

High-order harmonic generation from a linear chain of ions

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Abstract. The high-order harmonic generation process due to the interaction of a multi-well quantum system with an intense laser field is examined. A plateau extension up to a photon energy of $I_p + 8U_p$ and the generation of attosecond pulses are demonstrated.

1. Introduction

When an atom is submitted to an intense laser field, it gives rise to strongly nonlinear effects such as above-threshold ionization (ATI) or the generation of highly energetic photons (high-order harmonic generation) [1]. In the harmonic generation process, the spectrum of the emitted light always has the same profile, a rapid decrease followed by a plateau (where the harmonics have nearly the same intensity). Numerical simulations and experimental results [2–5] both agree on the maximum photon energy that can be emitted (which sets the end of the plateau)

$$N_m \omega_L = I_p + 3.17U_p. \quad (1)$$

In this so-called cut-off law, N_m is the maximum harmonic order, I_p is the ionization potential and U_p is the ponderomotive energy. $U_p = E_0^2/4\omega_L^2$ (atomic units), with E_0 and ω_L being the peak amplitude and the frequency of the laser field, respectively. Beyond the understanding of the fundamental principles which are at the origin of the harmonic generation process, a strong effort is devoted to its optimization, that is having the strongest conversion efficiency from the laser energy to the harmonics and having the most energetic photons in order to achieve the extreme-ultraviolet (XUV) spectral region.

Because of the higher numbers of degrees of freedom, the study of the interaction of an intense laser field with more complex systems such as molecules or clusters not only revealed new mechanisms related to the nucleus such as above-threshold dissociation [6, 7] or Coulomb explosion [8, 9] but also enriched the electron dynamics. It brought out phenomena such as the enhanced ionization [10–13], gave rise to the possibility of increasing the number of generated harmonics [14–16] and demonstrated the production of trains of attosecond (10^{-18} fs) pulses [17].

Multi-well potentials have been used to model a wide number of physical systems such as linear multi-atomic molecules [18, 19], ionized clusters [20] or metals [21]. These potentials have been mostly used to investigate the enhanced ionization and the fragmentation

of polyatomic molecules [22] or to study the ionization and harmonic generation processes in metals [21].

It has already been shown that a system with two atoms is able to generate photons with energies up to $I_p + 8U_p$ [14] and to produce attosecond pulses [17]. The harmonics corresponding to photon energies between $I_p + 3.17U_p$ and $I_p + 8U_p$ form what is called the plateau extension. In this paper we study the high-order harmonic generation process due to the interaction of a linear chain of ions with an intense, linearly polarized laser pulse. We want to test the robustness of the mechanisms at the origin of the plateau extension (and attosecond pulse production) when the system is more complex than a small molecule. For a large number of ions (a maximum of 15), we want to know whether it is advantageous in terms of the conversion efficiency or if it can create, due to the multiple recombination sites, a loss in the number of harmonics generated and/or in the phase relation between the different harmonics (the phase relation which is at the origin of the attosecond pulse production obtained in a two-ion system [17]). To this day, systems capable of creating a plateau extension of energies up to $I_p + 8U_p$ and a train of attosecond pulses still only exist as theoretical models [17, 18], and no experiments have been carried out to the best of our knowledge. The system we use here can, in our opinion, give some information on the behaviour of an ionized cluster of intermediate size in terms of harmonic generation. Our goal is to show that these more complex systems also allow us to produce attosecond pulses and a plateau extension. They are therefore new candidates for experiments. The paper is divided into two main parts, the first one uses a seven-ion chain in order to find the optimal conditions for the generation of the harmonics as a function of the ionization probability and therefore of the enhanced ionization process. The second part uses 15 atoms in order to understand the influence of the intermediate ions (i.e. the ions between the central and the external ones) on the spectrum. Calculations have already been done to note the plateau extension in a chain of, for example, five atoms [18], but the number of intermediate ions is too small in order to understand their influence. Moreover, studies concerning the production of attosecond pulses have never been made for these more complex systems.

2. The model

In the single-atom case, the $3.17U_p$ term of the cut-off law (equation (1)) has been understood within a simple three-step model. The first step is the ionization of the atom, the second step is the motion of the free electron in the electromagnetic field (which can be described classically) and the third step is the recombination, which leads to the generation of the harmonics. Using the equations of motion of the classical electrodynamics (equation (2)), it has been shown [3] that the maximum kinetic energy of the electron when it recombines with the atom is $3.17U_p$, which explains the cut-off law.

