int_{t_t}^{t} dt' a(t'), \tag{6}$$

and $a(t) = a_1 \cos(\omega_1 t + \varphi)$ with φ a constant laser phase. Here contributions to order $O(a^4)$ as well as space-charge fields in the ion cavity have been neglected. The transverse space-charge field may be neglected compared to the laser field if $(k_1 w_1)(k_p/k_1)^2 \ll a_1$. The longitudinal space-charge force may be neglected if the longitudinal displacement from the atom is sufficient small $k_1 \Delta z \ll \alpha$, where α is the fine structure constant. This condition will be satisfied if $(k_1 L_1)(k_p/k_1)^2 \ll \alpha$, where L_1/c is the HHG drive laser duration and assuming the HHG drive laser is located near the center of the ion cavity.

As an example of this concept, consider cavity formation using a short pulse, 10-μm CO_2 laser pulse. Progress in CO_2 laser technology has opened the possibility of sub-ps pulse durations that would enable efficient (i.e., resonant, with duration of order the plasma period) plasma wake field excitation at plasma densities $\sim 10^{15}$ cm^{-3}, and such laser systems are expected to become available in the next several years [12]. A frequency-doubled Ti:Al_2O_3 laser, delayed and co-propagating in the ion cavity, may be considered to drive HHG. For example, a 10 μm CO_2 laser pulse, with laser parameters $a_0 = 1$ (circularly polarized), 75 μm spot size, and 500 fs RMS duration, propagating in Ar gas of atomic density 1.4×10^{14} cm^{-3} will ionize the gas to Ar^{7+} yielding a plasma electron density of 10^{15} cm^{-3}. This long-wavelength pulse satisfies Equation 3 and will form an electron-free, co-propagating ion cavity (as shown in Figure 1). A linearly-polarized 0.4-μm pulse with $a_1 = 0.05$ propagating in the ion cavity will drive HHG using the bound electron with $U_i = 143.5$ eV. Figure 2 shows the HHG spectrum (Fourier transform of the dipole moment $|r(\omega)|$) calculated using Equation 4–6. Equation 1 predicts the maximum harmonic $\omega_{\max}/\omega_1 = 373$, with maximum photon energy of $\hbar\omega_{\max} = 1.2$ keV.

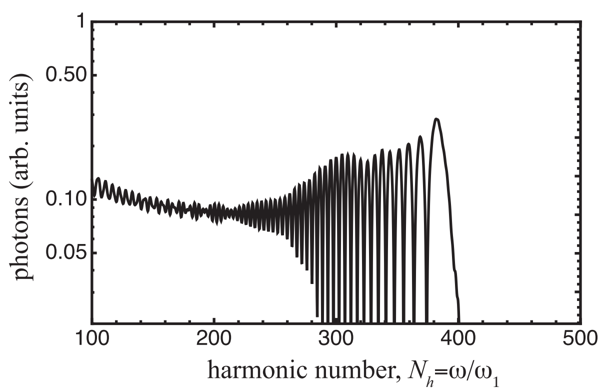

FIGURE 2. HHG spectrum generated in an electron free, Ar^{7+} ion cavity ($U_i = 143.5$ eV) driven by a 0.4-μm laser pulse with $a_1 = 0.05$.

SUMMARY AND DISCUSSION

In this paper we have described a method for producing coherent x-rays via high-harmonic generation using an laser interacting with highly-stripped ions in cavitated plasma wakefields. This method uses two laser pulses of different colors. A long-wavelength pulse propagating in a high-Z gas generates some level ionization (preparing the ions in a state with the remaining bound electrons having a large ionization potential) and expels the ionized electrons via the large ponderomotive force, forming an electron free ion cavity. A short-wavelength pulse is delayed with respect to the long-wavelength pulse and co-propagates in the co-moving ion cavity. The short-wavelength pulse has a large electric field and can generate higher laser harmonics, using the remaining bound electrons, with high efficiency. This method enables efficient laser harmonic generation in the sub-nm regime.

Using this method, the laser intensity may be increased to extend the maximum HHG photon energy, until the laser drives the electron motion to relativistic velocities. At relativistic laser intensities, the magnetic field of the laser strongly influences the electron motion via the nonlinear $v \times B$ Lorentz force, and this force produces motion of the electron in the direction of laser propagation. As a result, the electron wave packet will not return to the atom and hence there will be no recombination and harmonic emission [13].

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