Enhancement of high harmonic generation by confining electron motion in plasmonic nanostructures

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Abstract: We study high-order harmonic generation (HHG) resulting from the illumination of plasmonic nanostructures with a short laser pulse of long wavelength. We demonstrate that both the confinement of the electron motion and the inhomogeneous character of the laser electric field play an important role in the HHG process and lead to a significant increase of the harmonic cutoff. In particular, in bow-tie nanostructures with small gaps, electron trajectories with large excursion amplitudes experience significant confinement and their contribution is essentially suppressed. In order to understand and characterize this feature, we combine the numerical solution of the time-dependent Schrödinger equation (TDSE) with the electric fields obtained from 3D finite element simulations. We employ time-frequency analysis to extract more detailed information from the TDSE results and classical tools to explain the extended harmonic spectra. The spatial inhomogeneity of the laser electric field modifies substantially the electron trajectories and contributes also to cutoff increase.

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References and links
1. M. Protopapas, C. H. Keitel, and P. L. Knight, “Atomic physics with super-high intensity lasers,” Rep. Prog. Phys. 60(4), 389–486 (1997).
2. T. Brabec and F. Krausz, “Intense few-cycle laser fields: frontiers of nonlinear optics,” Rev. Mod. Phys. 72(2), 545–591 (2000).
3. F. Krausz and M. Ivanov, “Attosecond physics,” Rev. Mod. Phys. 81(1), 163–234 (2009).
4. P. B. Corkum and F. Krausz, “Attosecond science,” Nat. Phys. 3(6), 381–387 (2007).
5. M. Lein, “Molecular imaging using recolliding electrons,” J. Phys. B 40(16), R135–R173 (2007).
6. P. B. Corkum, “Plasma perspective on strong field multiphoton ionization,” Phys. Rev. Lett. 71(13), 1994–1997 (1993).
7. M. Lewenstein, P. Balcou, M. Y. Ivanov, A. L’Huillier, and P. B. Corkum, “Theory of high-harmonic generation by low-frequency laser fields,” Phys. Rev. A 49(3) 2117–2132 (1994).
8. S. Kim, J. Jin, Y.-J. Kim, I.-Y. Park, Y. Kim, and S.-W. Kim, “High-harmonic generation by resonant plasmon field enhancement,” Nature 453(7196), 757–760 (2008).
9. I.-Y. Park, S. Kim, J. Choi, D.-H. L. Y.-J. Kim, M. F. Kling, M. I. Stockman, and S.-W. Kim, “Plasmonic generation of ultrashort extreme-ultraviolet light pulses,” Nat. Phot. 5(11), 677–681 (2011).
10. M. Mühlischlegel, H.-J. Eisler, O. J. F. Martin, B. Hecht, and D. W. Pohl, “Resonant optical antennas,” Science 308(5728), 1607–1609 (2005).

11. P. J. Schuck, D. P. Fromm, A. Sundaramurthy, G. S. Kino, and W. E. Moerner, “Improving the mismatch between light and nanoscale objects with gold bowtie nanoantennas,” Phys. Rev. Lett. 94(1), 017402 (2005).

12. R. J. Jones, K. D. Moll, M. J. Thorpe, and J. Ye, “Phase-coherent frequency combs in the vacuum ultraviolet via high-harmonic generation inside a femtosecond enhancement cavity,” Phys. Rev. Lett. 94(19) 193201 (2005).

13. A. Husakou, S.-J. Im, and J. Herrmann, “Theory of plasmon-enhanced high-order harmonic generation in the vicinity of metal nanostructures in noble gases,” Phys. Rev. A 83(4), 043839 (2011).

14. I. Yavuz, E. A. Blesa, Z. Altun, and T. Topçu, “Generation of a broadband xuv continuum in high-order-harmonic generation by spatially inhomogeneous fields,” Phys. Rev. A 85(1), 013416 (2012).

15. M. F. Ciappona, J. Biegert, R. Quidant, and M. Lewenstein, “High-order-harmonic generation from inhomogeneous fields,” Phys. Rev. A 85(3), 033828 (2012).

16. T. Shaaran, M. F. Ciappona, and M. Lewenstein, “Quantum-orbit analysis of high-order-harmonic generation by resonant plasmon field enhancement,” Phys. Rev. A 86(2), 023408 (2012).

17. M. Sivos, M. Duwe, B. Abel, and C. Ropers, “Nanostructure-enhanced atomic line emission,” Nature 485(7397), E1–E3 (2012).

18. S. Kim, J. Jin, Y.-J. Kim, I.-Y. Park, Y. Kim, and S.-W. Kim, “Kim et al. reply,” Nature 485(7397), E1–E3 (2012).

19. G. Baffou and R. Quidant, “Thermo-plasmonics: using metallic nanostructures as nano-sources of heat,” Laser & Photon. Rev. (in press) (2012).

