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Letter

# Extending the high-order harmonic generation cutoff by means of self-phase-modulated chirped pulses

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**Abstract**

In this letter we propose a complementary approach to extend the cutoff in high-order harmonic generation (HHG) spectra beyond the well established limits. Inspired by techniques normally used in the compression of ultrashort pulses and supercontinuum generation, we show this extension can be achieved by means of a nonlinear phenomenon known as self-phase-modulation (SPM). We demonstrated that relatively long optical pulses, around 100 fs full-width half maximum (FWHM), non linearly chirped by SPM, are able to produce a considerable extension in the HHG cutoff. We have also shown it is possible control this extension by setting the length of the nonlinear medium. Our study was supported by the numerical integration of the time-dependent Schrödinger equation joint with a complete classical analysis of the electron dynamic. Our approach can be considered as an alternative to the utilization of optical parametric amplification (OPA) and it can be easily implemented in usual facilities with femtosecond laser systems. This technique also preserves the harmonic yield in the zone of the plateau delimited by $I_p + 3.17Up$ law, even when the driven pulses contain larger wavelength components.

Keywords: high harmonic generation (HHG), self phase modulation (SPM), chirped pulses

(Some figures may appear in colour only in the online journal)

In recent years there has been a great interest in the laser community to obtain and handle coherent radiation in the XUV soft-x-ray spectral region. This fact can be understood since this radiation has countless applications in biology, biochemistry, high-resolution spectroscopy, among others [1–3]. One of the subjects, in which it has acquired a major impact, is the generation of attosecond (1 as $= 10^{-18}$ s) pulses, i.e. laser pulses with durations in the sub-fs/as range. These short bursts of light configure an instrumental tool to study the physical phenomena in their natural time scale [4] and represent a key aspect in the fundamental progress in atomic and molecular physics [5]. High-order harmonics generation in atoms (HHG)

 

allows us to obtain radiation with the characteristics described above. HHG is a nonlinear phenomenon that takes place when an intense and ultrashort pulse interacts with matter, i.e. atom and molecules, generally in their gaseous phase [5, 6]. This process can be easily explained by invoking the three step model [7, 8], but it should be endorsed with a quantum mechanical description [9]. Both the classical and quantum approaches predict that the maximum photon energy (cutoff) can be obtained by employing [7] (atomic units will be used unless otherwise stated):

$$E_{\text{cut-off}} = I_p + 3.17U_p \tag{1}$$

where $I_P$ is the ionization potential of the atom or molecule under consideration. $U_P$ is the so-called ponderomotive energy, given by $U_P = \frac{E_0^2}{4\omega_0^2} \propto I\lambda^2$, being $E_0$ the peak amplitude and $\omega_0$ and $\lambda$ the central frequency and laser wavelength, respectively. According to equation (1) there are two possible ways to extend the harmonic cutoff (i.e. to achieve high energetic photons), namely, to increase the laser wavelength or the laser intensity. However, it must be mentioned that, a longer wavelength will imply a sudden decrease in the HHG efficiency, governed by the well known $\lambda^{-5.5}$ law [10, 11]. The reasons of this reduction can be traced out in the spreading of the electron wave-packet effect also observed when the intensity of driven pulses are increased. In addition this increment will also entail a loss of efficiency as a consequence of the barrier suppression [12]. The standard form to generate HHG is through commercial Ti-Sapphire lasers based on the chirp pulse amplifier technique [13], whose pulses ranging from few to several femtoseconds at a wavelength $\lambda = 800$ nm with an optical cycle of 2.7 fs, focused on a gas jet [6]. There are peer reviewed articles showing several strategies to extend the cutoff region beyond the $3.17U_P$ value with femtosecond laser pulses. However, only a few experimental approaches have reported appreciable cutoff extension with ultrashort pulses [14–17]. In turn, from a theoretical point of view different ways have been proposed to achieve an extended HHG cutoff, e.g. by employing spatial-temporal field synthesized by the mean of the plasmonic effect on nanoparticles fields [18, 19] or by using nonlinear chirped pulses [20, 21].

