

# High-Order Harmonic Extension and Generation of Single Isolated Attosecond Pulse in Hydrogen Gas by Using Plasmonic Field Enhancement

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**Abstract-** By using numerical solving of the 1D time dependent Schrödinger equation, we present theoretical study of high-order harmonic generation using plasmonic field enhancement. We show that the strong inhomogeneity feature of field enhancement, plays an important role in HHG process and as a result we obtain a short isolated pulse.

**Keywords-** 1D time dependent Schrödinger equation, high-order harmonic generation, isolated attosecond pulse, plasmonic field enhancement.

## I. Introduction

Recent advances in laser technology have enabled the full control of few-cycle optical fields, which have key applications including the production of isolated, attosecond ( $1\text{as}=10^{-18}\text{s}$ ) extreme ultraviolet (XUV) pulses via high-harmonic generation (HHG) [i] and the study of nanosystems in the ultrafast regime [ii,iii].

At the present time, the most well-known theory to understand the HHG is the classical three-step (or *simple man's*) model [iii]. According to this model, the electron first tunnels through the barrier formed by the Coulomb potential and the laser field together. Then it oscillates and accelerates by the laser field. Finally, it can recombine with the parent ion releasing a photon with the maximum energy given by  $E_{\text{cutoff}} = I_p + 3.17U_p$ , where  $I_p$  is the ionization potential and  $U_p$  is the ponderomotive energy of the free electron in the laser field.

Another alternative way of producing attosecond pulses at high repetition rates is to exploit the plasmonic field enhancement occurring in metallic nanostructures [iv, v]. The plasmonic resonance of free electrons boosts the intensity level of a modest femtosecond laser pulse readily by a factor of more than 100, leading to generation of high-order harmonics without reducing the repetition rate [vi–viii].

For high-order harmonic generation to occur, one needs a laser field with intensity greater than  $10^{13}\text{W}/\text{cm}^2$ , two orders of magnitude larger than the output of the current modest femtosecond oscillator ( $10^{11}\text{W}/\text{cm}^2$ ). It means an additional process like chirped-pulse amplification is needed to reach the required intensity for generation of high harmonics using noble gases. Even so, the XUV based on HHG has low duty cycle and efficiency [iii]. The recent demonstration based on surface

Plasmon resonances as light amplifiers could overcome these difficulties [iii,ix]. By maneuvering surface Plasmon resonances, the laser electric fields can locally be enhanced by more than 20 dB [x, xi] without the need of extra cavities or laser pumping process.

The physical mechanism of HHG based on plasmonics can be explained as follows (for full explanation see [iii]). A femtosecond low intensity laser pulse couples to the Plasmon mode and initiates a collective oscillation among the free charges within the metal. This causes a large resonant enhancement of the local field inside and at the nanostructure vicinity. This enhancement is well above the threshold intensity for generating high harmonics. Consequently, by injecting rare gases into the site of the enhanced field, HHG can be produced. In here, the enhanced field is spatially inhomogeneous in the region where electron dynamics take place.

## II. Material and Methodology

In our calculations, the HHG and the attosecond pulse generation can be investigated by solving the one dimensional time-dependent Schrödinger equation (TDSE) based on single-active electron approximation [xii-xiv]. In the dipole approximation and the length gauge, the TDSE is given by (atomic units are used throughout this paper unless stated otherwise)

$$i \frac{\partial \psi(x,t)}{\partial t} = H(t)\psi(x,t) \tag{1}$$

$$= \left[ -\frac{1}{2} \frac{\partial^2}{\partial x^2} + V_A(x) + V_L(x,t) \right] \psi(x,t)$$

Where  $V_A(x)$  is the atomic potential and  $V_L(x,t)$  represents the potential due to the laser electric field. Here, we use for  $V_A$  the "quasi-Coulomb" or "soft-core" potential [xv] for H atom