20. S. L. Stebbings, F. Süßmann, Y.-Y. Yang, A. Scrinzi, M. Durach, A. Rusina, M. I. Stockman and M. F. Kling, “Generation of isolated attosecond extreme ultraviolet pulses employing nanoplasmonic field enhancement: optimization of coupled ellipsoids,” New Journal of Physics 13(7), 073010 (2011).

21. F. Süßmann and M. F. Kling, “Attosecond nanoplasmonic streaking of localized fields near metal nanoparticles,” Phys. Rev. B 84(12), 121406(R) (2011).

22. S. Zherebtsov et al., “Controlled near-field enhanced electron acceleration from dielectric nanospheres with intense few-cycle laser fields,” Nat. Phys. 7(8), 656–662 (2011).

23. M. Hommelhoff, Y. Sortais, A. Aghajani-Talesh, and M. A. Kasevich, “Field emission tip as a nanometer source of free electron femtosecond pulses,” Phys. Rev. Lett. 96(7), 077401 (2006).

24. M. Schenk, M. Krüger, and P. Hommelhoff, “Strong-field above-threshold photoemission from sharp metal tips,” Phys. Rev. Lett. 105(2), 257601 (2010).

25. M. Krüger, M. Schenk, and P. Hommelhoff, “Attosecond control of electrons emitted from a nanoscale metal tip,” Nature 475(7354), 78–81 (2011).

26. M. Krüger, M. Schenk, M. Förster, and P. Hommelhoff, “Attosecond physics in photoemission from a metal nanopipet,” J. Phys. B 45(7), 074006 (2012).

27. G. Herrink, D. R. Solli, M. Gulde, and C. Ropers, “Field-driven photoemission from nanostructures quenches the quiver motion,” Nature 483(7388), 190–193 (2012).

28. P. Salières, A. L’Huillier, P. Antoine, and M. Lewenstein, “Study of the spatial and temporal coherence of high-order harmonics,” Advances in Atomic, Molecular and Optical Physics, eds. B. Bederson and H. Walther 41, 83–142 (1999).

29. A. L’Huillier and M. Lewenstein, “Principles of single atom physics: high-order harmonic generation, above-threshold ionization and non-sequential ionization,” Strong Field Laser Physics ed. T. Brabec, Springer Series in Optical Sciences (Springer, 2008).

30. J. A. Pérez-Hernández, M. F. Ciappona, M. Lewenstein, L. Roso, A. Zait, “Beyond Carbon K-edge harmonic emission using spatial and temporal synthesized laser field,” arXiv:1207.4653v1 (2012).

31. C. Su and J. H. Eberly, “Model atom for multiphoton physics,” Phys. Rev. A 44(9), 5997–6008 (1991).

32. J. L. Krause, K. J. Schafer, and K. C. Kulander, “Calculation of photoemission from atoms subject to intense laser fields,” Phys. Rev. A 45(7), 4998–5010 (1992).

33. K. J. Schafer and K. C. Kulander, “High harmonic generation from ultrafast pump lasers,” Phys. Rev. Lett. 78(4), 638–641 (1997).

34. A. Thai, M. Hemmer, P. Bates, O. Chalus, and J. Biegert, “Sub-250-mrad, passively carrier-envelope-phase-stable mid-infrared OPCPA source at high repetition rate,” Opt. Lett. 36(19), 3918–3920 (2011).

35. S. S. Acimović, “Introduction to nanoparticle characterization in COMSOL” (available from http://sdjancosl.com, 2011).

36. P. B. Johnson and R. W. Christy, “Optical Constants of the Noble Metals,” Phys. Rev. B 6(12), 4370–4379 (1972).

37. D. Gabor, “Theory of communication,” J. Inst. Electr. Eng. 93, 429–441 (1946).

38. C. C. Chirilă, I. Dreissigacker, E. V. van der Zwan, and M. Lein, “Emission times in high-order harmonic generation,” Phys. Rev. A 81(3), 033412 (2010).

39. L. V. Keldysh, “Ionization in the field of a strong electromagnetic wave,” Sov. Phys. JETP 20(5), 1307–1314 (1965).

40. M. V. Ammosov, N. B. Delone, and V. P. Krainov, “Tunnel ionization of complex atoms and of atomic ions in an alternating electromagnetic field,” Sov. Phys. JETP 64(6), 1191–1194 (1986).
1. Introduction

When atoms and molecules are subject to intense laser radiation, new phenomena appear as a consequence of this interaction. Among them, high-harmonic generation (HHG), above-threshold ionization (ATI), and non-sequential double ionization (NSDI) can be mentioned as the most important ones [1, 2]. In particular, HHG has attracted significant interest, since it represents the most reliable pathway to coherent light sources in the ultraviolet (UV) to extreme ultraviolet (XUV) spectral range. These tools are in high demand nowadays for basic research, material science, biology and possibly lithography [3]. Furthermore, HHG opens the possibility of generating coherent attosecond pulses [4] enabling the extraction of temporal and spatial information with attosecond and sub-Ångström resolution, respectively [5].