At this point it must be mentioned the pioneering work of Tong *et al*, where only the chirp effects are analyzed on the shape of the harmonics peak but not in the cutoff, since any extension effect can be appreciated [22]. In this letter we put forward a new route to extend the HHG cutoff beyond the $3.17U_P$ limit. In order to obtain this extension we will exploit the well-known nonlinear phenomenon called self-phase-modulation (SPM) for the fundamental pulse [23]. This strategy is commonly used in the compression of ultrashort pulses and supercontinuum generation [24, 25]. In a previous work [26] we showed that the chirp that allows the cutoff to be extended, must be generated by a nonlinear phenomena because it introduces new spectral components in the driven pulse.

In the region where many materials are transparent and for high enough laser intensities, the refractive index depends nonlinearly on the propagating field (this phenomenon is the so-called self-phase modulation (SPM)) which is used to change the frequency components of the pulse. The described dependence affects the optical pulse phase that propagates through the medium. One of the main parameters that characterize this phenomenon is the $n_2$ index, called the nonlinear index coefficient. It represents the strength of the coupling between the electric field and the refractive index [23]. The dependence of the refractive index with the field intensity can be expressed as:

$$n(I) = n_0 + n_2 I(t). \tag{2}$$

When SPM occurs (neglecting other nonlinear effects), we have supposed that the interaction takes place in a length shorter than the dispersion length $L_{\text{D}}$, (this parameter reflects the typical length where the pulse is temporally stretched in a noticeable form) and the response of the electric field with the medium is instantaneous. This means that the response time of the material should be about the length of the pulse [23]. Under these assumptions, the pulse phase $\phi(t, z)$ after passing through the medium at a distance $z$ can be written as:

$$\phi(t, z) = \omega_0 t - \frac{n_2 k_0 z}{n_0} I(t), \tag{3}$$

where $n_0$ the refractive index of the material, $k_0$ is the wave number and $\omega_0$ is the central frequency of the input laser pulse. In order to characterize the SPM it is convenient to introduce a nonlinear interaction length

$$L_{\text{NL}} = \frac{n_0}{n_2 k_0 I_{0m}} \tag{4}$$

where $I_{0m}$ is the maximum laser intensity. As we have shown, SPM allows us to get a time-dependent phase and this dependence is strongly determined by the pulse shape, see equation (3). An interesting fact is that the SPM does not modify the temporal shape of the pulse that propagates through a nonlinear medium. For the previous analysis we have only considered a quasi uniform spatial intensity distribution.

In the following we will study the HHG driven by femtosecond pulses once they have propagated through a nonlinear material and were distorted by the SPM phenomenon. Because our laser pulses will be linearly polarized and due to its large duration (100 fs FWHM) it is legitimate to simulate the HHG spectra by using the time-dependent Schrödinger equation in reduced dimensions (1D-TDSE) and to complement the quantum mechanical results by a classical analysis of the electron trajectories [27]. The input pulse employed to perform the simulation was a Gaussian one, with 100 fs FWHM, wavelength $\lambda = 800$ nm and a model argon atom as the active medium. The laser intensity was set to $I_2 = 10^{14}$ W cm$^{-2}$.

For this reason, only certain materials, that behave as a Kerr medium, are suitable to produce SPM in this experiment, which possess $n_2$ on the order of $n_2 \sim 10^{-16}$ cm$^2$ W$^{-1}$ [23]. Other materials to be mentioned are photonics crystal fibers which bear high intensities, based on the fact that some of them have zero dispersion in the range of 800 nm [28]. It is worthwhile to mention that in a previous work, similar considerations were used to describe this propagation phenomena despite the fact that they use a smaller pulse width [29].

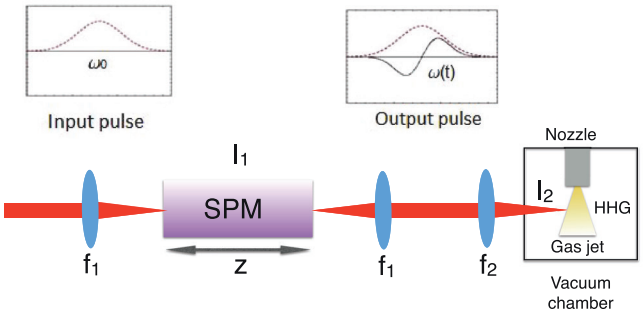


**Figure 1.** Potential experimental set-up proposed for SPM and HHG. Each inset contains the envelope of the driven pulse as a dashed line; in the left one the frequency $\omega_0$ is shown as the solid line and in the right one $\omega(t)$ (equation (7)).

