

# Harmonic generation by atoms interacting with intense ultrashort laser pulses

Nguyen Hong Shon, Akira Suda, and Katsumi Midorikawa  
Laser Technology Laboratory, RIKEN

The interaction of intense ultrashort laser pulses with a gas of argon atoms is numerically investigated. The investigation utilizes a model that solves both the time-dependent Schrödinger equation and the wave equation self-consistently. The time-dependent Schrödinger equation for single-active electron in the effective static potential and in the potential of electron-laser interaction is solved using alternating direction implicit (Peaceman-Rachford) method. The Maxwell's wave equation is solved using an explicit finite-difference technique. We show that, under some conditions, propagation effects can significantly change and clean up the single-atom emission spectrum. In few-cycles regime, a strong fluctuation of the XUV emission in the cut-off region is observed. This is due to change of carrier phase during the propagation.

## Introduction

The availability of intense ultrashort Ti: sapphire laser pulses (less than ten optical cycles) opens up a promising way to generate a coherent x ray in the “water window” region and to produce a single attosecond extreme ultraviolet (XUV) pulse.<sup>1–3)</sup> In this new and fast developing area of nonlinear optics many important phenomena have been observed. For example, unexpected high harmonics were generated in various noble gases by using 25 fs Ti: sapphire laser.<sup>4)</sup> The XUV radiation extending to the “water window” were generated by focusing 5 fs Ti: sapphire laser pulse on He gas<sup>5)</sup> and phase-matched high-harmonic generation in noble gases were demonstrated with 20 fs laser pulses.<sup>6)</sup> However, theoretical investigations of laser-atoms interaction in this nonperturbative, nonadiabatic regime are still behind experimental achievements and represent a challenge for physicists. Rigorously, the theoretical description of harmonic generation involves two aspects: the single-atom process, in which the harmonics are generated, and multi-atom (macroscopic) effects, which occur during propagation of signal through nonlinear medium. The calculated single-atom harmonic spectra in this regime fall to describe the experimental results (compare for example the theoretical<sup>7)</sup> and experimental<sup>4)</sup> results). This leads to a common opinion that propagation effects should play a crucial role in this ultrashort regime. The most direct and extensively used model to treat propagation effects is a slowly-varying envelope (SVE) approximation.<sup>8,9)</sup> The main advantage of the SVE approximation is that it reduces propagation equation to a relatively simple one, which requires not so much computation efforts to be solved. When the rate of ionization is low (less than about 1% per cycle) and the laser pulse is relatively long ( $\simeq 1$  ps) such a method is highly successful at quantitatively reproducing experimental results. However, with pulse duration as short as 30 fs, a SVE approximation is no longer valid and new methods should be used. As far as we know, there exist two approaches in this field: an extended SVE approximation and a self-consistent model. In the first method<sup>10,11)</sup> by using special mapping techniques, the harmonic strength as a function of the laser intensity is extracted from the single-atom spectrum which is then used in the propagation calculations. The advantage of this method is that one has only to perform the single-

atom calculation at one intensity and this remarkably reduces computation times. Using this method, the harmonic spectrum generated by 27 fs laser driver in argon was calculated<sup>10)</sup> which shows a qualitative agreement with the experimental results.<sup>4)</sup> To our best knowledge, this is the only theoretical work which clearly demonstrate how phase matching can clean up the emission spectra during propagation. However, as was mentioned by the authors, their method works well within the adiabatic assumption and the validity of their results may reach the limit when the pulse duration is as short as 27 fs. The self-consistent model (free of this limitation) solves both the time-dependent Schrödinger equation (TDSE) and the wave equation *ab initio*. In this model the single-atom responses must be calculated at each points in the interaction region and for the pulse as a whole. This is a complex and time-consuming program and very few theoretical investigations utilize this method. When the self-consistent treatment is required, in order to reduce computation time, various simplifications (1D TDSE,<sup>12)</sup> strong field approximation,<sup>13,14)</sup> soft Coulomb potential<sup>15)</sup> and short propagation distances *etc.*) are used.