The physical mechanism behind the generation of high-order harmonics has been well established in the so-called three step or simple man’s model [6, 7]. The first step is the tunnel ionization of an atomic or molecule electron as a consequence of the nonperturbative interaction with the coherent electromagnetic radiation. The classical propagation of the electron in the laser field establishes the second step of this model. In the third step, the electron returns to its origin and recombines with its parent ion, emitting a high-energy photon with energy equal to the sum of the ionization potential and the electron kinetic energy.

In noble gases, high-order harmonic generation only occurs if the laser intensity exceed $10^{13}$ W cm$^{-2}$, two orders of magnitude larger than the output of the current femtosecond oscillators. Nowadays, chirped-pulse amplification is employed to reach the threshold intensity. In addition, improving the efficiency and duty cycle of XUV radiation based on HHG is challenging. The recent demonstration based on surface plasmon resonances as light enhancers could provide a potential solution to this problem [8,9]. Indeed, field enhanced HHG using plasmonics, generated starting from engineered metal nanostructures, requires no extra cavities or laser pumping to amplify the power of the input pulse. By exploiting surface plasmon resonances, local electric fields can be enhanced by more than 20 dB [10, 11]. Consequently, the intensity of the locally enhanced electric field is strong enough to exceed the threshold laser intensity for HHG generation in noble gases. In particular, using gold bow-tie shaped nanostructures, it has been shown that the enhancement was sufficient to produce XUV wavelengths from the 7th (114 nm) to the 17th (47 nm) harmonics while the pulse repetition rate remains unaltered without any extra pumping or cavity attachment (for cavity enhancement production of XUV radiation see e.g. [12]). Furthermore, the high harmonics radiation generated from each nanostructure acts as a point-like source, enabling collimation or focusing of this coherent radiation by means of constructive interference. This would open a wide range of possibilities to spatially arrange nanostructures to enhance and even shape the HHG spectra [8].

The mechanism behind of HHG based on plasmonics can be explained as follows (the full explanation can be found in [8]). When a femtosecond low intensity laser pulse couples to the plasmon mode, it initiates a collective oscillation among free charges within the metal. A spot of highly amplified electric field is created while these free charges redistribute the electric field around the metal nanostructure. The enhanced field is well above the threshold for generating high harmonics. As a result, by injection of noble gases into this localized region, HHG can be produced. In here, the enhanced field is not spatially homogeneous in the region where the electron dynamics take place. Additionally, the movement of the electron in the enhanced field would be restricted in space. These two features imply strong modifications in the harmonic spectra, as was shown recently by several authors [13–16].

The outcome of experiment of Kim et al. [8], however, has been recently under an intense examination [17,18]. Among the matters to be resolved are the real shape of the local enhanced field, the actual intensity enhancement of the input laser field, the damage threshold of the gold bow-tie nanostructures and the characteristics of the radiation emitted by these nanosources, i.e.
if the high harmonic radiation is in fact coherent (HHG) or merely an incoherent atomic line emission [17, 18]. Regarding the field enhancement, for instance, the authors of Ref. [8], based on their own finite-element simulations, reported an intensity enhancement up to 4 orders of magnitude (40 dB). It means for input laser intensities of the order of $10^{11}$ W cm$^{-2}$ the local field could reach to the order of $10^{15}$ W cm$^{-2}$ at the vicinities of the bow-tie nanostructure. Then again, from the measured high-order harmonic spectra it seems that these numbers are not realistic, since no harmonics beyond 17$^{th}$ were observed, a limit corresponding to intensities of the order of $5 \times 10^{13}$ W cm$^{-2}$ (we have estimated this value using the well established three step or simple man’s model [7]). It is clear that the experimental field enhancement is about 2 order of magnitude smaller than the finite elements simulations. In our theoretical models we considered this point using a reduction factor in the field enhancement obtained from our own finite-element simulations.

Another aspect to consider is the potential for photo-damage to the metal bow-tie nanostructures by strong and short laser pulses. Heat generated by the Joule effect could indeed be high enough to modify the shape of the metal nanostructure and even melt it. We have made estimations of the absorbed energy density for gold nanoantennas resonating at both $\lambda = 800$ nm and $\lambda = 1800$ nm and compared them with the absorption of a 50-nm-thick continuous gold film [19]. When these systems are excited with a resonant monochromatic plane wave of $\lambda = 800$ nm, with intensity $1.4 \times 10^{11}$ W cm$^{-2}$, the absorbed power per unit volume of nanoantennas and continuous film are $6.14 \times 10^3$ nW/nm$^3$ and $1.16 \times 10^5$ nW/nm$^3$, respectively; while for $\lambda = 1800$ nm they are $3.83 \times 10^4$ nW/nm$^3$ and $1.07 \times 10^5$ nW/nm$^3$, respectively. Our results show that the heat produced by nanoantennas engineered to resonate at $\lambda = 1800$ nm is smaller compared to those that resonate at $\lambda = 800$ nm. This lower absorbency, combined with the lower electric field enhancements required to observe the same harmonics, make infrared antennas particularly advantageous compared to visible antennas. Alternative approaches employing different kinds of metallic nanostructures, e.g. nanoparticles (see e.g. [20–22]) or nanotips (see e.g. [23–27]), have been recently used to generate high energy electrons as a way to study the distinct and new characteristics of these nanosystems. However, to the best of our knowledge, there exist no experiments demonstrating high-order harmonic generation in any of the above mentioned nanosystems.