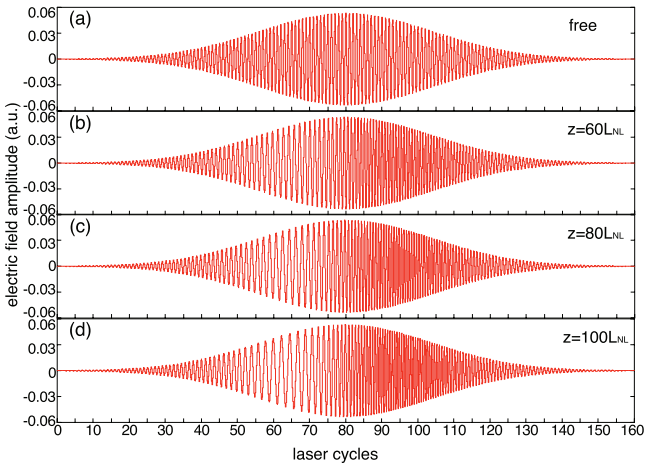


**Figure 2.** Free case (a) and chirped pulses (b)–(d) resulting from the SPM for three different values of $L_{\rm NL}$. The longer periods (i.e. long wavelengths) appear in the first half of the pulse (up-chirp). Note that the laser electric field amplitude, i.e. laser intensity, remains invariant in all cases.

In figure 1 we show our proposed experimental setup. The input pulse is focused onto the material to achieve an intensity $I_1$, high enough to generate SPM in the incoming pulse, as established by equation (3).

Once the pulse is propagated at a distance $z$ into the material, the beam is collimated by using a lens ($f_1$). As a consequence we obtain a pulse whose envelope remains unchanged, but its phase is modified following equation (3). The pulse with the reconfigured phase is then focused by a second lens( $f_2$), whose focal distance is short enough to obtain a high pulse intensity into the gas jet. Under these assumptions, the obtained intensity $I_2$ will be able to drive HHG in a noble gas atom.

The driving laser pulse used in our simulations can be written as

$$E(t,z) = E_0 \exp\left[-2\ln 2\left(\frac{t}{\tau_0}\right)^2\right]\cos[\phi(t,z)], \qquad (5)$$

where $E_0$ is the peak electric field amplitude, $\tau_0$ is FWHM and $\phi(t,z)$ is the phase defined in equation (3). $\phi(t,z)$ can be rewritten as (in terms of the definition of the nonlinear interaction length $L_{\rm NL}$, equation (4)):

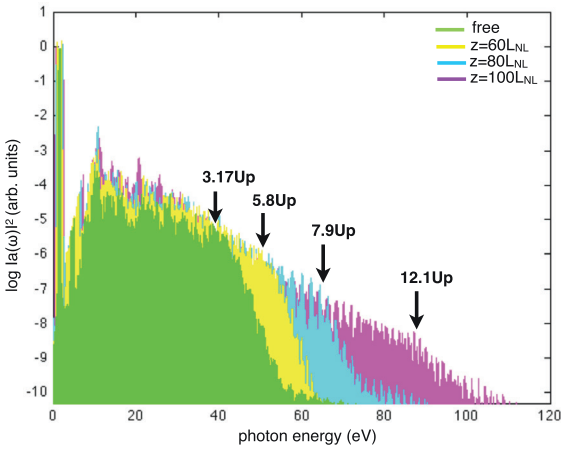


**Figure 3.** Spectra in argon extracted by the numerical integration of the 1D TDSE using the corresponding driving laser pulses of figure 2. The different arrows show the estimated harmonic cutoffs predicted by the semiclassical model (see figure 4).

$$\phi(t,z) = \omega_0 t - \frac{z}{L_{\rm NL}}\left|\exp\left[-2\ln 2\left(\frac{t}{\tau_0}\right)^2\right]\right|^2. \qquad (6)$$