If one uses two atoms, it has been shown in a previous paper [23] that when the nuclei have an internuclear distance $R = (2n - 1)\pi\alpha_0$ (where $\alpha_0 = E_0/\omega_L^2$ and $n = 1, 2, \dots$) the cut-off limit extends beyond $I_p + 3.17U_p$ up to $I_p + 8U_p$. The harmonics corresponding to photon energies between $I_p + 3.17U_p$ and $I_p + 8U_p$ form what is called the plateau extension. This limit of $8U_p$ corresponds to the maximum kinetic energy reachable by the electron during its classical excursion. This upper limit is easy to check [23], starting from the equations of motion of a classical electron in an electric field with initial position and velocity equal to zero

(in atomic units):

$$\begin{aligned}\frac{d^2z}{dt^2} &= -E_0 \sin(\omega_L t + \varphi) \\ \frac{dz}{dt} &= \frac{E_0}{\omega_L} [-\cos \varphi + \cos(\omega_L t + \varphi)] \\ z &= \frac{E_0}{\omega_L^2} [-\omega_L t \cos \varphi + \sin(\omega_L t + \varphi) - \sin \varphi].\end{aligned}\quad (2)$$

Since we do not know the value of the electric field when the electron is ionized, the phase φ allows us to simulate different times of ionization. The kinetic energy being $T = \frac{1}{2}4U_p[-\cos \varphi + \cos(\omega_L t + \varphi)]^2$, its maximum value is obtained when the expression in square brackets is equal to two and gives $T_{\max} = 8U_p$. The position where the kinetic energy of the electron is maximum is $z_{E_{\max}} = (2n - 1)\pi\alpha_0$. Then, when an electron is ejected by an atom (called the parent atom and positioned at $z = 0$) and recombines in another one situated at a distance $z = (2n - 1)\pi\alpha_0$ (the atom is then positioned where the kinetic energy of the electron is maximum), photons with an energy up to $I_p + 8U_p$ are emitted due to the higher recombination energy.

In order to simulate the dynamics of a bounded electron in interaction with a linearly polarized intense laser field, we solve the one-dimensional Schrödinger equation (in atomic units):

$$i \frac{\partial}{\partial t} \psi(z, t) = -\frac{1}{2} \frac{\partial^2}{\partial z^2} \psi(z, t) + V(z; R) \psi(z, t) - zE(t) \psi(z, t). \quad (3)$$

The equation is solved by wavepacket propagation with the ground state of the system as the initial state. z is the electronic coordinate, $E(t)$ is the amplitude of the electric field and $V(z; R)$ is the system potential which depends parametrically on the inter-well distance R .

The electric field is given by

$$E(t) = f(t) \sin \omega_L t \quad (4)$$

where

$$f(t) = \begin{cases} 0 & \text{for } t < 0 \\ E_0 \sin^2\left(\frac{t\pi}{2\sigma}\right) & \text{for } 0 < t < \sigma \\ E_0 & \text{for } \sigma < t < T + \sigma \\ E_0 \sin^2\left(\frac{(t-T)\pi}{2\sigma}\right) & \text{for } T + \sigma < t < T + 2\sigma \\ 0 & \text{for } t > T + 2\sigma \end{cases} \quad (5)$$

σ and T are the turn-on (off) and plateau durations of the electric field envelope. In many situations of intense, linearly polarized, laser-matter interactions it has been shown that it is a good approximation to consider one-dimensional models [24] in the line of the widely used one-dimensional hydrogen model with a soft-core Coulomb potential [25] (this kind of potential allows us to avoid the Coulomb singularity). In our case, the multi-well potential is simply a sum of softened Coulomb potentials:

$$V(z; R) = \sum_{n=-N}^{+N} -\frac{1}{\sqrt{1 + (z + nR)^2}}. \quad (6)$$

The number of wells is then $2N + 1$. These potentials are widely used to simulate small molecules [16, 26], ionized clusters [27] or metals [21].

The power spectrum of the emitted light is calculated as

$$S(\omega) \propto |TF[\ddot{d}(t)]|^2 \quad (7)$$

where $TF[\ddot{d}(t)]$ is the Fourier transform of the acceleration of the dipole moment $d(t) = \langle \psi(t) | z | \psi(t) \rangle$ [24].