Up to now numerical and semiclassical approaches to study laser-matter processes in atoms and molecules, in particular high-order harmonic generation (HHG), have been largely based on the assumption that the laser electric field ($\mathbf{E}(\mathbf{r}, t)$) and its vector potential associated ($\mathbf{A}(\mathbf{r}, t)$) are spatially homogeneous in the region where the electron dynamics takes place, i.e. $\mathbf{E}(\mathbf{r}, t) = \mathbf{E}(t)$ and $\mathbf{A}(\mathbf{r}, t) = \mathbf{A}(t)$ [1, 2]. On the other hand, the fields generated using plasmonics are spatially dependent and can not be described by this supposition. From a theoretical viewpoint, the HHG process using homogeneous fields can be tackled using different approaches (for a summary see e.g. [28, 29] and references therein).

In our previous work [15], we extended the Time Dependent Schrödinger Equation (TDSE) in reduced dimensions in order to study the harmonic radiation generated by a model atom when it is illuminated by an inhomogeneous electric field, having a linear spatial dependency. Our TDSE in reduced dimensions model is certainly not able to reproduce any atomic structural detail present in a real high-order harmonic spectra, but it could predict exactly the HHG cutoff (for first attempts to study HHG driven by nonhomogeneous fields using a full 3D TDSE approach see [14, 30]). In the present work, we much further improve the model presented in Ref. [15], by including the actual field, which is obtained directly from 3D finite elements simulations where the real parameters of bow-tie shaped nanostructures, as those used in [8], are considered.

The paper is organized as follows. In the next section (Sec. 2) we present our theoretical
2. Theoretical method

For a linearly polarized field, which is the case of our study, the dynamics of an atomic electron is mainly along the direction of the field and as a result it is a good approximation to employ the time-dependent Schrödinger equation in one spatial dimension (1D-TDSE) \[1\] which reads:

\[
\frac{i}{\hbar} \frac{\partial \Psi(x,t)}{\partial t} = \mathcal{H}(t) \Psi(x,t) \tag{1}
\]

\[
= \left[ -\frac{1}{2} \frac{\partial^2}{\partial x^2} + V_{\text{atom}}(x) + V_{\text{laser}}(x,t) \right] \Psi(x,t) \tag{2}
\]

where \( V_{\text{atom}}(x) \) is the atomic potential and \( V_{\text{laser}}(x,t) \) represents the potential due to the laser electric field. In here, we use for \( V_{\text{atom}}(x) \) the quasi-Coulomb potential

\[
V_{\text{atom}}(x) = -\frac{1}{\sqrt{x^2 + \xi^2}} \tag{3}
\]

which first was introduced in \[31\] and has been widely used in the 1D studies of laser-matter processes in atoms. The required ionization potential can be defined by varying the parameter \( \xi \) in Eq. (3). The potential \( V_{\text{laser}}(x,t) \) due to the laser electric field \( E(x,t) \), is given by

\[
V_{\text{laser}}(x,t) = E(x,t)x. \tag{4}
\]

In here the spatial dependency of \( E(x,t) \) can be defined in terms of a perturbation to the dipole approximation and it reads:

\[
E(x,t) = E_0 f(t) \left( 1 + h(x) \right) \sin \omega t, \tag{5}
\]

which is linearly polarized along the \( x \)-axis. In Eq. (5), \( E_0 \), \( \omega \) and \( f(t) \) are the peak amplitude, the frequency of the coherent electromagnetic radiation and the pulse envelope, respectively. Furthermore, \( h(x) \) represents the functional form of the nonhomogeneous electric field and it can be written as a series of the form

\[
h(x) = \sum_{i=1}^{N} b_i x^i \tag{6}
\]

where the coefficients \( b_i \) are obtained by fitting the real electric field that results from a finite element simulation considering the real geometry of different nanostructures. In this work we use for the laser pulse a trapezoidal envelope given by

\[
f(t) = \begin{cases} 
\frac{t}{t_1} & \text{for } 0 \leq t < t_1 \\
1 & \text{for } t_1 \leq t \leq t_2 \\
\frac{(t-t_2)}{(t_3-t_2)} & \text{for } t_2 < t \leq t_3 \\
0 & \text{elsewhere} \end{cases} \tag{7}
\]

where \( t_1 = 2\pi n_{\text{on}} / \omega \), \( t_2 = t_1 + 2\pi n_p / \omega \), and \( t_3 = t_2 + 2\pi n_{\text{off}} / \omega \). In here, \( n_{\text{on}} \), \( n_p \) and \( n_{\text{off}} \) are the number of cycles of turn on, plateau and turn off, respectively.