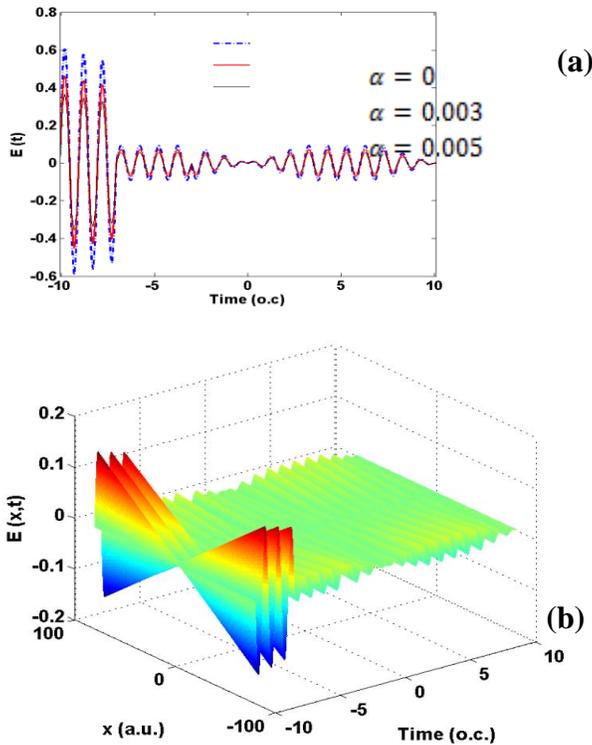
$$V_A(x) = -\frac{1}{\sqrt{(x^2+1)}} \tag{2}$$

The laser potential  $V_L(x,t)$  of the laser electric field  $E(x,t)$  is given by

$$V_L(x,t) = E(x,t)x \tag{3}$$

In Eq. (3), the spatial dependency of  $E(x,t)$  can be defined in terms of a perturbation to the dipole approximation and it reads

$$E(x,t) = E_0 f(t)(1 + \alpha h(x)) \sin(\omega_0 t) \quad (4)$$



**Fig.1.** Electric field for H atom inside the bow-tie nanostructure gap when it has been influenced by linear polarization plane wave with 800 nm wavelength. Panel (a) electric field in  $x = 0$  for  $\alpha = 0, \alpha = 0.003$  and  $\alpha = 0.005$ , panel (b) functional form of the field as function of time and space for  $\alpha = 0.003$ .

which is linearly polarized along the x-axis. In Eq. (4),  $E_0$ ,  $\omega_0$  and  $f(t)$  are the peak amplitude, the frequency of the coherent electromagnetic radiation and the pulse envelope, respectively. In addition,  $h(x)$  represents the functional form of the nonhomogeneous electric field and it can be written as a power series of the form  $h(x) = \sum_{i=1}^N b_i x^i$ . The coefficients  $b_i$  are obtained by fitting the actual electric field that results from a finite element (FE) simulation considering the real geometry of different nanostructures [xvi]. The envelope function is

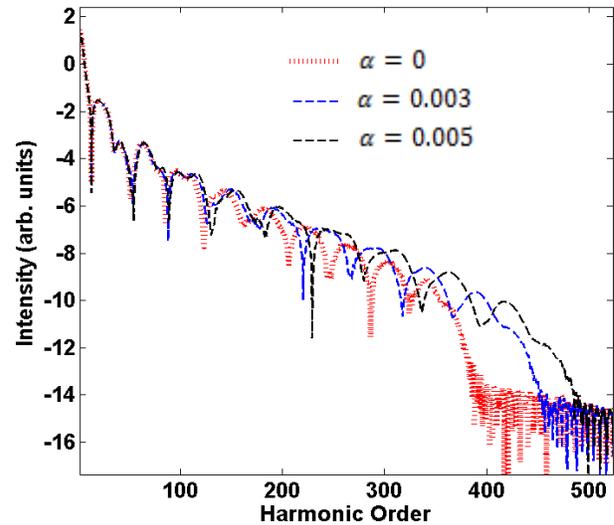
$$f(t) = \begin{cases} \frac{t}{t_1} & \text{for } 0 \leq t < t_1 \\ 1 & \text{for } t_1 \leq t \leq t_2 \\ -\frac{(t-t_3)}{(t_3-t_2)} & \text{for } t_2 < t \leq t_3 \\ 0 & \text{elsewhere} \end{cases} \quad (5)$$

