Strong increasing the efficiency of high-order harmonic generation using counter-propagating laser pulses

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ABSTRACT

We present results of computer simulations showing dramatic increases in the production and intensity of high-order harmonics (in the cutoff region) in rare gases interacting with counter-propagating pulses. Using two different models of medium, classical anharmonic oscillators (for Ne, Ar, Kr) as well as quantum oscillators (for Ne^+, Ar^+, Kr^+), we demonstrate the increase of efficiency by four and by six orders in these models, respectively. This is at least by two orders of magnitude more than obtained with other methods.

Keywords: High-order harmonics, rare gases, efficiency, electronic Raman scattering

1. INTRODUCTION

High-order harmonic generation (HHG) by a strong laser field in a gaseous medium,(see, e.g. [1-4]) is an attractive source for experiments and practical applications requiring coherent extreme ultraviolet (XUV) and x-ray emission. However, the efficiency of converting the energy from the driving laser pulse to HHG has reached about 10^{-8}(at 20 nm), and less than 10^{-10}(at < 10 nm) [5,6]. HHG was studied for different media and their attributes as well as parameters of driven laser pulse (see, for example, [1-9] and references therein) to increase the photon number. It was found some improvements to the efficiency of HHG can be made by decreasing the laser pulse duration [2-4] or by designing laser pulse temporal profiles to enhance the atomic response [9]. The efficiency of converting laser field into HHG is mainly limited by phase mismatches (see, e.g., [5, 10-13] and references therein). Tamaki at al. [14] for phase matching of HHG used a technique of self-guided beam, and demonstrated the increase in the conversion efficiency by 40 times around the 49th harmonic (in the cutoff region). Recently, Tempea et al. [15] proposed to use mechanism nonadiabatic self-phase matching (NSFM) to enhance HHG above 1 keV by 4 orders of magnitude. However, NSFM works predominantly for wavelengths in x-ray regime, moreover, very high peak intensities ($I_0 = 1.5 \times 10^{16} W/cm^2$) and very short laser pulse duration (5 fs) are required [15]. Improvements to the conversion efficiency of HHG through quasi phase matching (QPM) [16] based on a modulated density of neutral and ionized gases, were developed and it was demonstrate theoretically [10,13] that harmonic intensity might raise about two orders. However, such method [10] would compensate for geometrical phase mismatches, but it could not deal with index-related phase mismatches, assuming them to be inseparably linked with the density modulation. Furthermore, the manipulation of the atomic density to create a good enough periodic gas vacuum structure in the focus poses some technical challenges. An alternative approach for QPM at HHG was proposed in [11]. Rather than attempting to modulate spatially either the density of the atomic medium or the density of free electrons, the approach uses counter-propagating pulses (CPP) to disrupt the phase matching microscopically in specific regions to frustrate harmonic production in selected zones with undesirable phase, while remaining regions then constructively interfere, and harmonic production is expected to increase. The CPP-based QPM at HHG was studied [11] for the simplest phenomenological model of HHG under the assumption of perturbation theory and for relatively weak CPP (one hundredth the intensity) without the investigations of the nonlinear dynamics of propagation in medium and its polarization.

In this paper we report the results of numerical investigations the nonlinear dynamics of laser pulses and HHG, including CPP with the same peak intensity, as exact solutions of self-consistent equations, namely Maxwell's equations for laser fields and equations for two different models of gaseous media, as ensembles of (I) classical anharmonic

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ICONO 2001: Ultrafast Phenomena and Strong Laser Fields, Vyacheslav M. Gordienko, Anatoly A. Afanas'ev, Vladimir V. Shuvalov, Editors, Proceedings of SPIE Vol. 4752 (2002) © 2002 SPIE · 0277-786X/02/$15.00

