Commun. Comput. Chem. Vol.**2**, No.**2**, pp.47-53
doi: 10.4208/cicc.2014.v2.n2.1 June 2014

COMMUNICATION

Plasmonic Nanostructures Assisted Generation of x-ray Sources

Liqiang Feng[1,2,*] and Hang Liu[3]

[1] *College of Science, Liaoning University of Technology, Jinzhou, 121000, China*
[2] *State Key Laboratory of Molecular Reaction Dynamics, Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian, China, P. R. 116023*
[3] *School of Chemical and Environmental Engineering, Liaoning University of Technology, Jinzhou, 121000, China*

Received 3 April 2014; Accepted (in revised version) 13 May 2014

Abstract: A promising way to generate the isolated attosecond x-ray sources has been theoretically investigated emerging from the concept of nanostructures plasmonic field enhancement. It is found that by properly modulating the inhomogeneity of the input two-color weak field, not only the harmonic cutoff has been extended to the x-ray region, but also the single short quantum path has been selected to contribute to the harmonic. As a consequence, a series of sub-50as attosecond x-ray pulses have been obtained.

AMS subject classifications: 35Q41, 78A10, 81V55

Key words: High-order harmonic generation, x-ray source, plasmonic field enhancement

Ultrashort x-ray pulses are a key tool for probing the ultrafast electronic dynamics in atoms [1,2], attosecond time-resolved spectroscopy [3], and tomographic imaging of molecular orbitals [4,5], etc. High-order harmonic generation (HHG) as the most promising way to produce the isolated attosecond x-ray pulses by direct frequency upconversion of femtosecond near-infrared pulses has been widely investigated in the past two decades [6-8]. Currently, the HHG process can be well depicted in terms of the semiclassical three-step model [9]: ionization, acceleration, and recombination of the electrons in the intense laser field. During the recombination, a maximum harmonic cutoff with $E_{max}=I_p+3.17U_p$ can be obtained, where I_p is the ionization potential and $U_P = I/4\omega^2$ is the ponderomotive energy of the free electron in the laser field.

*Corresponding author. *Email addresses*: lqfeng_lngy@126.com (L.-Q. Feng)

However, due to the limitation of the harmonic cutoff, it is hard to produce the isolated x-ray sources from the single weak (10^{14}W/cm^2) laser pulse. Thus, in the last decade, much effort has been paid to extending the harmonic cutoff, such as the two-color or the three-color field scheme [10-12], the chirp pulse scheme [13], and Thz controlling method [14] etc. However, in most of the above methods, the fundamental fields are also beyond the 10^{15}W/cm^2, which are not easier to be obtained in laboratories.

Recently, the plasmonic field enhancement in the vicinity of metallic nanostructures has attracted a lot of interest [15-18]. As an alternative technique for harmonic generation and extension, it is not necessary to utilize extra cavities to amplify the input pulse power, and the local electric fields can be enhanced by more than 20 dB [19]. The underlying mechanism of the plasmonic field enhancement harmonic emission can be well described as follows: when a low intensity input pulse couples to the plasmon mode, as shown in **Figure 1**. There is a collective oscillation of free charges around the vicinity of the metal nanostructure, and the negative charges are redistributed around one apex and the positive charges around the other one, resulting in a large resonant enhancement of the local field. Consequently, by injecting rare gases into this enhanced field, the HHG can be produced or extended [15,20]. For instance: (i) from the experimental side, Kim et al [15] shows that the output laser field has been enhanced by three orders of magnitude compared with the input pulse (10^{11}W/cm^2), and a 17th order harmonic has been obtained. However, the outcome of the Ref 15, has been put under controversy more recently [16-18], i.e. whether the harmonic emission is in fact the coherent (HHG) or only an incoherent atomic line emission. (ii) From the theoretical side, by using the linearly spatial-dependent laser fields, the generation of even harmonics, the selection of quantum paths and the wavelength dependence of the harmonic yields, etc, have also been investigated [21,22].