The plateau extension has been demonstrated in different systems with one- and three-dimensional simulations [15, 16, 18, 23]. The simulations demonstrated, however, that the intensity of the harmonics forming the plateau extension is lower than the intensity of the harmonics corresponding to photons with an energy smaller than $I_p + 3.17U_p$. Some features, such as, for example, the possibility of producing attosecond pulses [17], justify the interest in the study of this plateau extension despite the lower conversion efficiency. The next section presents the results of the study of a multi-well structure in interaction with an intense linearly polarized laser field. The possibility to produce the plateau extension (and the attosecond pulses) despite the multiple recombination sites and the relation between the harmonic generation process and other nonlinear effects such as the enhanced ionization are discussed.

3. Results

3.1. Enhanced ionization and harmonic generation

We investigate the harmonic spectra generated by a linear chain of seven ions submitted to an intense laser pulse ($I = 2 \times 10^{14} \text{ W cm}^{-2}$) of wavelength $\lambda = 800 \text{ nm}$. The pulse has a turn-on and off (σ) of three cycles and a constant amplitude over six cycles (equation (5)). Figure 1 shows the ionization probability of the electron as a function of the inter-well distance. We observe that around an inter-well distance of 4 au the probability of ionization is maximum. This is what is called the enhanced ionization effect [10–13]. The harmonic spectra are then calculated for different internuclear distances ($R = 3, 4$ and 11 au) in order to cover the various regimes of ionization (see figure 1).

Figure 2, left-hand column, displays the potential $V(z; R)$ (thin curves) and the static potential $V(z; R) - zE_0$ (which corresponds to the maximum distortion) with the initial ground state energy (the initial wavefunction of these systems is maximum at $z = 0$) and on the right-hand column the corresponding spectra.

- For $R = 3 \text{ au}$, the ionization probability reaches 31% (figure 1) and nearly 60 harmonics are generated (figure 2(d)). The system, which extends over 18 au (figure 2(a)), ionizes due to tunnelling through the barrier B1 and the plateau nearly follows the cut-off law (equation (1)). In figure 2(d), we marked the energy of $I_p + 3.17U_p$ as a reference, but the cut-off of this spectrum is at a close but higher value. Effectively, the electron can recombine with an ion other than the central one, then creating a small plateau extension (small because of the small distance between the central and the peripheral atom). This can be understood with the help of figure 3. This figure plots the kinetic energy of a free electron as a function of its coordinate (equation (2)) [14] for different times of ionization (i.e. different phases φ). Since we are interested in the maximum kinetic energy as a function of the electron position regardless of the time of ionization, a large number of trajectories with different phases have been computed in order to obtain the envelope. In this figure we can see that when the electron crosses the position $z = 0$, the maximum kinetic energy of recombination is $3.17U_p$ (thin curves). This is why when

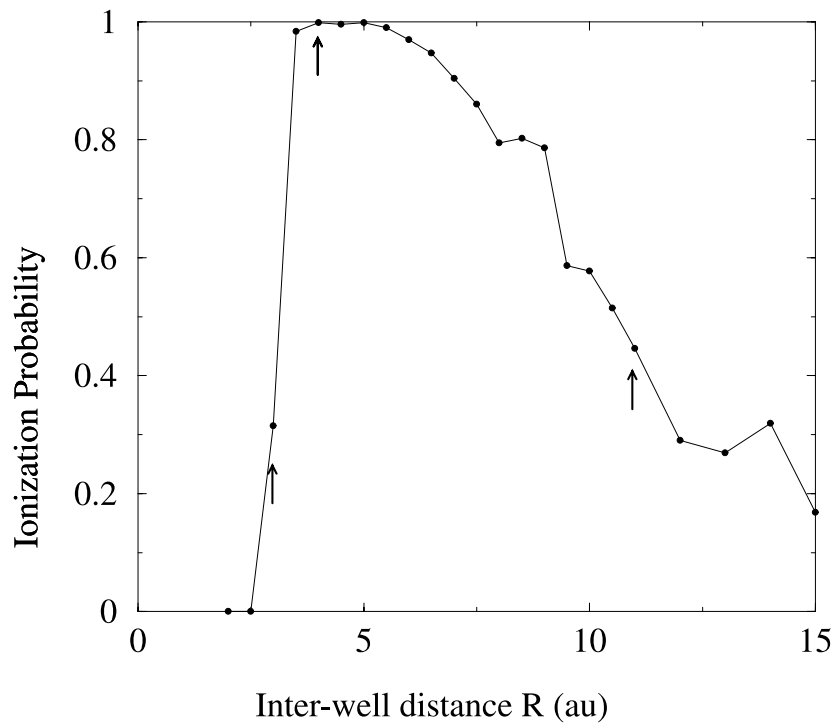


Figure 1. Ionization probability of a seven-ion system as a function of the inter-well distance. The arrows show the inter-well distances used in the calculations.