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oscillators [17] for atoms as well as (II) two-level systems relating to fine structure of ground state of their ions [18]. The most prominent (but needing significant computational resources) method that used to describe the atom response at HHG was the integration of the time-dependent Schrödinger equation for an atom in the single active electron approximation [19,20]. However, a number of simplified model, such as classical anharmonic oscillators (see, e.g. [7, 17]), two-level systems under direct transitions (see, e.g. [21]), two-level systems under Raman transitions [18], give results that are qualitatively similar to those of the more exact calculation [19,20] . In addition, recently it was shown [22] that ionizing two-level system in which the ionized population is considered as classical particles moving inside the electric fild, give results that are in reasonable agreement with [19,20]. It is well known [23] that emission from ions must be invoked to explain the HHG. The theoretical model for HHG based on stimulated electronic Raman scattering (SERS) at the fine structure of singly ionised noble gases was studied [18] and shown to be in reasonable agreement with attributes of HHG. To show that our results do not depend qualitatively on the model of the medium, we studied the HHG, at single pulse as well as at CPP, on the two different models previously mentioned. We show that both models (I) and (II) give similar qualitatively results for CPP: the increase of a number of harmonics, the increase of intensities of higher harmonics in plateau (i.e. the harmonics of more higher order) by several orders of magnitudes, and the decrease of intensities of lower harmonics.

2. CLASSICAL ANHARMONIC OSCILLATORS AND HHG AT CPP

In this paper, to describe the laser field polarized linearly along the x-axis and propagating on the z-axis, we start from one-dimension (1D) Maxwell's equations, which we write in gaussian units in the form

$$\frac{1}{c}\frac{\partial E_x}{\partial t} = -\frac{\partial H_y}{\partial z} - \frac{4\pi}{c}\frac{\partial P}{\partial t}; \quad \frac{1}{c}\frac{\partial H_y}{\partial t} = -\frac{\partial E_x}{\partial z}, \tag{1}$$

where, for (I) model of medium (atoms of noble gases as classical anharmonic oscillators) , the polarization is $P = -N_{atom}eq$, N_{atom} is the density of polarizable atoms, $-e$ is the electronic charge, q is the displacement from equilibrium of an electron in the atom. The equation of motion for the anharmonic oscillator of centrosymmetric media, is [7,17,24]

$$\ddot{q} + \Gamma\dot{q} + \omega_{ao}^2 q = -em_e E_x(t,z) - \beta q^3 \tag{2}$$

where Γ, ω_{ao}, β are a damping term, a characteristic frequency, and the parameter of the anharmonicity, respectively. As the model based on Eq. (2) for HHG drived by laser pulse with frequency ω, the ratio ω/ω_{ao} is considered to be of the order of 0.1 [17], corresponding to the typical ratio between the photon energy of a terawatt laser using usually (Ti:sapphire), and the ionization potential of a noble gas. It should be noted, that in this paper we don't touch relativistic intensities ($10^{18}W/cm^2$) of laser pulses, and thus the influence of the magnetic field on HHG is neglected.

2.1. Results for (I) model of medium

We used standard fourth-order Runge-Kutta algorithms to get the solutions of Eq. (2). And the spectra of HHG obtained by us agree with the results [17] at same conditions. It is necessary to stress that the HHG in process of propagation of laser pulse, as well as collective effects and phase matching ones, was not studied in [17]. For numerical integrating of 1D Maxwell's equations (1) we use well known algorithms [25]. We have performed the numerical simulations on the model described by the system of the self-consistent Eqs. (1)-(2) for different durations and intensities of laser pulses, and we found that the model also show a rise to the efficiency of HHG with decreasing of the laser pulse duration. These results are in reasonably good qualitative agreement with the experimentally deduced regularities on HHG, as well as with the known behaviour of other models (see, for example [17,26]). Because, one of the ways to increase the efficiency of HHG, as it was mentioned above, is to decrease of the laser pulse duration, (the best experimental results being obtained with 26 fs and less pulse durations [2-4]), we use the initial $\tau_0 = 20$ fs FWHM pulse with a Gaussian envelope. These 20 fs pulses from Ti:sapphire laser (wavelength $\lambda \sim 800$ nm), contain only 8 optical cycles FWHM.