We use \( \xi = 1.18 \) in Eq. (3) such that the binding energy of the ground state of the 1D Hamiltonian coincides with the (negative) ionization potential of Argon, i.e. \( E_{\text{GS}} = -15.7596 \).
Moreover, we assume that the noble gas atom is in its initial state (ground state (GS)) before we turning the laser \((t = -\infty)\) on. Equation (1) is then solved numerically by using the Crank-Nicolson scheme [1]. In addition, to avoid spurious reflections from the spatial boundaries, at each time step, the electron wave function is multiplied by a mask function [32].

The harmonic yield of an atom is proportional to the Fourier transform of the so-called dipole acceleration \(a(t)\) of its active electron [33]. That is,

\[
D(\omega) = \left| \frac{1}{\tau} \frac{1}{\omega^2} \int_{-\infty}^{\infty} dt e^{-i\omega t} a(t) \right|^2 \tag{8}
\]

with \(a(t)\) is obtained by using the following commutator relation

\[
a(t) = \frac{d^2\langle x \rangle}{dt^2} = -\langle \Psi(t) | [\mathcal{H}(t), [\mathcal{H}(t), x]] \rangle \Psi(t) \rangle. \tag{9}
\]

In here, \(\mathcal{H}(t)\) and \(\Psi(x,t)\) are the Hamiltonian and the electron wave function defined in Eq. (1), respectively. The function \(D(\omega)\) is called the dipole spectrum, which gives the spectral profile measured in HHG experiments. For solving Eq. (1), the gap size \(g\) of the gold bow-tie nanostructure is taken into account by restricting the spatial grid size (see Fig. 1 for a sketch of the gold bow-tie nanostructure including the typical dimensions and the geometry).

### 3. Results and discussion

#### 3.1. Nanostructure

The antenna is formed by two identical (isosceles) triangular gold pads (longest altitude of 600 nm and the smallest acute angle of \(30^\circ\)) separated by an air gap \(g\) (as shown in Fig. 1). The apices at corners were rounded (10 nm radius of curvature) to account for limitation of current fabrication techniques and avoid nonphysical fields enhancement due to tip-effect. The out of plane thickness is set to 25 nm. These parameters yield to a dipolar bonding resonance centered at around \(\lambda = 1800\) nm. This particular value of \(\lambda\) was chosen according to the availability of laser sources [34]. On the other hand, the selected laser wavelength gives the electron a larger excursions and will allow us to better explore the inhomogeneity of the electric field. Classically the electron excursion in an oscillating electric field is given by the so-called quiver radius \(\alpha_0\), which is \(\propto \sqrt{I\lambda^2}\) where \(I\) is the laser intensity. For instance, in here, for intensities \(I\) of \(\sim 10^{14}\) W·cm\(^{-2}\), \(\alpha_0\) can have a value about \(\pm 80\) a.u. (\(\pm 4.5\) nm).

The electric field intensity distribution inside the gap of the gold bow-tie nanoantenna was computed numerically by a 3D finite-element method (COMSOL Multiphysics) [35], using the gold optical properties taken from Ref. [36]. The enhancement of the field depends on the size of the gap; the smaller the gap, the larger the enhancement will be. Based on finite-element calculations, the enhancement factor decays exponentially with the size of the gap. In this paper, we focus on the intermediate region, where there is no such asymptotic behavior, i.e between \(g = 10\) nm and \(g = 20\) nm. In particular, we study HHG for the cases with \(g = 12\) nm and \(g = 15\) nm.

The insets of Figs. 2 and 3 display the calculated electric field intensity enhancement in the gap of the bow-tie structures when illuminated by a linearly polarized (x-axis) plane wave at 1800 nm for \(g = 12\) nm and \(g = 15\) nm, respectively. The field-enhancement profile is extracted for the bow-tie long axis through the middle of the gap, so the successive problem is reduced to 1D. Additionally we normalize the electric field by setting \(E(0,t) = 1\). We observe amplifications of about 39 and 37 dB, between the input intensity and the intensity at center of the gap for \(g = 12\) nm and \(g = 15\) nm, respectively.
3.2. Spectra and time analysis