In this paper we utilize the self-consistent model to investigate the harmonic generation in a gas of argon atoms in ultrashort regime. The main improvement of our model when compared with other self-consistent models<sup>12–15)</sup> is that, in single-atom dynamic calculations, we use, for the first time, single active electron model.<sup>16)</sup> This improvement allows to simulate experiments with realistic rare gas atoms. Using this model we are able to show that, propagation effects remarkably clean up the single-atom harmonic spectra providing new one with well-resolved harmonic structure. In the single-cycles regime, the strong fluctuation of XUV emission in the cut-off region was observed. We assume that this phenomenon is due to change of carrier phase during the propagation.

## Method

For modeling the interaction of an atom with an intense linearly polarized laser field (in the  $x$  direction) we use the single active electron (SAE) model.<sup>16)</sup> This model involves the

exact solution of the TDSE for a single electron in the effective static potential and in the time-dependent potential of electron-laser interaction:

$$i\frac{\partial}{\partial t}\psi(\vec{r}, z, t) = \left( -\frac{1}{2}\nabla^2 + V_{eff}(r) + \vec{r} \cdot \vec{E}(z, t) \right) \times \psi(\vec{r}, z, t), \quad (1)$$

Here  $\vec{r}$  denotes the coordinate in the frame of atom and  $z$  defines the position of atoms in the interaction region. The effective potential  $V_{eff}(\vec{r})$  for the active electron is generated from Hartree-Slater calculations for the ground and excited states,<sup>17)</sup> with different potentials for different orbital angular momenta  $l$ . For the argon  $3p$  orbitals, we obtain the  $l = 1$  potential from a ground state calculation and the  $l = 0$  and  $l = 2$  potentials from  $3p^54s$  and  $3p^53d$  calculations, respectively. For  $l > 2$  we use  $l = 2$  potential. In the Hartree-Slater calculations the scaling parameter in the exchange-correlation potential is varied to obtain an accurate ionization energy for the states. Also, the modification of the Hartree-Slater equations to incorporate the correct, long-range Coulomb tail has been included. This method results in quite accurate excited-state potential energies, much better than normally obtained in either Hartree-Fock or Hartree-Slater calculations. We assume that the system is initially in the ground state and express the wave function in spherical harmonics. The Eq. (1) is discretized variationally by a Lagrangian formulation that allows to consider the  $r = 0$  boundary explicitly, then it is solved by using alternating direction implicit (Peaceman-Rachford) method. In all our calculations we use a grid with a maximum radius of 100 a.u. (which is about three time larger than the amplitude of electron oscillation in laser field) and maximum number of partial waves  $L_{max} = 30$ . The grid spacing was 0.1 a.u. and the time step was  $1/2048$  of an optical cycle. To prevent reflection of the wave function from the grid boundary, after each time step, the wave function was multiplied by a  $\cos^{1/8}$  mask function that varied from 1 to 0 over a width of 20 a.u. at the outer radial boundary. The dipole acceleration is given by

$$a(z, t) = -\frac{d^2\langle x \rangle}{dt^2} = -\langle \psi(\vec{r}, z, t) | [H, [H, x]] | \psi(\vec{r}, z, t) \rangle, \quad (2)$$

where  $H$  is the full Hamiltonian.

The simulation of propagation effects utilizes the solution of the wave equation for the linearly polarized plane wave that propagates along the  $z$  axis

$$\frac{\partial^2 \vec{E}(z, t)}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 \vec{E}(z, t)}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \vec{P}(z, t)}{\partial t^2} \quad (3)$$

where

$$\ddot{\vec{P}}(z, t) = N\vec{a}(z, t), \quad (4)$$

$\vec{P}(z, t)$  is the polarization vector and  $N$  is the atomic density.

