

# Chapter 2

## Enhancement of Efficiency of XUV Generation in Atomic Gases Irradiated by Intense Laser Fields

A.V. Andreev, S.Y. Stremoukhov, and O.A. Shoutova

**Abstract** We present the results of the theoretical study of the high-order harmonic generation (HHG) in atomic gases. It is shown that the photoemission spectra exhibit unusual behavior when the laser field strength approaches near-atomic values. In subatomic field strength the cut-off frequency increases linearly with laser pulse intensity. However, when the field strength approaches near-atomic region firstly cut-off frequency slows down and then saturates. To interpret such kind of photoemission spectrum behavior we have proposed the light-atom interaction theory based on the use of eigenfunctions of boundary value problem for “an atom in the external field” instead of the traditional basis of the “free atom” eigenfunctions.

### 1 Introduction

In spite of the twenty-year history, the effect of the HHG is still under a great interest of both experimentalists and theoreticians. From a practical point of view, the HHG is one of the effective mechanisms for producing a coherent emission in broad region of electromagnetic wave spectrum. The presence of plateau region in the harmonic amplitude distribution in extreme ultraviolet (XUV) region affords grounds for development of subfemtosecond pulse formation methods. As a result, the new frontiers are opened up in science by extending the nonlinear optics and time-resolved spectroscopy to the XUV region [1] and pushing ultrafast science to the attosecond domain, enabling XUV spectroscopy and imaging of molecular orbitals [2], surface dynamics [3], and electron motion. The HHG is the reliable route to produce attosecond light pulses [4, 5] and is therefore fundamental to attosecond science [6].

At present days, the efficiency of conversion to high-order harmonics is really too small to consider this emission as a real coherent XUV source for biology, plasma diagnostics, medicine, microscopy, photolithography, etc. Hence, the search

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A.V. Andreev · S.Y. Stremoukhov (✉) · O.A. Shoutova  
Physics Department, M.V. Lomonosov Moscow State University, Leninskie Gory, 1, build. 2,  
119991, Moscow, Russia  
e-mail: [sustrem@gmail.com](mailto:sustrem@gmail.com)

for ways of increasing the cut-off frequency and the HHG efficiency in the XUV spectral range is still among the most topical problems of nonlinear optics.

The HHG effect was observed with the large number of periodic table elements having usually small and middle atomic numbers [7]. Up to now, there has been developed a number of different theoretical models to describe the dynamics of an atomic electron in a strong laser field. These models are based on different approximations, which are usually valid in the restricted area of laser pulse field strength. Here, we use the non-perturbative theory of light-atom interaction [8] which is equally applicable for both weak and strong laser fields.

## 2 Basic Statements of Theory

In the non-relativistic case, the theory of light-atom interaction is based on the time dependent Schrodinger equation TDSE:

$$i\hbar \frac{\partial \psi(\vec{r}, t)}{\partial t} = \left[ \frac{1}{2m} \left( \vec{p} - \frac{e}{c} \vec{A}(t) \right)^2 + U(r) \right] \psi(\vec{r}, t), \quad (1)$$

where  $U(\vec{r})$  is the potential energy of electron in the intra-atomic field, and  $\vec{A}(t)$  is the vector potential of the external electromagnetic wave. The traditional approaches to the analysis of the TDSE are based on the expansion of the wave function in the series of “free atom” eigenfunctions.

The eigensolutions of the boundary value problem for Hamiltonian of the TDSE,

$$\left[ \frac{1}{2m} \left( \vec{p} - \frac{e}{c} \vec{A}(t) \right)^2 + U(r) \right] \varphi_N(\vec{r}, t) = E_N \varphi_N(\vec{r}, t), \quad (2)$$

have been found in [8]. There are the following relationships between the eigenfunctions of the problem (2) and the “free atom” eigenfunctions  $u_n(\vec{r})$ :

$$\varphi_N(\vec{r}, t) = u_n(\vec{r}) \hat{V}^{-1}, \quad \hat{V} = \exp \left( -i \frac{q}{\hbar c} \vec{A}(t) \vec{r} \right). \quad (3)$$

The eigenvalues of the problem (2) coincide exactly with the “free atom” eigenvalues  $E_N = E_n$ .