In a real experiment, the field enhancement would be smaller than our finite-element calculations [8]. As a result, we assume an amplification of 29 dB for the case with $g = 12$ nm case and 30 dB for the $g = 15$. We use a reduction factor to account for the difference between the plasmonic field enhancement obtained in the finite elements results and the one experimentally observed at $\lambda = 800$ nm and not at $\lambda = 1800$ nm, which is the value used in our simulations. Obviously, the reduction factors obtained for $\lambda = 800$ nm not necessarily apply for $\lambda = 1800$ nm. Nevertheless, we think that our estimations are very conservative and that, in fact, potential wavelength dependence of the discrepancy between experimental and theoretical observations should be less pronounced at longer wavelengths. For instance, if e.g. the quality of the nanos-structuring process was responsible for this discrepancy at $\lambda = 800$ nm, this will be less severe at $\lambda = 1800$ nm, where the structures are larger and the field enhancement less sensitive to small fabrication deviations. We will consider a laser with initial input intensities of $1 \times 10^{11}$ W cm$^{-2}$ which would lead to enhanced field of $I = 8 \times 10^{13}$ W cm$^{-2}$ and $I = 1.25 \times 10^{14}$ W cm$^{-2}$ at the center of the spot ($x = 0$), for the former and later cases, respectively. In order to be consistent with our finite-element calculation, in which we used a monochromatic field, we used a trapezoidal shaped pulse with three optical cycles during the ramp up ($n_{on} = 3$) and the ramp down ($n_{off} = 3$), with a plateau with 4 optical cycles ($n_p = 4$), i.e. 10 optical cycles in total which is roughly 60 fs for a wavelength $\lambda = 1800$ nm.

We compute HHG spectra considering an homogeneous electric field, i.e $E(x,t) = E(t)$, and a nonhomogeneous electric field, using Eq. (5). These results are displayed in Figs. 2 and 3 for $g = 12$ nm and $g = 15$, respectively.

For the homogeneous case we have a harmonic cutoff at around $139\omega$ and $204\omega$ as shown by the arrows in Figs. 2 and 3, respectively. In fact, our calculation are in excellent agreement with the semiclassical model [7]. For nonhomogeneous cases, however, we observe a substantial increase in the harmonic cutoff, which is about 50 % higher than the cutoff generated by a conventional homogeneous electric field. This new feature emerges due to the combination of the nonhomogeneous character of the electric field and the confinement of the electron motion [15]. From a theoretical point of view, the spatial grid of our 1D-TDSE defines the
Fig. 2. High-order harmonic generation (HHG) spectra for Ar with ionization potential $E_{\text{GS}} = -0.58$ a.u., laser wavelength $\lambda = 1800$ nm and intensity $I = 8 \times 10^{13}$ W/cm$^{-2}$ at the center of the gap $x = 0$. We use a trapezoidal shaped pulse, Eq. (7), with $n_{\text{on}} = 3$, $n_{\text{off}} = 3$ and $n_p = 4$ (about 60 fs). The gold bow-tie nanostructure has a gap $g = 12$ nm (226 a.u.). The black line indicates the homogeneous case while the red line indicates the nonhomogeneous case. The arrow indicates the cutoff predicted by the semiclassical model for the homogeneous case [7]. The top left inset shows the functional form of the electric field $E(x, t)$, where the solid lines are the raw data obtained from the finite element simulations and the dashed line is a nonlinear fitting. The top right inset shows the intensity enhancement in the gap region of the gold bow-tie nanostructure.

The confinement region, which corresponds to the gap of the bow-tie nanostructure. In principle, for generating HHG the confinement region should be at least on the order of electron excursion. If the confinement region is smaller than the excursion of the electron then the released electron will not have a chance to recombine with its parent ion. On the other hand, for a given wavelength the electron excursion depends on the field intensity. The larger the intensity, the larger the excursion. For the nonhomogeneous system that we are studying in here the electron accelerates in the field which parabolically increases over position. As a result, it will have a larger excursion relative to the case where it accelerates in an homogeneous field.

In the following, we employ time-analysis in order to investigate the harmonic spectra shown in Figs. 2 and 3. We employ the Gabor transformation which was developed in the 1940s by D. Gabor [37]. It has been proven that this technique is appropriate to estimate the emission times of harmonic spectra in atoms and molecules and to discriminate the different electron trajectories [38]. Starting from the dipole acceleration $a(t)$ of Eq. (9), the Gabor transform is defined as

$$a_G(\Omega, t) = \int dt' a(t') \frac{\exp \left[ -(t - t'^2)/2\sigma^2 \right]}{\sigma \sqrt{2\pi}} \exp(i\Omega t')$$

where the integration is usually taken over the pulse duration. In our studies we use $\sigma = 1/3\omega$, with $\omega$ being the central laser frequency. The chosen value of $\sigma$ allows us to achieve an adequate balance between the time and frequency resolutions (see Ref. [38] for details). In Fig. 4 we display the Gabor analysis of the harmonic spectra of Figs. 2 and 3. Panel (a) and (c) represent the homogeneous cases corresponding to Figs. 2 and 3, respectively, while panels (b) and (d)
show their nonhomogeneous counterparts.

For the homogeneous case, both short and long electron trajectories are present and contribute to the harmonic spectra, while for nonhomogeneous cases the long trajectories are absent. In here we confine the electron motion to the region formed by the bow-tie nanostructure, which is in the range of the electron excursion. For the homogeneous case, the electron excursion is smaller than the size of the gap thus the electron excursion along the long trajectories will also be in the allowed limit. For the nonhomogeneous case, the electron excursion is larger thus only the short trajectories will be in the allowed limit. Our classical calculation support this argument as we will discuss this point in the next section.