To solve the wave Eq. (3) we first, change to the moving reference frame  $\tau = t - z/c$ ,  $\xi = z$  and then Fourier transform the equation to frequency domain. The obtained equation is solved numerically using an explicit finite-difference technique. We start with the laser pulse in vacuum region and the polarization vector equals zero. As the laser pulse prop-

agates through an interaction region, the local electric field  $E(\xi, \tau)$  at each grid point can be obtained, which, in turn, is used as the input to the TDSE (1). A Fourier transform of the electric field  $E(\xi, \tau)$  exiting the interaction region yields the harmonic spectrum  $|E(\omega)|^2$ . The temporal profile of laser pulse is obtained by Fourier synthesis of the frequency components  $E(\omega)$  within the band  $\omega_{min} < \omega < \omega_{max}$ . Because of computation limitations we have used a relatively short propagation distance and a high atomic intensity to achieve a similar density-length product of realistic experimental conditions. In our calculations a space step in the propagation direction is  $0.01\lambda/\pi$  ( $\lambda$  is the wavelength of laser driver). The interaction region consists of total 600 atoms. As the atoms are spaced by one grid point, the maximum size of the interaction region is  $6\lambda/\pi$ . To minimize the effect of reflected waves from the plasma, we choose the atomic density is  $2 \times 10^{20} \text{ cm}^{-3}$  which is an order of magnitude less than the plasma critical density (for  $\lambda \simeq 800 \text{ nm}$ ). Our model corresponds to the density-length product 5 Torr-1.7mm, which is typical for rare-gas jet experiments.

## Results and discussions

In Fig. 1 we show the single-atom (a) and propagated (b) and (c) harmonic spectra for gas of argon atoms exposed to the Ti:sapphire laser pulse with the peak intensity  $3 \times 10^{14} \text{ W/cm}^2$  and the pulse width of 21 fs FWHM. The similar single-atom harmonic spectrum (as shown in Fig. 1(a)) was obtained in Ref. 7 for 27 fs laser pulse. We see that only the last few harmonics around the cutoff are distinct and that they are broad and regular. In contrast, there is hardly any harmonic structure left in the plateau region. This is typical single-atom harmonic spectrum of the ultrashort pulse regime. The physical origin of a noisy and almost continuous spectra in the plateau region can be qualitatively understood from a strong field approximation model.<sup>18,19)</sup> For a harmonic belonging to the plateau region, there are several electronic trajectories that contribute to the generation process. One of these trajectories has a phase that varies quite slowly as a function of the laser intensity. It is associated with the nominal (odd) harmonic frequencies. Other trajectories have a rapidly-varying phases and their interference giving rise to an unusual, continuous frequency components. With increasing of laser intensity and/or reducing of pulse length, the contributions of these trajectories increase leading to a noisy and continuous spectra in the plateau region. Since the different trajectories are phase matched differently, it is commonly believed that the nominal (odd) harmonics can be selected by phase matching during propagation. To verify this we propagated the single-atom harmonic spectrum through the nonlinear medium and followed its changes. We have found that at some optimum propagation distance (over 50 atoms in our simulation) the phase mismatch can significantly clean up “noises” (coming from trajectories with rapidly-varying phase), provides a spectrum with well resolved odd harmonics. This situation is shown in Fig. 1(b). If we increase propagation distance further, the phase mismatch begin to affect on nominal harmonics and the spectrum is gradually distorted. The harmonic structure of spectrum disappears again at relatively long distance. It is demonstrated in Fig. 1(c).