Equation (6) is a convenient form to write the phase $\phi(t,z)$, because it explicitly shows the functional dependence with $L_{\rm NL}$. In addition, we could write $z$ as $z = nL_{\rm NL}$. In our simulations we use three propagation lengths, $z = 60L_{\rm NL}$, $z = 80L_{\rm NL}$, $z = 100L_{\rm NL}$, obtaining in this way the different chirped pulses whose features can be appreciated in figure 2. Note that the chirp induced in the pulse makes the frequency increase in time. This kind of chirp is known in the literature as an *up chirp* (i.e. the longer wavelengths appear before the shorter ones along the pulse). Our quantum simulations performed by numerical integration of the 1D-TDSE provide the single atom spectra shown in figure 3. For a complete understanding a phase matching effect study should be included but this task falls out of the scope of this letter. In figure 3 a clear increasing on the HHG cutoff can be observed when the propagation length is increased, or in equivalent form, as a function of the pulse chirp. The spectra show a clear decrease on the HHG efficiency for long cutoff extensions. However all spectra present one property in common, the efficiencies are similar in all cases in the zone of the harmonic plateau delimited by the $I_p + 3.17U_p$ law. Even a slight increase of the harmonic yield can be appreciated correspondingly to an increase of the chirp. This fact can be attributed to the shorter wavelengths present in the chirped pulses. On the other hand, the wavepacket spreading suffered by the electrons ionized by short frequencies is considerably reduced, making the electron recombination more efficient. The different cutoff extensions are originated by the largest wavelengths also contained in the chirped pulses. The pulses in figure 2 increase the largest wavelengths in agreement with the increase of the nonlinear interaction length. The classical law $I\lambda^2$ establishes that the larger the wavelength, the higher the cutoff. As the pulse propagates with an intensity of $I_1$ through the nonlinear medium, new frequencies (or wavelengths) are added to the pulse spectrum, so it results in a broadening of the pulse

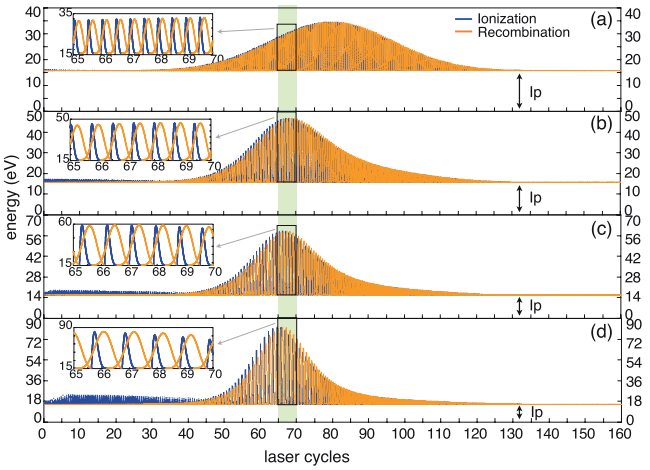


**Figure 4.** (a) Classical ionization/recombination electron energies generated by the pulses of figures 2(a)–(d), respectively. $I_p$ is the ionization potential and correponds to 15.7 eV.

bandwidth, characteristic of the SPM process. As we have shown previously, the chirped pulses show higher and lower frequencies (see figure 2) than the central one $\omega_0$. The latter, are responsible for the cutoff increasing in the HHG spectrum and the former are responsible for the efficiency preserving. It must be considered that these new frequencies must be generated in a temporal zone of the pulse such that their energy range allows the ionization by the tunnel effect of the atomic electron [26]. Given the symmetry of the Gaussian pulse, the phase, proportional to this waveform, results symmetrical. The instantaneous frequency $\omega(t)$ as the temporal derivative of the phase $\phi(t,z)$ (see equation (6)) can be written as:

$$\omega(t) = \frac{\partial\phi(t,z)}{\partial t} = \omega_0 + \frac{8\ln(2)tz}{L_{\mathrm{NL}}\tau_0^2}\left|\exp\left[-2\ln 2\left(\frac{t}{\tau_0}\right)^2\right]\right|^2. \tag{7}$$

We complement the quantum outcomes with the classical analysis in figure 4 by integration the Newton–Lorentz equation for the four pulses plotted in figure 2.