3.3. Classical analysis

In this section we employ classical and semiclassical tools in order to study the main features present in the quantum-mechanical computed spectra of Sec. 3.2. We first use the semiclassical three-step model [6,7] in order to explain the extension of the harmonic cutoff. As was already pointed out in [15], this new attribute appears as a consequence of the combination of two factors, namely the nonhomogeneous character of the laser electric field and the confinement of the electron motion. It is well-established that the position of the high-order harmonic cutoff obeys

\[ n_c = \frac{(3.17 U_p + I_p)}{\omega} \]  

(11)

where \( n_c \) is the harmonic order at the cutoff, \( \omega \) the laser frequency, \( U_p \) the ponderomotive energy (defined by \( U_p = I/4\omega^2 \), \( I \) being the laser intensity in a.u.) and \( I_p \) the ionization potential of the atom or molecule.

The relationship (11) can be obtained by solving the classical Newton equation for an electron moving in a linearly polarized, e.g. in the \( x \)-axis, electric oscillating field under the following initial conditions: (i) the electron starts at position zero at time \( t = t_0 \) with zero velocity, i.e.

\[ x(t_0) = 0 \]

(12)

\[ \dot{x}(t_0) = 0 \]  

(13)
Here, $t_0$ is known as the birth or ionization time; (ii) when the electric field reverses its direction, the electron returns to its initial position (i.e. the electron *recollides* or recombines with the parent ion) at a later time $t = t_1$, i.e.

$$x(t_1) = 0.$$  

(14)

In Eq. (14) $t_1$ defines the recollision or recombination time. The electron kinetic energy at the $t_1$ time is calculated from

$$E_k(t_1) = \frac{\dot{x}(t_1)^2}{2}$$

(15)

and finding the value of $t_1$ (as a function of $t_0$) that maximizes this energy, Eq. (11) is fulfilled.

We solve the Newton equation numerically for an electron moving in an electric field with the same parameters used in the 1D-TDSE calculations of Section 3.2. We thus find the numerical solution of

$$\ddot{x}(t) = -\nabla_x V_{\text{laser}}(x, t) = \ddot{E}(x, t)$$

(16)

where $\ddot{E}(x, t)$ is the effective electric field along the electron trajectory $x(t)$. In Eq. (16) $V_{\text{laser}}(x, t)$ and $E(x, t)$ are defined by Eqs. (4) and (5), respectively. Fixing the value of ionization time $t_0$ it is possible to compute the classical trajectories and to numerically calculate the recollision times $t_1$, i.e. the $t_1$ when $x(t_1) = 0$.

Additionally, for a given ionization time $t_0$ the electron trajectory is completely determined. For comparison purposes we present here the following set of results, namely, (i) calculations with $E(x, t) = E(t)$, i.e. the homogeneous case and without restriction in the electron motion;
Fig. 5. Total energy of the free electron (in terms of the harmonic order) in the laser field when it recollides with its parent ion obtained from Newton’s second law and plotted as a function of the ionization time (green filled circles) or the recollision time (red open circles). Panel (a) homogeneous case without restriction in the electron motion, (b) non-homogeneous case without restriction in the electron motion, (c) idem (a) restricting the electron motion to the region $[-\alpha_0, \alpha_0]$ and (d) idem (b) restricting the electron motion to the region $[-\alpha_0, \alpha_0]$. The laser parameters are $I = 8 \times 10^{13}$ W cm$^{-2}$, $\lambda = 1800$ nm and a trapezoidal shaped pulse with $n_{on} = 3$, $n_p = 4$ and $n_{off} = 3$. The nonhomogeneous electric field is that corresponding to a bow-tie shaped nanostructure with $g = 12$ nm.

(ii) calculations with the nonhomogeneous fields $E(x,t)$ of Eq. (5) and without restriction in the electron motion; (iii) idem (i) but restricting the electron motion to the region $[-\alpha_0, \alpha_0]$, $\alpha_0$ being the quiver radius defined by $\alpha_0 = E_0/\omega^2$; (iv) idem (ii) but restricting the electron motion to the region $[-\alpha_0, \alpha_0]$. 

In Fig. 5 panels (a)-(d) we plot the dependence of the harmonic order on the ionization time ($t_0$) and recollision time ($t_1$), calculated from $n = (E_k(t_i) + I_p)/\alpha_0$ with $i = 0$ and $i = 1$, respectively, and for the first case presented in Sec. 3.2, i.e. for $g = 12$ nm. Panel (a) is the homogeneous case without restriction in the electron motion (case (i)); (panel b) is the nonhomogeneous case without restriction in the electron motion (case (ii)); (panel c) is the homogeneous case, but now restricting the electron motion (case (iii)); and (panel d) is the nonhomogeneous case by including the restriction in the electron motion (case (iv)). From panel (a) it is possible to observe that the maximum kinetic energy of the returning electron is in perfect agreement with Eq. (11) (no harmonic order beyond $n_c \sim 140$ is reached). On the other hand panel (b) shows how the nonhomogeneities of the field modify the electron trajectories and that no clear high-order harmonic cutoff is observed. This behaviour is consistent with the predictions of the 1D-TDSE simulations presented in [15].