In Fig. 2 we plot the single-atom (a) and propagated (b) harmonic spectra for gas of argon atoms exposed to shortest

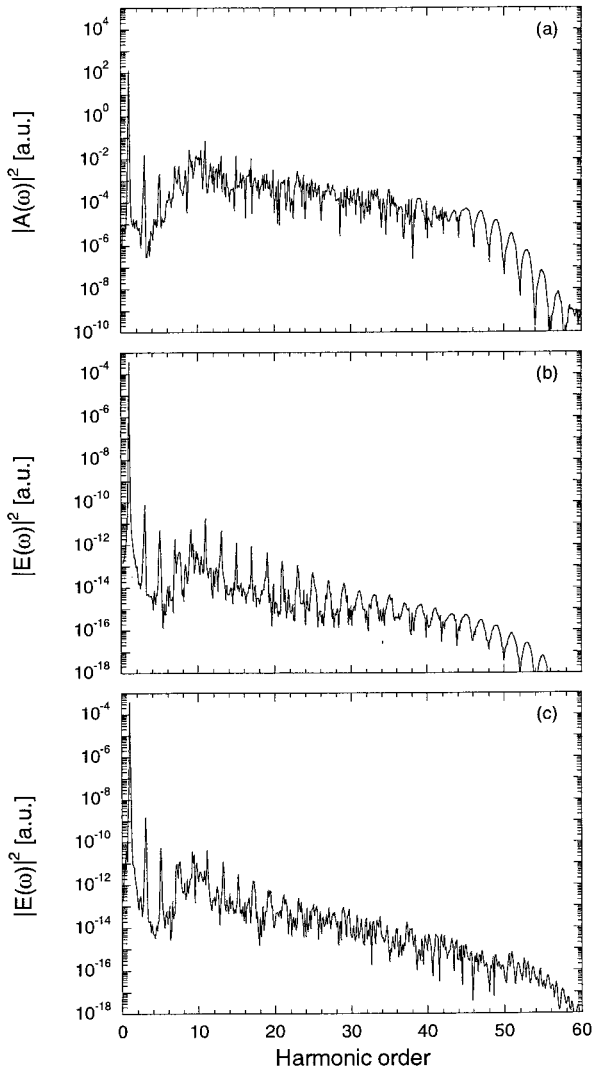


Fig. 1. Single-atom (a) and propagated (b),(c) harmonic spectra for argon atoms exposed to 794 nm laser with the peak intensity  $3 \times 10^{14}$  W/cm<sup>2</sup> and 21 fs FWHM. The propagation distances are 126.4 nm (50 atoms) (b) and 631.6 nm (250 atoms) (c).

(5.3 fs FWHM) Ti: sapphire laser pulse with the peak intensity  $8 \times 10^{14}$  W/cm<sup>2</sup>. In this single-cycle regime, the single-atom spectrum near the cutoff is very smooth and broad and the harmonic structure is not distinct (Fig. 2(a)) (cf. also with the single-atom spectrum for helium atom in the single-cycle regime<sup>20</sup>). After the propagation the single-atom spectrum remarkably changed. The propagated spectrum exhibits strong oscillations with the cutoff is shifted toward longer wavelengths. Figure 2(b) shows the spectrum after propagation the distance of 380 nm (150 atoms). This evolution of the harmonic spectrum during the propagation is very similar to the change of single-atom spectrum when the carrier phase  $\phi$  varies from 0 to  $\pi/2$ .<sup>2,21</sup> (We note that the carrier phase determines the position of the carrier wave relative to the envelope and we use the driver pulse with  $\phi = 0$ .) As was pointed out in these works<sup>2,21</sup> the origin of the phase sensitivity becomes clear from inspection of the time profiles of the harmonic in the cutoff. For  $\phi = 0$  the cutoff harmonics are generated at a single position in the pulse (the main