Figure 1(a) shows the 20-fs pulse high harmonic spectrum after passing 6 μm in medium with $N_{atom} = 4 \times 10^{18} atoms/cm^3$, and at the initial intensity of the pulse $I_0 = 1.6 \times 10^{14} W/cm^2$. We used in Eq. (2) the same parameter of the anharmonicity $\beta = 2 \times 10^{35} (\mathring{A}\ s)^{-2}$ in Kr that was obtained in [7] from experimental estimations, at $\omega_0/\omega_{ao} = 0.1$ (ω_0 is initial frequency of the pulse), and without damping ($\Gamma = 0$). This spectrum in Fig. 1(a) has the characteristic cutoff in the area of 35th harmonic and also the so-called "hyper-Raman" peaks especially

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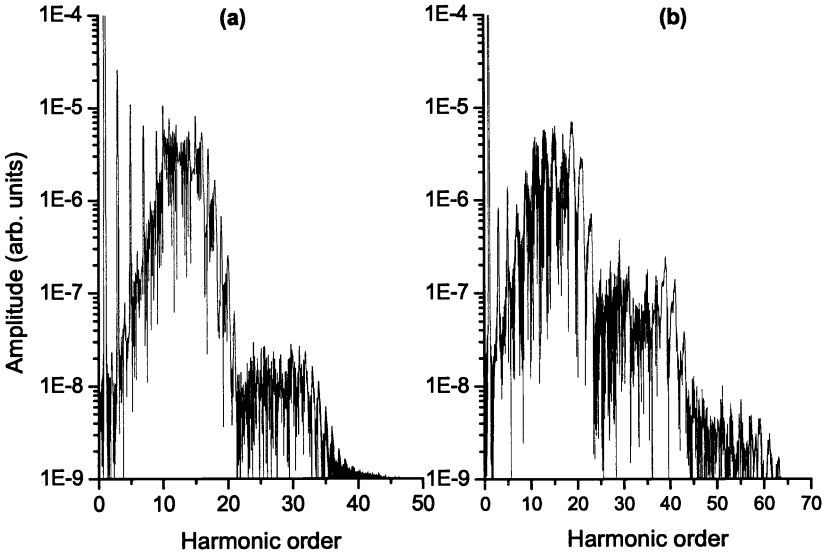


Figure 1. Amplitude spectra vs ω/ω_0 of 20 fs FWHM Gaussian pulse, $\lambda = 800$ nm fundamental, of peak intensity $1.6\times10^{14}W/cm^2$, passed 6 μm in medium Duffing anharmonic oscillators with $N_{atom} = 4 \times 10^{18} atoms/cm^3$, $\omega_0/\omega_{ao} = 0.1$, and parameter of the anharmonicity $\beta = 2 \times 10^{35}(\AA\ s)^{-2}$ typical for Kr, (a) without using the 160 fs FWHM Gaussian counter-propagating pulse and (b) with using one.

around harmonics with orders higher than 7. Hyper-Raman peaks in spectra of harmonics, both in the classical (anharmonic oscillators) and quantum case, was discussed in [17, and Refs. therein] and was shown to be connected with dynamical coherence effects. Figure 1(b) shows the spectrum of the same pulse, but after passing the same medium in the presence of a 160-fs counter-propagating pulse with the same I_0. A comparison Figs. 1(a) and 1(b) reveals that the CPP alters radically the spectrum of the 20-fs pulse: (A) the increase in harmonic photon energy by more than 60% (the 39th harmonic is the maximum order when the CPP is not used (Fig. 1(a)), and the 65th harmonic is the maximum order when the CPP is used (Fig. 1(b))); (B) the increase of the amplitudes of higher harmonics (for instance, the amplitudes of the 39th and 37th harmonics are increased by two orders of magnitudes ,that is, the intensities of these harmonics are increased by four orders of magnitudes); (C) the decrease of amplitudes of lower harmonics. We performed the computer simulations for different durations of the CPP, but give, in Figs.1, the results with 160-fs CPP. This longer CPP is able to interact with the forward-traveling pulse through comparatively longer time and distanse. Our simulations also shown that if the duration of the CPP is decreased then HHG is increased from this CPP while the efficiency of HHG from the 20-fs pulse is fallen. The latter is easy to understand, taking into consideration that the decrease in the duration of the CPP lead to increasing HHG from this pulse (opposite to propagating 20-fs pulse), and, at the same time, an interaction length for the both pulses is shortened, decreasing the efficiency of converting 20-fs field into high harmonics.