The corresponding insets in the different panels represent a zoom of the fixed temporal zone in the four cases. It is worthwhile to stress that in this case the physical information provided by the classical analysis is extremely rich. On the one hand it confirms the cutoff extensions reported in figure 3. By comparing both figures we could observe an excellent matching between the classical and quantum mechanical approaches. In addition, note that in figures 4(a) and (b) the maximum recombination energy presents a 'shift' to the left side of the pulse and it takes place before the peak amplitude. This fact, and the constancy of the envelope amplitude for all up-chirped pulses, turn to be important for HHG at the high intensity regime close to the threshold of saturation. On this manner, the *up chirped* pulses proposed here produce higher HHG cutoff than a normal pulse with the same intensity, delaying harmful effects (such as the degradation of the harmonic efficiency) caused by the barrier suppression when the saturation limit is exceeded. In the same way, this asymmetry in the maximum of ionization/recombination process also favours the HHG process. As we mentioned before the excess

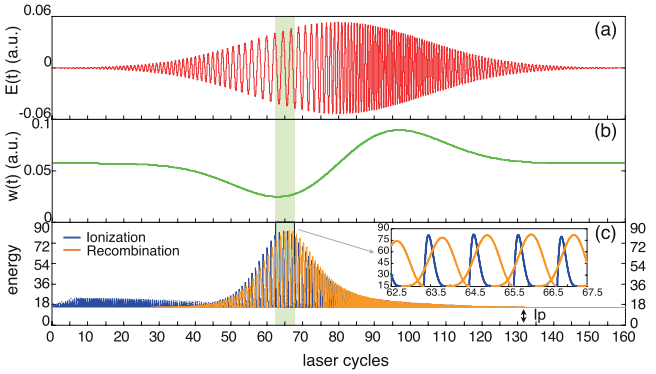


**Figure 5.** (a) Chirped pulse for $z = 100L_{\mathrm{NL}}$ and (b), in green, its corresponding first derivative of the phase, $\omega(t)$. (c) Classical trajectory analysis. The inset shows the zone of maximum energy of recombination.

of ionization causes degradation in the harmonic efficiency due to the plasma effects. The displacement of the maximum of ionization/recombination from the middle to the first part of the pulse (for up chirped pulses) is beneficial because it makes the maximum recombination event occur before in time, with lower amplitude, in comparison with the ordinary driving pulse, and consequently with lower degree of ionization and high harmonic generation efficiency.

In figure 5 we analyze in detail the chirped pulse for the case of $z = 100L_{\mathrm{NL}}$. In figure 5(b) we depict the frequency function $\omega(t)$, in green, obtained as the first derivative of the phase (equation (7)) of the waveform represented in figure 5(a). Figure 5(c) represents the corresponding classical analysis. Note that the largest wavelengths (i.e. maximum recombination energies) appear in a temporal region where $\omega(t)$ presents a minimum. Consequently it is clear the relation between larger wavelengths generated by SPM chirping and the extension of the HHG cutoff. For all driven pulses considered, the largest wavelength occurs at the same time. This can be deducted by setting the first derivative to zero. In addition, the largest wavelength is proportional to the $z$ coordinate as can also be seen from the derivative of equation (7).

In conclusion, we have presented some guidelines to perform an experimental approach that allows an effective extension in the HHG cutoff using chirped pulses obtained when they propagate in a medium where only the SPM phenomenon takes place. These cutoff extensions predicted scale with the largest wavelength and are proportional to the propagation length $z$. The main advantage this technique presents is that, while the longer wavelengths are responsible for the cutoff extension the shorter wavelengths are responsible for preserving the harmonic efficiency, at least, in the harmonic plateau governed by the $I_p + 3.17U_p$ law. To implement this experiment two fundamental aspects must be considered: first, the FWHM of the pulses must be greater than the relaxation time of the system and second, the propagation length $z$ should be less than the dispersion length $z \ll L_{\mathrm{D}}$. The fulfilment of these two hypothesis ensures the realization of the desired chirped pulses. This experimental scheme can be considered as optional for HHG instead of systems based on conventional optical parametric amplification (OPA) [30].

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