an electron is ionized from an atom, it comes back to its initial position and produces harmonics, the cut-off is positioned at an energy of $I_p + 3.17U_p$. In our case, the maximum distance between the central atom ('electron source') and a possible recombination ion is 9 au. When it recombines at this distance, the electron has a kinetic energy of nearly $3.7U_p$ (see the broken curves in figure 3), then allowing a plateau extension up to photon energies of nearly $I_p + 3.7U_p$. This small plateau extension is then not clearly observable (figure 2(d)).

- For $R = 4$ au we are at the enhanced ionization maximum with a probability of $\simeq 99\%$ (figure 1). We can see in figure 2(b) that not only is the potential barrier B1 completely lowered, but what we can call the inter-well barrier B2 also allows the ionization. In this situation of total barrier suppression, we observe that the spectrum (figure 2(e)) reaches, for the same reason as previously explained, a maximum photon energy of just beyond $I_p + 3.17U_p$ but, due to the 'over-the-barrier ionization', the resolution of the harmonics is very low.
- For $R = 11$ au, the ionization probability lowers again to nearly 44% (figure 1) because the superposition of the different atomic potentials decreases and allows the potential barrier B2 to increase (figure 2(c)). The spectrum displays 60 harmonics (figure 2(f)) that are very well defined due to the tunnelling ionization regime. Here we must note that the spectra obtained for $R = 3$ au (S1) and $R = 11$ au (S2) display the same number of harmonics, but they are generated in different ways. This can be seen because the spectrum S1 (figure 2(d)) has its cut-off at an energy of $\simeq I_p + 3.7U_p$, while S2 (figure 2(f)) has its cut-off at $\simeq I_p + 5.3U_p$. These different cut-off limits can be understood with the help of