Where  $t_1 = \frac{2\pi n_{on}}{\omega}$ ,  $t_2 = t_1 + \frac{2\pi n_p}{\omega}$  and  $t_3 = t_2 + \frac{2\pi n_{off}}{\omega}$ .  $n_{on}$ ,  $n_p$  and  $n_{off}$  are the number of cycles of turn on, plateau and turn off, respectively.

Equation (1) is solved numerically by using the Crank- Nicolson scheme [xvii]. The time-dependent induced dipole acceleration can be given by means of Ehrenfest's theorem [xviii],

$$d(t) = \frac{d^2 \langle x \rangle}{dt^2} = - \left\langle \psi(x,t) \left[ H(t), [H(t), x] \right] \psi(x,t) \right\rangle \quad (6)$$

In here,  $H(t)$  and  $\psi(x,t)$  are the Hamiltonian and the electron wave function defined in Eq. (1), respectively. And the HHG spectrum  $I(\omega)$  is obtained by Fourier transforming the time-dependent dipole acceleration  $d(t)$ ,



**Fig.2.** High-order harmonic generation (HHG) spectra for a model atom with EGS  $= -0.67$  a.u. generated using the 1D-TDSE. The laser parameters are  $I = 3 \times 10^{14} \text{ w/cm}^2$  and  $\lambda = 800$  nm. We have used a trapezoidal shaped pulse with ten optical cycles, for  $\alpha = 0, \alpha = 0.003$  and  $\alpha = 0.005$ .

$$I(\omega) = |d_\omega|^2 = \left| \frac{1}{\sqrt{2\pi}} \int d(t) e^{-i\omega t} dt \right|^2 \quad (7)$$

Then, attosecond pulse can be generated by superposing harmonics,

$$I(t) = \left| \sum_{\omega} d_\omega e^{i\omega t} \right|^2 \quad (8)$$

Where  $d_\omega = \int d(t) e^{-i\omega t} dt$  represents the inverse Fourier transformation.

### III. Results and Tables

We only consider the linear term of the series for  $h(x)$ , then

$$E(x,t) = E_0 f(t)(1 + \alpha x) \sin(\omega_0 t) \quad (9)$$

where  $\alpha \ll 1$  is a parameter that characterizes the strength of the inhomogeneity and has dimensions of inverse length.

We use  $I = 3 \times 10^{14} \text{ w/cm}^2$ . We have used a trapezoidal shaped pulse with three optical cycles turn on ( $n_{on} = 3$ ) and turn off ( $n_{off} = 3$ ) and a plateau of four constant-amplitude optical cycles ( $n_p = 4$ ).