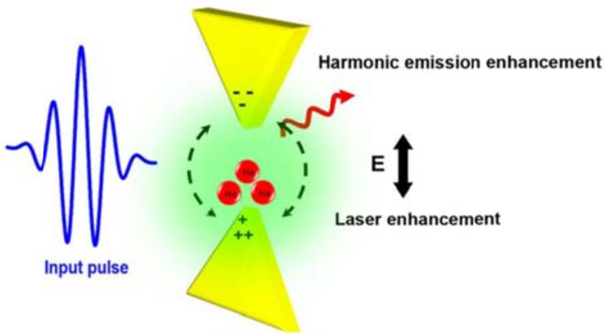

Figure 1: Schematic illustration of linear electric field enhancement and harmonic emission using a nanostructure of bow-tie elements.

However, in these previous studies, the optimal harmonic extension has scarcely been reported. Moreover, the obtained sources are almost around the UV region and the generated pulse durations are much larger than 1 atomic unit of time (24as). Thus, in this paper, based on the numerical solutions of the three dimensional-time-dependent Schrödinger equation (3D-TDSE), we further investigate the harmonic extension by using the two-color weak field combined with the plasmonic field enhancement scheme. The details of the numerical solution of the TDSE can be found in Refs [21-24]. Due to the strong confinement of plasmonic hot spots, the enhanced field is not spatially homogeneous, which can be expressed as $E(t) = f_0(t)E_0(1+\beta r)\cos(\omega_0 t) + f_1(t)E_1(1+\beta r)\,cos(\omega_1 t)$. Here E_i, and ω_i(i=0,1) are the amplitude and the frequency of the 800nm fundamental pulse and the 1200nm controlling pulse. β determines the order of inhomogeneity of the fields and the unit is in the reciprocal length. The envelope function can be expressed as $f_i(t) = \exp[-4ln(2)t^2/\tau_i^2]$, where τ_i (i=0,1) is the pulse durations of the two pulses.

Figure 2(**a**) shows the HHG spectra driven by two-color weak field with different inhomogeneous parameters. The laser fields are chosen to be 5fs/800nm, I_0=5.0×10^{14}W/cm^2 and 10fs/1200nm, I_1=5.0×10^{13}W/cm^2. It shows that with the increasing of the inhomogeneous parameter β, the maximum harmonic cutoff has been remarkably enhanced. However, when β>0.0024 (i.e. β=0.003), we see that the modulation on the harmonic cutoff is remarkably enhanced, which is unbeneficial to the generation of the isolated pulse. Thus, through our calculations, β=0.0024 (which corresponds to an inhomogeneous region of 22.04nm) is the optimal inhomogeneous parameter for the harmonic extension, which resulting in a smooth ultrabroad 372eV bandwidth. According to the three step model, the maximum harmonic cutoff is $E_{max} = I_P + 3.17\, I/4\omega^2$, thus, it is possible to approximately extract the enhanced pulse intensity using a given harmonic cutoff, namely, $I = 4\omega^2/3.17\,(E_{max} - I_P)$. For instance, for the optimal inhomogeneous parameter case (β=0.0024), the maximum harmonic cutoff is E_{max}=317ω_0, and if we choose the single 5fs/800nm homogeneous pulse (β=0.0) as the reference, then the enhanced pulse intensity will be I′=2.45×10^{15}W/cm^2, which is nearly 4.9 times higher than the single 5fs/800nm, I_0=5.0×10^{14}W/cm^2 pulse. To illuminate the advantages for using the plasmon field enhancement scheme, the HHG spectra driven by the above two cases (i.e. two-color with β=0.0024 and single 5fs/800nm I′=2.45×10^{15}W/cm^2, β=0.0) have been shown in **Figure 2**(**b**). Clearly, the maximum harmonic cutoff of the two-color low intensity inhomogeneous field case (solid black line) is as large as the single-color high intensity homogeneous field case (solid red line). However, due to the inhomogeneous effect, the interference structure on the harmonic plateau of the inhomogeneous field case is much smaller than that of the homogeneous field case, which is beneficial to isolated x-ray source generation.