In order to classically simulate the 1D-TDSE results for real gold bow-tie shaped nanostructures, we restrict the classical electron trajectories to the domain $[-\alpha_0, \alpha_0]$. The $\pm \alpha_0$ values represent the starting point of the mask function and consequently a fair comparison is possible. The results are presented in panels (c) and (d) of Fig. 5. From these plots we can argue that only short trajectories contributes to the harmonic radiation. This is related with the elec-
Fig. 6. Idem Fig. 5 but with $I = 1.25 \times 10^{14}$ W cm$^{-2}$ and the nonhomogeneous electric field is that corresponding to a bow-tie shaped nanostructure with $g = 15$ nm.

electron motion restriction, i.e. the confinement we incorporate in the classical simulations that are the classical counterpart of the restriction in the 1D-TDSE spatial grid. Furthermore, a clear high-order harmonic cutoff is now observed for the nonhomogeneous case and its value is in excellent agreement with the 1D-TDSE predictions (see Fig. 2).

We perform similar calculations, but for the case of a gold bow-tie shaped nanostructure with $g = 15$ nm. We present the results in Fig. 6. From panels (a)-(d) similar conclusions to the previous case can be extracted.

Finally, we plot in Figs. 7 and 8 the recollision time $t_1$ of the electron as a function of the ionization time $t_0$ for all the cases considered above. Panels (a) (of both Figs. 7 and 8) represent the free space case, i.e. the electron motion is not restricted, and in panels (b) we confine the electron motion into the region $[-\alpha_0, \alpha_0]$. The long trajectories are those with recollision times $t_1 \gtrsim 4.25$ optical cycles and these are only for the homogeneous case (blue squares ($\square$)) these trajectories are clearly visible. On the other hand, short trajectories are characterized by $t_1 \lesssim 4.25$ optical cycles and these are present for both the homogeneous and nonhomogeneous cases. Our results are consistent with those shown in [14] and [15], but note that in the present work the actual functional form of the electric field is considered. From panels (a) of both figures we observe how the long trajectories are modified by the nonhomogeneities of the field, namely the homogeneous long trajectories (blue squares ($\square$)) with ionization times $t_0$ around the 3.25 and 3.75 optical cycles merge into unique trajectories ($\bigcirc$). Additionally the branch with $t_0 \sim 3.75$ has now ionization times smaller; hence, the time spent by the electron in the continuum increase and consequently a higher amount of kinetic energy is acquired [14].

When the electron motion is restricted, the long trajectories are absent (see panels (b) of Figs. 7 and 8) and only short trajectories develop the harmonic spectrum. Considering the electric field strength at the ionization time for short trajectories is higher than for the long ones and taking into account the ionization rate as a highly nonlinear function of this electric field [39, 40], long trajectories are much less efficient than the short ones. Consequently this fact explains why an harmonic spectrum constituted by modified short trajectories shows an
Fig. 7. Dependence of the semiclassical trajectories on the ionization and recollision times. Non confined case panel (a); confined case panel (b). Blue squares (●), homogeneous case; red circles (○) nonhomogeneous case. The laser parameters are $I = 8 \times 10^{13}$ W cm$^{-2}$, $\lambda = 1800$ nm and a trapezoidal pulse with $n_{on} = 3$, $n_p = 4$ and $n_{off} = 3$. The nonhomogeneous electric field is that corresponding to a bow-tie shaped nanostructure with $g = 12$ nm.

4. Conclusions and outlook

We present high-order harmonic generation of Argon produced by the fields generated when a gold bow-tie nanostructure is illuminated by a short laser pulse of long wavelength. The functional form of these fields is extracted from finite element simulations using both the complete geometry of the metal nanostructure and laser wavelength. We use the numerical solution of the time-dependent Schrödinger equation (TDSE) in reduced dimensions to predict the harmonic spectra. We observe an extension in the harmonic cutoff position that could lead to the production of XUV coherent laser sources and opening the avenue to the generation of attosecond pulses. This new feature is a consequence of the combination of a nonhomogeneous electric field, which modifies substantially the electron trajectories, and the confinement of the electron dynamics. Furthermore, our numerical results are supported by time-analysis and classical simulations. A more pronounced increment in the harmonic cutoff, in addition with an appreciable growth in the conversion efficiency, could be attained optimizing the nanostructure geometry and by choosing the adequate materials. These degrees of freedom pave the way to tailor the harmonic spectra according to specific requirements. On the other hand, the phase mismatch between the generated harmonics and the injected noble gas seems to does not play an important role in these kind of systems and furthermore the conversion efficiency could be improved.
Fig. 8. Idem Fig. 7 but with $I = 1.25 \times 10^{14}$ W·cm$^{-2}$. The nonhomogeneous electric field is that corresponding to a bow-tie shaped nanostructure with $g = 15$ nm.

by considering an array of point-like nanostructures sources.

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