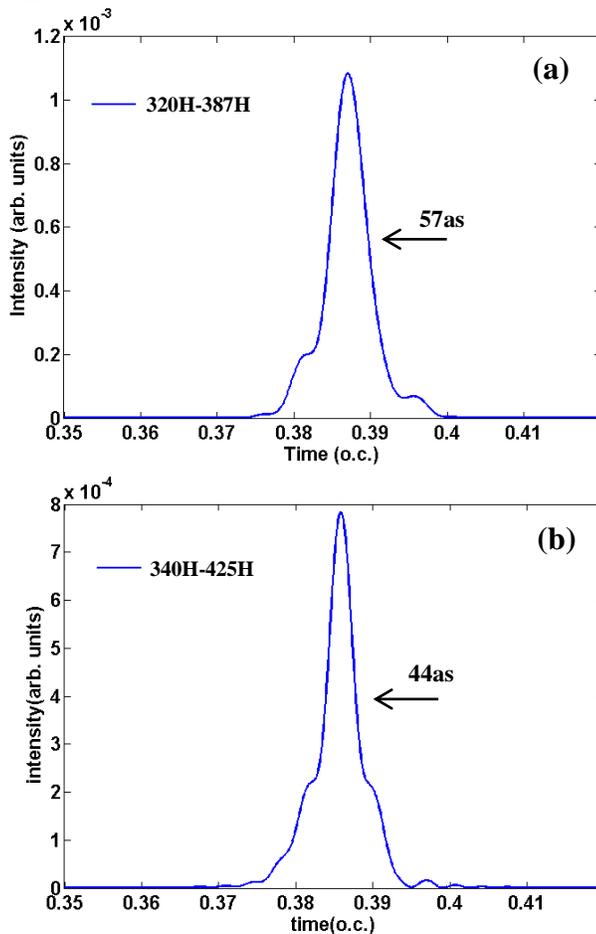


Fig.3. the single isolated attosecond pulse for (a)  $\alpha = 0.003$  by superposing 320th -387th harmonics obtained 57 attosecond and (b)  $\alpha = 0.005$  by superposing 340th -425th harmonics obtained 44 attosecond

Fig. 1(a), shows the electric field for Hydrogen atom inside the bow-tie shaped nanostructure in the origin of the gap for  $\alpha = 0.0$  (blue),  $\alpha = 0.003$  (red) and  $\alpha = 0.005$  (black).

The dependence of electric field on both time and x when  $\alpha = 0.003$  is shown on Fig. 1(b).

At first, we consider field with  $\alpha = 0.0$  then compare this result with result of enhance field. As seen in fig. 2, the cutoff order is in the 365th order of harmonic (red line). By using  $\alpha = 0.003$  and  $\alpha = 0.005$  the cutoff orders are 387th (blue line) and 425th (black line), respectively.

By comparing these results, we can see that by using enhanced field and increasing the inhomogeneity parameter, the cutoff order will increase. Increasing of the cutoff order is significant in producing attosecond pulse, whatever this content increase we have shortest attosecond pulse.

Then, we peruse the generation of attosecond pulses from HHG in enhanced field. Generation profile pulse for  $\alpha = 0.003$  is shown in Fig. 3(a). In this case, we obtain a single isolated 57 attosecond pulse by superposing several consecutive harmonics (320th -387th).

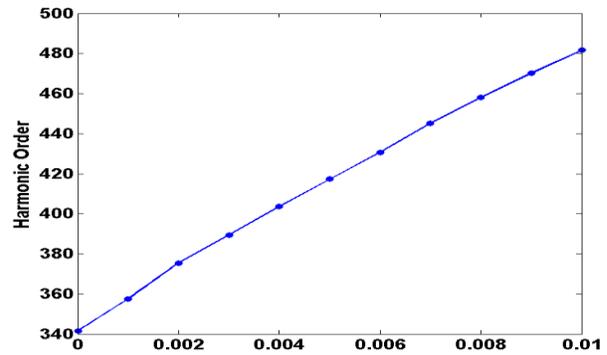


Fig. 4. The variation of the cutoff position of HHG as a function of field inhomogeneity  $\alpha$  between 0 and 0.01 a.u. The parameters are the same as in Fig.2.

Now, by superposing several consecutive harmonics in case  $\alpha = 0.005$  (340th -425th), we can obtain a single isolated attosecond pulse, 44 attosecond, as shown in Fig. 3(b).

Finally, we investigate the dependence of the cutoff on the field inhomogeneity  $\alpha$  of the driving field. The values of  $\alpha$  are chosen between 0.0 and 0.01 a.u. on a coarse mesh with 0.001 a.u. The results are presented in Fig. 4. As it can be seen from this figure, the increase in the order of the inhomogeneity translates into the cutoff extension linearly.

#### IV. Conclusions

When we use inhomogeneous field the cutoff of spectrum is extended. Also by increasing the value of  $\alpha$  parameter, the cutoff order increased. By superposing several consecutive harmonics of this part, a single isolated attosecond pulse produced by ideal time profile. Finally, the cutoff position increased almost linearly as the inhomogeneity factor grew.

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