Figure 3 shows the time-frequency distributions of harmonic emission spectra, obtained by using the wavelet transformation of the dipole acceleration a(t) [25]. It shows that there are three main energy peaks on the harmonic emission process for the present laser pulse. Particularly, for the two-color homogeneous field case (β=0.0), as shown in **Figure 3**(**a**), each energy peak receives two similar contributions from the short and the long quantum paths [26], which is responsible for the large interference on the harmonic spectrum. However, with the introduction of the inhomogeneous parameter, i.e. two-color inhomogeneous field with β=0.0024 as shown in **Figure 3**(**b**), not only the maximum harmonic cutoff has been remarkably extended as illuminated in **Figure 2**(**a**), but also the single short quantum path is well selected for harmonic emission, which is responsible for the small modulation on the harmonic plateau and will favorite to support an isolated x-ray pulse generation. **Figure 3**(**c**) shows the harmonic time-frequency of the single-color 5fs/800nm, $I'=2.45\times10^{15}$W/cm^2, β=0.0 case. We see that although the harmonic cutoff is the same as that in the two-color inhomogeneous field with β=0.0024 case, the similar contributions from the two quantum paths are unbeneficial to isolated x-ray source generation.

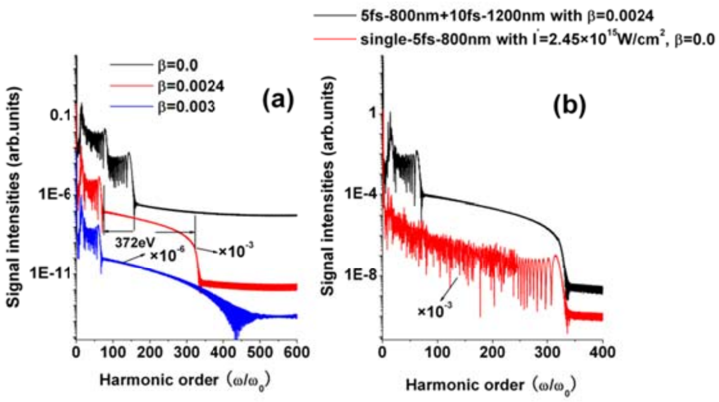

Figure 2: (**a**) Harmonic emission spectra driven by the two-color homogeneous field (β=0.0) and inhomogeneous field with β=0.0024 and 0.003. The laser parameters are chosen to be 5fs/800nm, $I_0=5.0\times10^{14}$W/cm^2 and 10fs/1200nm, $I_1=5.0\times10^{13}$W/cm^2. (**b**) Harmonic emission spectra driven by the above inhomogeneous field with β=0.0024 and the single-color 5fs/800nm homogeneous field with $I'=2.45\times10^{15}$W/cm^2.

Figure 4(**a**) shows the temporal profiles of isolated x-ray pulses. In particular, by properly superposing the optimal harmonics (**Figure 2**(**a**) solid red line) from the 200th to the 250th orders or the 250th to the 300th orders, two isolated x-ray pulses with 45as and 43as can be obtained. To show the advantages for using the plasmonic field enhancement scheme, we also superpose the harmonics of the single-color 5fs/800nm, $I'=2.45\times10^{15}$W/cm^2, β=0.0 case from the 250th to the 300th orders, and two attosecond pulse trains with durations of 64as have been obtained, as shown in **Figure 4**(**b**). Clearly, an isolated x-ray pulse is especially in favor of the field of the time-resolved dynamics measurement. Therefore, the plasmon field enhancement scheme with two-color low intensity pulse is more favorable for the generations of the isolated x-ray sources. It is noted that more recently, one of the authors and coworkers [20] have applied the two-color plasmonic field enhancement scheme to the attosecond x-ray source generation. In that work, the laser source is chosen to be two-color polarized gating with a polarized angle, and the theoretical method is the numerical solution of the 2D-TDSE. Whereas in this work, we presented the more accurate 3D results, and used the two laser pulses with the same polarized direction which is much better for experimental realization. Thus, it is expected that the present two-color pulse scheme may bring useful insight into the generation of attosecond x-ray pulse.