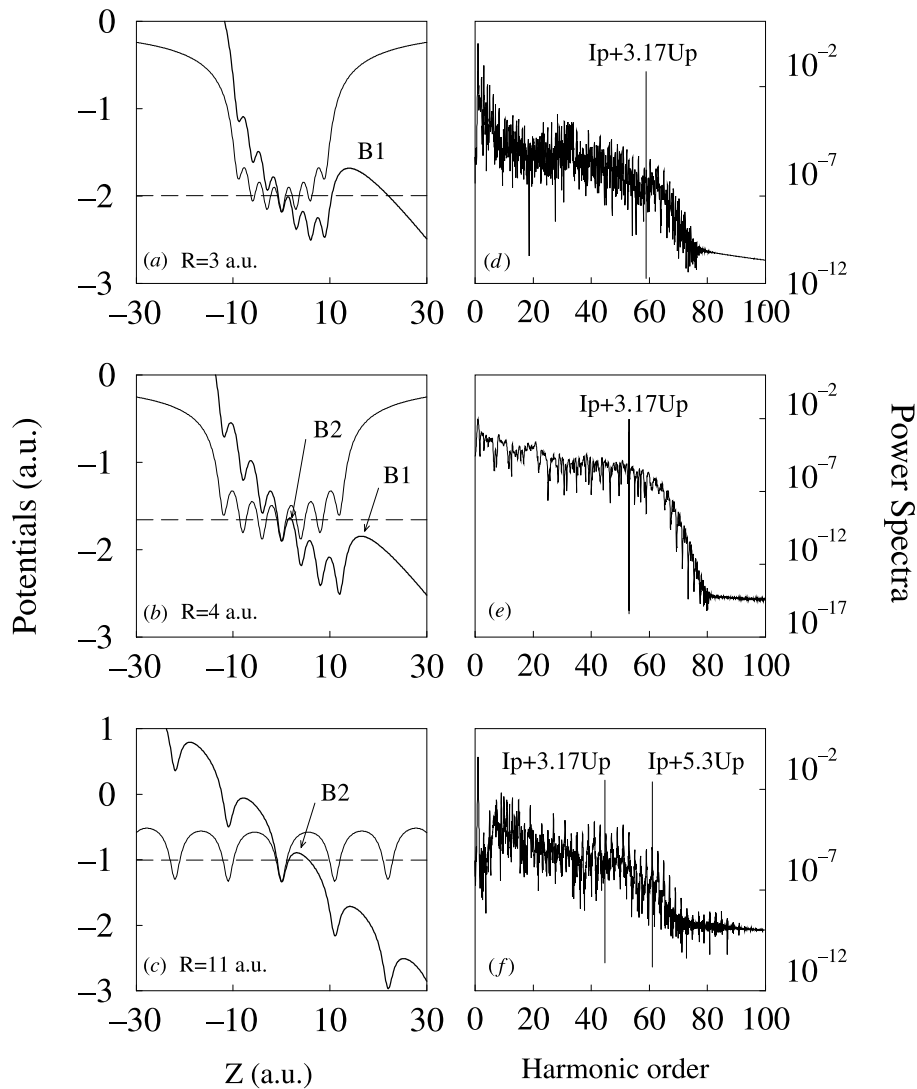


Figure 2. Left-hand column, the potential $V(z; R)$ (thin curves) and the static potential $V(z; R) - z \cdot E_0$ (thick curves) for different inter-well distances as a function of the electron coordinate Z . Broken lines, the energy of the initial ground state. Right-hand column, corresponding harmonic generation spectra. The vertical lines show the photon energy in terms of the so-called cut-off law. Parts (a) and (d) correspond to $R = 3$ au, (b) and (e) to $R = 4$ au and (c) and (f) to $R = 11$ au.

figure 3. In the spectrum S1, we have seen that the distance between the ions is not large enough to note the effect of the recombination of the electron with the external ions and the cut-off remains at an energy which is nearly $I_p + 3.17U_p$. In the spectrum S2 the maximum distance between the central atom and a possible recombination ion is 33 au. When it recombines at that distance, the electron has a kinetic energy of nearly $5.3U_p$ (see the thick curves in figure 3), then allowing a plateau extension up to photon energies of nearly $I_p + 5.3U_p$. This plateau extension is not observable (in this case) in terms of a number of generated harmonics because the ionization potential of the system decreases when