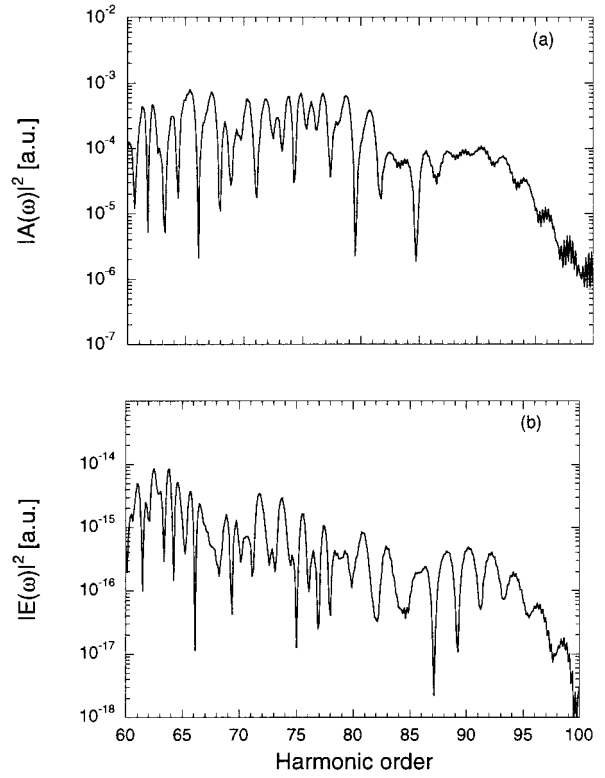


Fig. 2. Single-atom (a) and propagated (b) harmonic spectra for argon atoms exposed to 794 nm laser with the peak intensity  $5 \times 10^{14}$  W/cm<sup>2</sup> and 5.3 fs FWHM. The propagation distance is 379.1 nm (150 atoms).

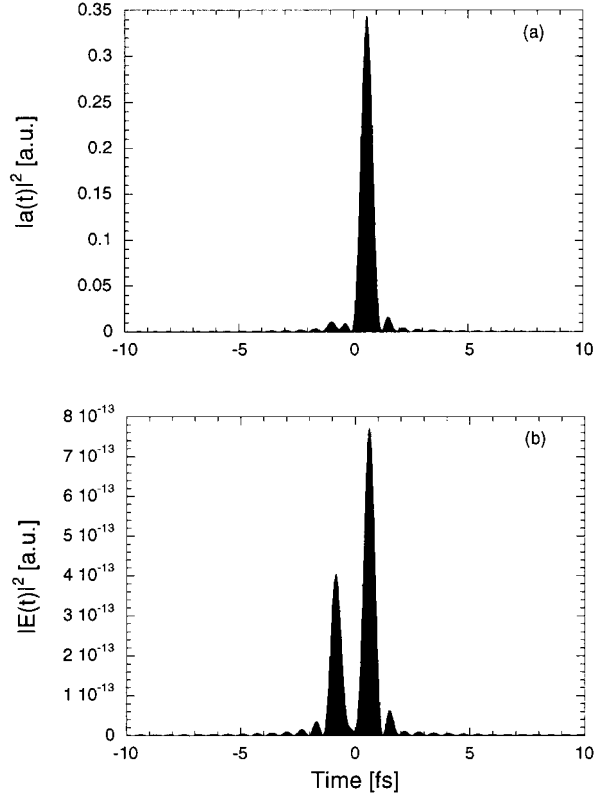


Fig. 3. Time profile of laser pulse obtained by Fourier synthesis of the frequency components between 88th and 92nd harmonics before (a) and after propagation the distance of 379.1 nm (150 atoms)(b). The driver pulse is the same as for Fig. 2.

peak), whereas for  $\phi = \pi/2$  the cutoff harmonics are generated at two positions in the pulse (one before and one after the main peak) that are approximately a half-cycle apart. We have followed the evolution of the cutoff pulse during the propagation and observed similar change. This is illustrated in Fig. 3. Based on these investigations we assume that the carrier phase changes during the propagation and this is the origin of strong fluctuation of the emission in the cutoff region.

In conclusion we have investigated the interaction of a gas of argon atoms with ultrashort laser driver. We have shown that at certain propagation distance, phase mismatch significantly cleans up the single-atom spectrum providing new spectrum with well-resolved harmonic structure. Further increasing of propagation distance distorts or even makes it structureless again. In the single-cycle regime, the strong fluctuation of XUV emission in the cut-off region was observed. This phenomenon is due to a change of carrier phase during the propagation.

The calculations were performed on a Fujitsu VPP700 at RIKEN.

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