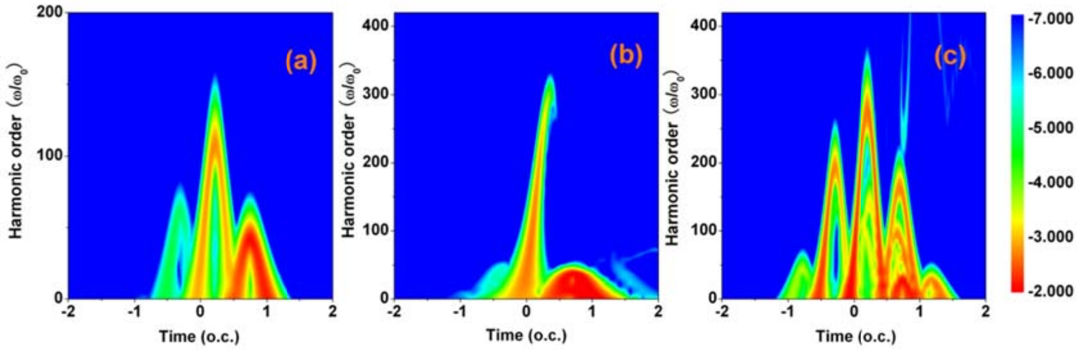

Figure 3: The time-frequency distributions of the harmonic emission spectra for the cases of (a) the above two-color homogeneous field; (b) the above inhomogeneous field with β=0.0024; (c) the single-color 5fs/800nm homogeneous field with I′=2.45×10^{15}W/cm^2.

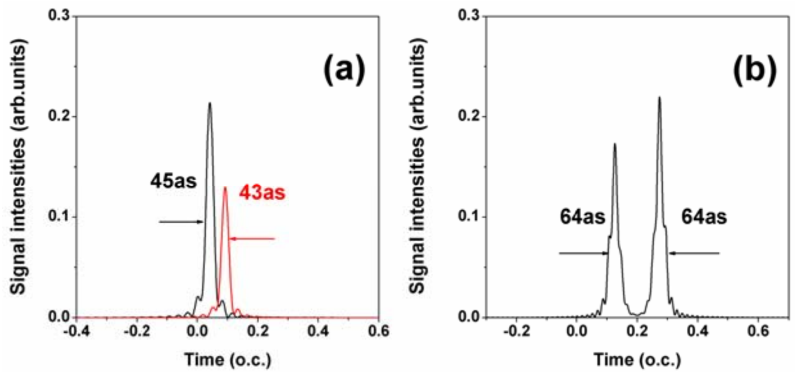

Figure 4: Temporal profiles of the x-ray pulses. The superposed harmonics are chosen (**a**) from the 200th to the 250th orders and from the 250th to the 300th orders of the above inhomogeneous field with β=0.0024 case; (**b**) from the 250th to the 300th orders of the above the single-color 5fs/800nm homogeneous field with I′=2.45×10^{15}W/cm^2 case.

In conclusion, we have theoretically investigated the plasmonic nanostructures assisted harmonic enhancement and the attosecond x-ray pulse generation. The results show that by properly choosing the inhomogeneity of the two-color field, not only the harmonic cutoff has been remarkably extended, but also the single short quantum path has been selected to contribute to the harmonic, further resulting in a series of sub-50as attosecond x-ray pulses, which is much better for experimental realization.

Acknowledgments

This work is supported by scientific research fund of Liaoning University of Technology, China (X201319 and X201312).

References

[1] F. Krausz and M. Ivanov, Attosecond physics, Rev. Mod. Phys., 81 (2009), 163–234.

[2] G. Sansone, E. Benedetti, F. Calegari, C. Vozzi, L. Avaldi, R. Flammini, L. Poletto, P. Villoresi, C. Altucci, R. Velotta, S. Stagira, S. D. Silvestri and M. Nisoli, Isolated single-cycle attosecond pulses, Science, 314 (2006), 443–446.

[3] M. Drescher, M. Hentschel, R. Kienberger, M. Uiberacker, V. Yakovelv, A. Scrinzi, Th. Westerwalbesloh, U. Kleineberg, U. Heinzmann and F. Krausz, Time-resolved atomic inner-shell spectroscopy, Nature, 419 (2002), 803-807.

[4] J. Itatani, J. Levesque, D. Zeidler, Hiromichi Niikura, H. Ppin, J. C. Kieffer, P. B. Corkum and D. M. Villeneuve, Tomographic imaging of molecular orbitals, Nature, 432 (2004), 867-871.

[5] M. Qin, X. Zhu, Q. Zhang and P. Lu, Tomographic imaging of asymmetric molecular orbitals with a two-color multicycle laser field, Opt. Lett., 37 (2012), 5208-5210.

[6] L. Q. Feng, and T. S. Chu, Role of Excited States in Asymmetric Harmonic Emission, Commun. Comput. Chem., 1 (2013), 52-62.