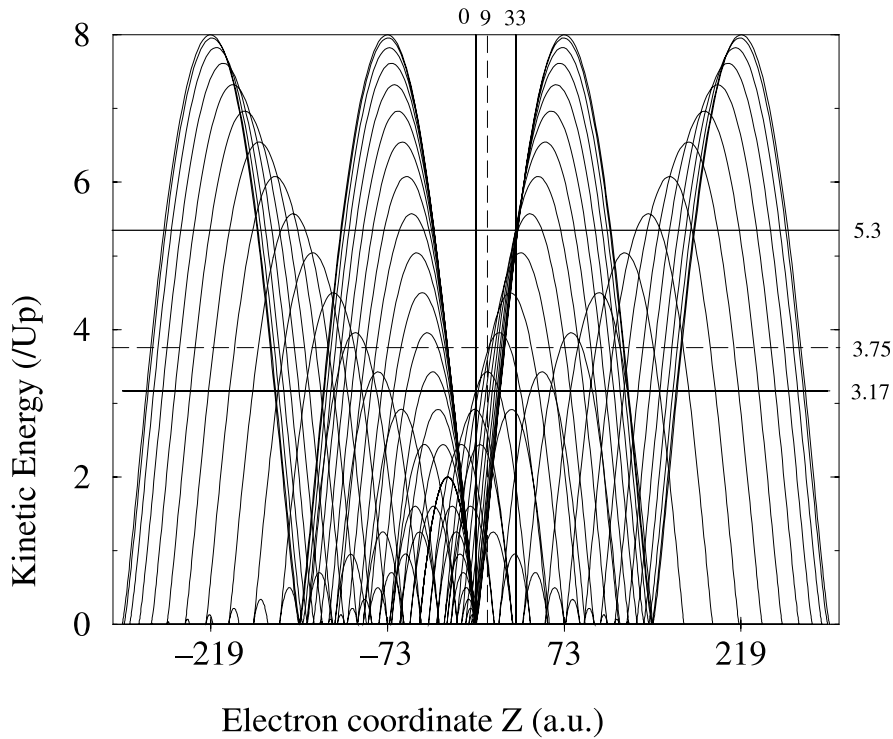


Figure 3. Kinetic energy (in units of U_p) of a free electron in interaction with an electric field of intensity $I = 2 \times 10^{14} \text{ W cm}^{-2}$ and wavelength $\lambda = 800 \text{ nm}$ as a function of its position Z . Various trajectories corresponding to different initial phases of the laser are plotted. The initial position and velocity of the electron is zero. The thin (thick) full lines correspond to the maximum kinetic energy of the electron at the position $z = 0 \text{ au}$ ($z = 33 \text{ au}$). The broken lines correspond to the maximum kinetic energy of the electron at the position $z = 9 \text{ au}$.

the inter-well distance increases and compensates (equation (1)) the plateau extension ($I_p = 1.99, 1.66, 1.17 \text{ au}$ for $R = 3, 4, 11 \text{ au}$).

To summarize, we noted that the region of enhanced ionization does not provide a good ‘resolution’ of the harmonics due to the total barrier suppression mechanism at the origin of the ionization. The regions around $R = 3$ and 11 au seem equivalent, but the region around $R = 3 \text{ au}$ is very narrow and is thus not very convenient. We can say that to produce well defined harmonics and energetic photons from a linear chain of ions in interaction with an intense linearly polarized field, the best conditions correspond to an inter-well distance larger than that corresponding to the enhanced ionization maximum.

3.2. Harmonic generation and attosecond pulses

The region around an inter-well distance of 11 au ensures a tunnelling regime which is good for the resolution of the harmonics and a sufficiently high rate of ionization. In order to maximize the plateau extension, we put 15 ions at an inter-well distance of 10.4 au (figure 4(a)). The distance between the central atom (‘electron source’) and the external ones is then 73 au (figure 4(a)). This situation fulfils the classical conditions (see figure 3) to obtain the maximum plateau extension (photon energy up to $I_p + 8U_p$). We can see in figure 4(b) that the harmonic