3. HHG AT THE FINE STRUCTURE OF SINGLY IONISED NOBLE GASES AND USING CPP

It is well known that at intensities $I_0 \sim 10^{14}W/cm^2$ and higher ones (and wavelength $\sim$ 800 nm), the ionization of noble gases occurs at every half optical cycle via a tunneling process. For example, at the intensity of $3 \times 10^{14}W/cm^2$ approximately half argon atoms will be ionized by the peak of the 27-fs pulse [26]. In addition, Krause et al. [23] have shown that at the intensities higher $10^{14}W/cm^2$, ions can contribute significantly to the tharmonic spectra.

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3.1. Fine structure of singly ionised noble gases as a two-level system for SERS and HHG

Next we turn to the above model for HHG that based on SERS [18] at fine structure of singly ionised noble gases.The SERS can be realized at transitions between fine structure $^2P_{3/2}$-$^2P_{1/2}$ levels of the Ne^+, Ar^+, Kr^+, Xe^+ ions. For example, in Ne^+ the ground state $^2P_{3/2}$ and the neighboring excited state $^2P_{1/2}$ form a two-level system where the frequency transition is $\Omega/(2\pi c) = 780, 4cm^{-1}$, and the transition are forbidden by parity in the electro-dipole approximation for one-quantum process, but permited for two-quantum prosess like SESS. And all other levels are removed on $\Delta > 217047$ cm^{-1} [27]. A similar situation exists in the ions of the other noble gases. Seeing that 20-fs pulse have spectral band no less then 1668 cm^{-1} the transition frequency Ω is contained within the spectrum of the pulse, i.e., its frequency range contains infinite set of components which are comparable on amplitude and satisfy the condition of the Raman resonance. This causes their effective stimulated Raman self-scattering (SRSS)[18]. The frequency Ω is much less than the central frequency ω_o of the initial laser pulse field $\mathcal{E}$. The average value of a polarizability $< \alpha >= Sp(\alpha\rho)$ and the difference of the level populations $w = \rho_{22} - \rho_{11}$, where α is the operator of polarizability of the medium, and ρ is the density matrix, are described by the equations [18,28]:

$$\frac{\partial^2 < \alpha >}{\partial t^2} + \frac{2}{T_2} \cdot \frac{\partial < \alpha >}{\partial t} + \Omega^2 < \alpha > = - \mid \alpha_{12} \mid^2 \frac{\Omega}{\hbar} \cdot \mathcal{E}^2 \cdot w \ , \tag{3}$$

$$\frac{\partial w}{\partial t} + \frac{(w - w_o)}{T_1} = \frac{1}{\hbar\Omega} \cdot \mathcal{E}^2 \cdot (\frac{\partial < \alpha >}{\partial t} + \frac{< \alpha >}{T_2}) \ , \tag{4}$$

where $\mid \alpha_{12} \mid$ is a value of the matrix element of the polarizability corresponding to the Raman transition with frequency Ω. The parameter T_2 is the dephasing time or the time of Raman coherence, and T_1 is the time of population relaxation. Since we will consider coherent interactions only ($\tau_0 \ll T_2, T_1$), we can neglect the terms related to the relaxation times and in Eqs. (3) and (4). The nonlinear dipole moment related to SRSS is $d = \alpha \cdot \mathcal{E}$. In this case, the nonlinear contribution to the polarization of the medium is given by $P = N_{ion} \cdot < \alpha > \cdot \mathcal{E}$, where N_{ion} is the number density of ions of noble gas. The 1D Maxwell's equations (1) using this expression for the polarization with equations (3) and (4) (describing the medium as an ensemble of the two-level oscillators under two-quantum transitions of Raman type) forms the system of the self-consistent equations for our (II) model.