[7] L. Q. Feng and T. S. Chu, Nuclear signatures on the molecular harmonic emission and the attosecond pulse generation, J. Chem. Phys., 136 (2012), 054102.

[8] E. Goulielmakis, M. Schultze, M. Hofstetter, V. S. Yakovlev, J. Gagnon, M. Uiberacker, A. L. Aquila, E. M. Gullikson, D. T. Attwood, R. Kienberger, F. Krausz and U. Kleineberg, Single-cycle nonlinear optics, Science, 320 (2008), 1614-1617.

[9] P. B. Corkum, Plasma perspective on strong field multiphoton ionization, Phys. Rev. Lett., 71 (1993), 1994-1997.

[10] L. Q. Feng and T. S. Chu, High-order harmonics extension and isolated attosecond pulse generation in three-color field: Controlling factors, Phys. Lett. A, 375 (2011), 3641-3648.

[11] Z. Zeng, Y. Cheng, X. Song, R. Li and Z. Xu, Generation of an extreme ultraviolet supercontinuum in a two-color laser field, Phys. Rev. Lett., 98 (2007), 203901.

[12] R. F. Lu, P. Y. Zhang and K. L. Han, Attosecond-resolution quantum dynamics calculations for atoms and molecules in strong laser fields, Phys. Rev. E, 77 (2008), 066701.

[13] L. Q. Feng and T. S. Chu, Generation of an isolated sub - 40 - as pulse using two-color laser pulses: Combined chirp effects, Phys. Rev. A, 84 (2011), 053853.

[14] L. Q. Feng and T. S. Chu, Quantum path control on the harmonic emission in the presence of a terahertz field, Chem. Phys., 405 (2012), 26-31.

[15] 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 (2008), 757-760.

[16] M. Sivis, M. Duwe, B. Abel and C. Ropers, Nanostructure-enhanced atomic line emission, Nature, 485 (2012), E1-E3.

[17] S. Kim, J. Jin, Y. J. Kim, I. Y. Park, Y. Kim and S. W. Kim, Kim et al. reply, Nature, 485 (2012), E1-E3.

[18] I. Y. Park, S. Kim, J. Choi, D. H. Lee, Y.-J. Kim, M. F. Kling, M. I. Stockman and S. W. Kim, Plasmonic generation of ultrashort extreme-ultraviolet light pulses, Nat. Photonics, 5 (2011), 677-681.

[19] P. Mühlschlegel, H. J. Eisler, O. J. F. Martin, B. Hecht and D. W. Pohl, Esonant Optical Antennas, Science, 308 (2005), 1607-1609.

[20] L. Q. Feng, M. H. Yuan, and T. S. Chu, Attosecond x-ray source generation from two-color polarized gating plasmonic field enhancement, Phys. Plasmas, 20 (2013), 122307.

[21] M. F. Ciappina, J. A. Pérez-Hernández, T. Shaaran, L. Roso and M. Lewenstein, Electron-momentum distributions and photoelectron spectra of atoms driven by an intense spatially inhomogeneous field, Phys. Rev. A, 87 (2013), 063833.

[22] I. Yavuz, Gas population effects in harmonic emission by plasmonic fields, Phys. Rev. A, 87 (2013), 053815.

[23] J. Hu, K. L. Han and G. Z. He, Correlation quantum dynamics between an electron and D2+ molecule with attosecond resolution, Phys. Rev. Lett., 95 (2005), 123001.

[24] T. S. Chu, Y. Zhang and K. L. Han, The time - dependent quantum wave packet approach to the electronically nonadiabatic processes in chemical reactions, Int. Rev. Phys. Chem., 25 (2006), 201-235.

[25] P. Antoine, B. Piraux and A. Maquet, Time profile of harmonics generated by a single atom in a strong electromagnetic field, Phys. Rev. A, 51 (1995), R1750.

[26] Y. Mairesse, A. D. Bohan, L. J. Frasinski, H. Merdji, L. C. Dinu, P. Monchicourt, P. Breger, M. Kovačev, R. Taïeb, B. Carré, H. G. Muller, P. Agostini and P. Salières, Attosecond synchronization of high-harmonic soft x-rays, Science, 302 (2003), 1540-1543.