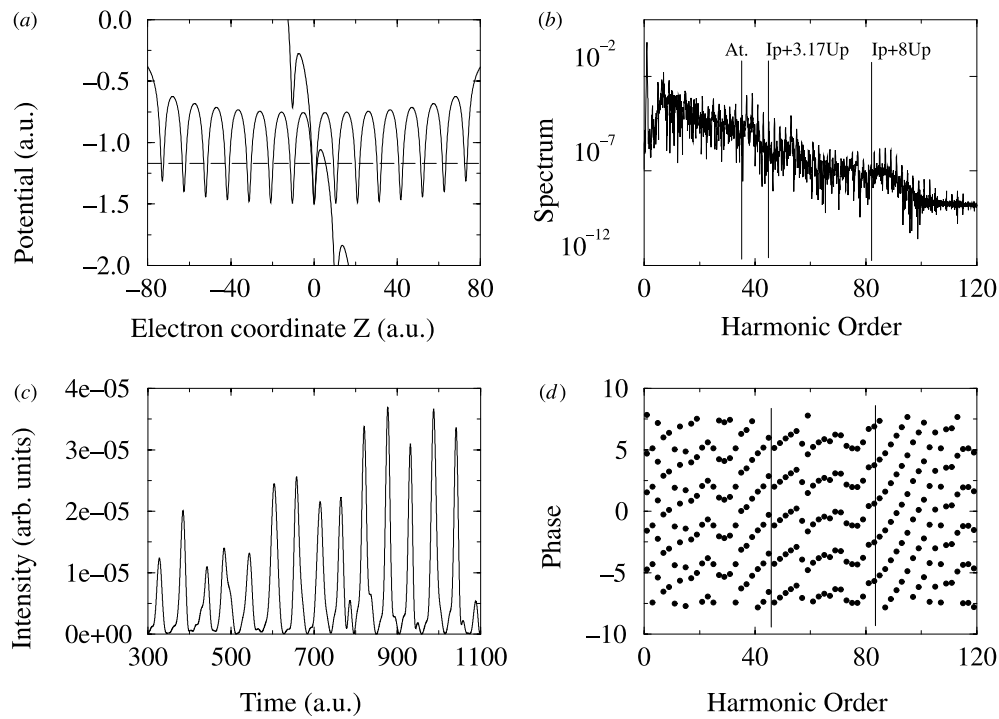


Figure 4. (a) Potential $V(z; R)$ (thin curve) and static potential $V(z; R) - z.E_0$ (thick curve) for an inter-well distance of $R = 10.4$ au. The broken line shows the ground initial-state energy. (b) Harmonic spectrum. The vertical lines correspond to the 35th harmonic (the maximum harmonic generated by an atom), the 45th and the 81st harmonics. (c) Train of attosecond pulses reconstructed from the 33rd to the 45th harmonics. (d) Phase of the harmonics (modulus π). The vertical lines are situated as in (b).

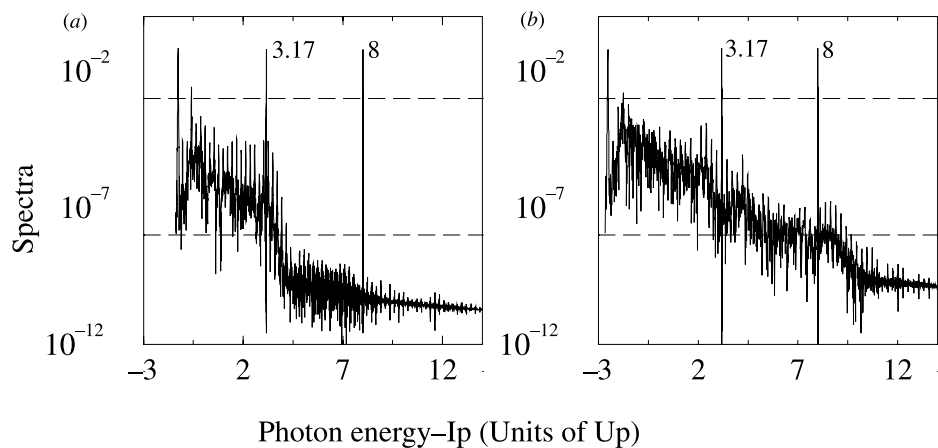


Figure 5. Spectra for a linear chain of three (a) and 15 (b) ions. The vertical lines show the cut-off energies at $I_p + 3.17U_p$ and $I_p + 8U_p$.

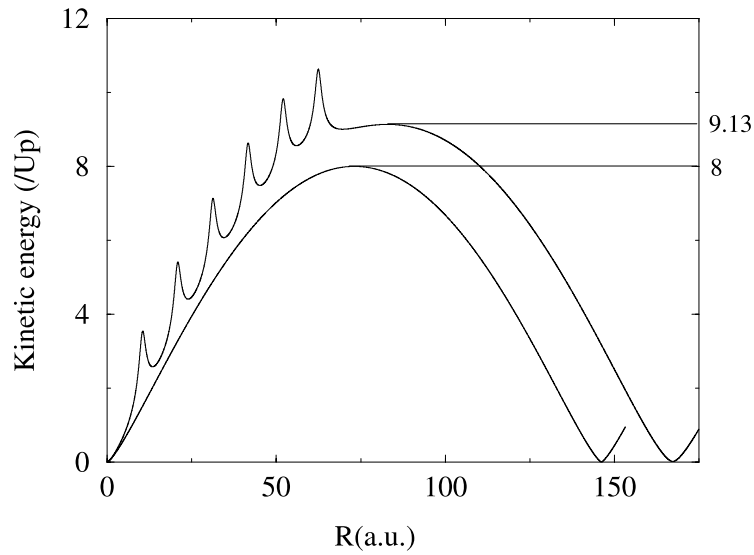


Figure 6. Kinetic energy in units of U_p as a function of the electron position for $\varphi = \pi$ (see equation (2)). The thin curve represents the free motion of the electron. The thick curve shows the electron motion with six potentials on his trajectory. The thin line shows the maximum value reachable ($8U_p$) without intermediate ions. The thick line shows the maximum value reachable ($9.13U_p$) after six ions.