3.2. Results for (II) model of medium

The results of our computer simulation for this model are shown in Fig. 2. The initial pulse and CPP have the parameters identical to those used for the (I) model above. The 20-fs pulse passed through the same 6 μm distance in medium with $N_{ion} = 3 \times 10^{18} ions/cm^3$. Low value of the matrix element $\mid \alpha_{12} \mid$ of the polarizability can be estimated from experimental data on multiphoton ionization in noble gases. For example, in experiments [29] with initial pulses $\tau_0 = 1$ ps and intensities $I_o \sim 10^{13} W/cm^2$ an effective change of the populations of the states $^2P_{3/2}$ and $^2P_{1/2}$ in Kr^+ ions was observed. This means that for the corresponding two-photon Rabi frequency we can estimate $\Omega_{2phR} \gg 1/\tau_0$, and taking into consideration that $\mid \alpha_{12} \mid \sim \Omega_{2phR} \cdot \hbar \cdot c/I_o$ [21], for $\Omega_{2phR} = 3 \times 10^{12} s^{-1}$ and $I_o \sim 10^{13} W/cm^2$ [29] we obtain $\mid \alpha_{12} \mid \sim 10^{-24} cm^3$. We used this value $\mid \alpha_{12} \mid$ in the calculations illustrated in Figs.2 and 3. Qualitatively the results illustrated in Fig. 2 are similar to those exhibited above by (I) model , but in this case we have: (A) the increase in harmonic photon energy by more than 2 times (the 39th harmonic is the maximum when the CPP was not used (Fig. 2(a)), and the plateau-cutoff is in the region of the 85th harmonic when the CPP was used (Fig. 2(b))); (B) the increase of the amplitudes of higher harmonics (for instance, the amplitudes of the 39th and 37th harmonics are increased by about three orders of magnitudes ,that is, the intensities of these harmonics are increased by about six orders of magnitudes; (C) the decrease of the amplitudes of the lower harmonics (up to 23rd). It is seen that the efficiency of HHG is much more always in the case (II)(Fig. 2) of quantum oscillators when compared to the case (I) (Fig. 1) of classical anharmonic oscillators. This is easy to understand in view of the fact that in this classical anharmonic oscillator (Eq. 2) the third-order nonlinearity is taken into account only, and in the Eqs. 3 and 4 for the two-level system all the higher-order nonlinearitеis are taken into account automatically (the difference of the level populations w is the intensity-dependent function (see Eq. 4)).

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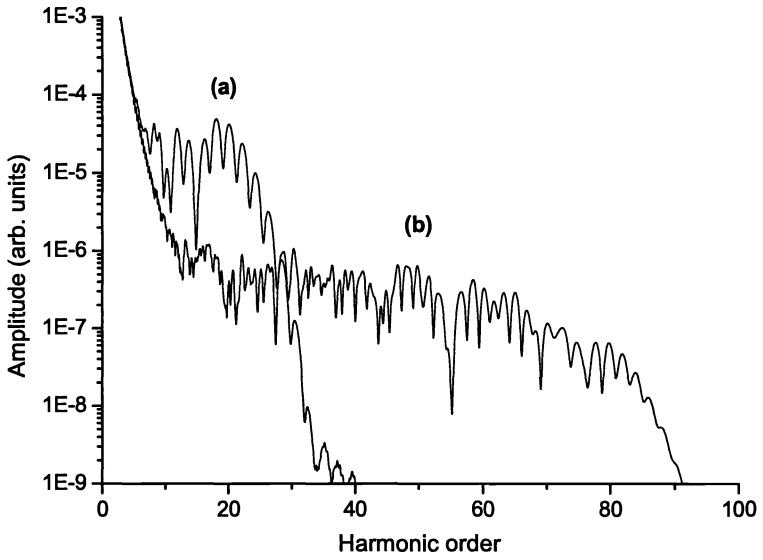


Figure 2. Amplitude spectra vs ω/ω_0 of 20 fs pulse, with the same parameters that in Fig. 1, passed under SERS at the fine structure, $\Omega/\omega_0 = 0.1$, of ions with $N_{ion} = 3 \times 10^{18} ions/cm^3$, and $\mid \alpha_{12} \mid = 10^{-24} cm^3$, (a) without using the 160 fs counter-propagating pulse and (b) with using one.

4. CONCLUSIONS

We have demonstrated, using two different models of media, classical anharmonic oscillators (for Ne, Ar, Kr) as well as quantum oscillators (for Ne^+, Ar^+, Kr^+), that the use of counter-propagating pulses leads to an increase in the generating efficiency of the highest harmonics at least by more than four orders of magnitude of their intensities (in the cutoff region) as well as allows to reach higher photon energies.

5. ACKNOWLEDGMENTS

This research was supported in part by INTAS, Projects No. 97-1058 and No. 97-2018, and by Belarus Basic Reseuch Foundation (Grant No. F98-319).

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