spectrum allows us to distinguish clearly the ‘conventional’ plateau and the plateau extension. At this point, it is interesting to compare the spectra generated by a chain of 15 ions (figure 5(b)) and a chain of three ions (figure 5(a)) in order to understand what is the contribution of the intermediate ions, that is, the ions which are between the central ion and the external ones (which produce the most energetic photons). In the case of three ions, there are no intermediate ions, whereas two lots of six intermediate ions are present in the chain of 15 ions. These ions are possible sites of recombination for the electron and will contribute to the emission of photons. The influence appears clearly in the spectrum (figure 5(b)), where the intensity of the harmonics between an energy of $I_p + 3.17U_p$ and $I_p + 8U_p$ is enhanced. The ionization probabilities are equivalent in the two cases, $\simeq 26\%$ for the three-ion chain and $\simeq 29\%$ in the 15-ion chain. Moreover, we can note that photons with an energy greater than $I_p + 8U_p$ appear. We can suppose that the electron, before recombining with the last ion of the chain, has gained kinetic energy during the previous interactions (with the intermediate ions). This possibility is confirmed by classical simulations as illustrated in figure 6. We can see in this figure the kinetic energy in units of U_p as a function of the position of the electron. Without any ions between the central ion and the external ones, the maximum kinetic energy reachable is $8U_p$. If six ions are placed in between as in the quantum case, we note that the maximum kinetic energy is now $\simeq 9U_p$. Due to the intermediate kicks, this value is no longer reached at $R = \pi\alpha_0$ but at a higher value. Because the difference in the maximum kinetic energy is very small and because we wanted to keep the internuclear distance constant, we did not change the position of the external atoms.

Another important characteristic of the spectrum is the phase of the harmonics. As is well known [28], there must be a phase relation in order to reconstruct a train of pulses from the harmonics. This is not the case in the atoms for ‘conventional’ harmonics (where the energy goes up to $I_p + 3.17U_p$). In contrast, it has been shown [17] that in the plateau extension

(energy from $I_p + 3.17U_p$ up to $I_p + 8U_p$) the harmonics are phase-locked and generation of a train of attosecond pulses is possible. In our situation, the phases of the harmonics in the plateau extension are not as clearly linked (figure 4(d)) as in the case of only two atoms [17]. This is because the recombination of the electron with each ion of the chain contributes to the spectrum. It is interesting to note, however, that just before the cut-off at $I_p + 3.17U_p$, there is a region (between the 33rd and 45th harmonics) where the phase changes linearly with the harmonic order. By filtering this region with a function which equals one between these harmonics and goes to zero as sine squared over two harmonics and making an inverse Fourier transform, we show in figure 4(c) that a train of attosecond pulses is produced. Consecutive peaks are separated by half of the laser period ($\pi/\omega_L \simeq 55$ au), since the harmonics are separated by $2\omega_L$. The carrier wave frequency is equal to $45\omega_L$, 45 being the order of the highest harmonic involved in the reconstruction. We can note other linearly related phases, but our interest is to take that from the most intense harmonics. The harmonics beyond an energy of $I_p + 8U_p$ are phase-locked, but their intensity decreases, which is not an optimal condition for the reconstruction. In order to quantify the gain obtained with this kind of system, we can compare with an atom described by the same kind of potential ($V(z) = -1/\sqrt{(1+z^2)}$). The maximum number of harmonics generated by the atom is $N_{\max}^{\text{At}} \simeq (I_p + 3.17U_p)/\omega_L = 35$ ($I_p = 0.67$ au, $U_p = 0.438$ au, $\omega_L = 0.057$ au). This limit is represented in figure 4(b) by a vertical line. For the linear chain of atoms, $N_{\max}^{\text{Ch}} \simeq (I_p + 8U_p)/\omega_L = 81$ ($I_p = 1.17$ au, same U_p and ω_L). The increase of the maximum number of generated harmonics is mainly due to the change of the cut-off law but also to the superposition of the potentials of the different atoms which increase the ionization potential of the central atom.

4. Conclusion

A linear chain of ions in interaction with an intense polarized laser field can produce a spectrum with a maximum photon energy equal to or even larger than $I_p + 8U_p$. We showed that, in contrast with the case of two-/three-atom systems, the presence of a high number of ions increases the conversion efficiency in the plateau extension and allows a larger plateau extension (with photon energies over $I_p + 8.0U_p$), due to the acceleration of the electron by the intermediate ions. In order to have well defined harmonics, the inter-atomic distance must not correspond to the enhanced ionization maximum. Another important characteristic of the spectrum produced is the phase relation existing between the harmonics, a signature of the existence of attosecond pulses.

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