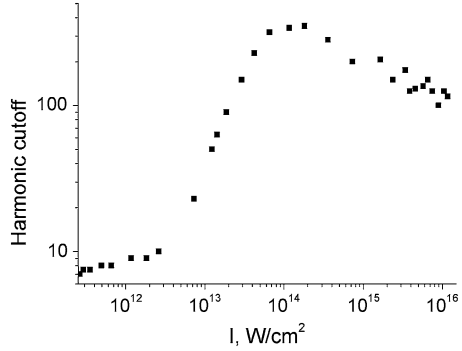
The bases of the eigenfunctions of a “free atom” boundary value problem and the basis of  $\varphi_n(\vec{r}, t)$  are related one-to-one by the transformations [9, 10]. So, we can expand the wavefunction of the TDSE (1) into series of free atom eigenfunctions

$$\psi = \sum_{nlm} a_{nlm}(t) u_{nlm}(\vec{r})$$

and then use the relations of one-to-one transformations [9, 10] to calculate the matrix elements of operators. Executing some evident transformations we get the following set of differential equations for probability amplitudes of discrete and continuum spectrum states

$$i\hbar \frac{da_n}{dt} = \sum_{m,k} V_{nk}^{-1} E_k V_{km} a_m.$$

**Fig. 1** Cut-off frequency as a function of the laser pulse intensity



It is well known, that the number of discrete states of any hydrogen-like atom is infinite and there is an innumerable number of continuum spectrum states. The developed approach provides the consistent mathematical procedure of a selection of atomic levels playing the main role in evolution of a state of the atom in an external laser field.

### 3 Numerical Research

In numerical simulations we assume that a single silver atom interacts with the pulse of Ti:Sapphire laser (the wavelength  $\lambda = 800$  nm). The pulse duration is assumed as  $\tau = 27$  fs. The amplitude of the laser field strength  $E_0$  is varied in the wide range starting from a significantly subatomic to near-atomic values. As we have discussed in our previous work [9] if the laser field strength lies in the interval  $I < 1.83 \cdot 10^{16}$  W/cm<sup>2</sup> then the approximately complete basis of states forms the following 284 levels of a silver atom:  $5s$  (GS),  $5p$ ,  $5d$ ,  $6p$  of the discrete spectrum and 280 states of continuum spectrum with orbital quantum numbers  $l = 0-3$ .

The spectrum of atomic response field in the far-field zone is proportional to the spectrum of the atomic current, which is given by

$$\vec{J}(t) = \frac{q}{m} \sum_{n,m,p,q} a_n^*(t) a_m(t) V_{np}^{-1}(t) \vec{p}_{pq} V_{qm}(t). \quad (4)$$

Our numerical calculations yield a series of harmonic emission spectra. In sub-atomic region the spectrum consists of the sequence of the odd harmonics of incident field carrier frequency. When the laser field strength approaches to near-atomic ones, the shape of the response spectrum is changed. There appear tendencies for growing of its width and formation of plateau. Further increase in the amplitude of the external field strength allows us to observe well-defined plateau with the definite cut-off frequency (CF). The results of computer calculations of the CF as a function of the laser pulse intensity is presented in Fig. 1. One can see that in the weak field range ( $I < 2.9 \cdot 10^{13}$  W/cm<sup>2</sup>) there is the quadratic growth of cut-off frequency with the field strength. However, at laser pulse intensity  $I > 10^{14}$  W/cm<sup>2</sup> the CF

is saturated, i.e. it ceases to be intensity dependent. The reason of such behavior is quite obvious if we take into account that the probability of electron ionization approaches to unity in this region of pulse intensity. It means that the atomic electron is mostly localized in the continuum spectrum states and it does not collide with its parent ion. So, the results of computer modeling show that the most probable reason of the cutoff frequency saturation is the total ionization of the irradiated atom. The behavior of cut-off frequency dependence presented in Fig. 1 is in a good agreement with experimental observed results [7].

## 4 Propagation and Dispersion Effects

In order to illustrate qualitatively the difference between the frequency-angular spectra of a single atom response and an ensemble of atoms we shall use further the simplest dispersion model.

In the case when the atoms of an ensemble are identical, the spectrum of the atomic current (4) is

$$\vec{J}(\vec{k}, \omega) = \sum_{i=1}^N \vec{J}_i(\omega) \exp \left[ i \frac{\omega}{c} (\vec{n}(\omega) - \vec{n}_0(\omega_0)) \vec{r}_i \right] = \vec{J}_0(\omega) f(\vec{k}, \vec{k}_0),$$

where  $\vec{J}_0(\omega)$  is the spectrum of a single atom current and the form factor  $f(\vec{k}, \vec{k}_0)$  is defined by

$$f(\vec{k}, \vec{k}_0) = \int \rho_\omega(\vec{r}) \exp \left[ i \frac{\omega}{c} (\vec{n}(\omega) - \vec{n}_0(\omega_0)) \vec{r} \right] dV. \quad (5)$$

Here,  $\rho_\omega(\vec{r})$  is an effective density of responding atoms, which is the product of density of atoms and the  $\omega/\omega_0$  power of the amplitude spatial distribution of driving laser pulse in irradiated volume.

According to the Lorentz classical theory the refractive index for electromagnetic wave propagating in ionized gaseous medium is given by

$$n(\omega) \approx 1 - \frac{1}{2} \left[ \sum_{\alpha} \frac{\omega_{p\alpha}^2}{\omega^2 - \omega_{0\alpha}^2 + i\omega\gamma_{\alpha}} + \frac{\omega_{pe}^2}{\omega^2 + i\omega\gamma_e} \right],$$

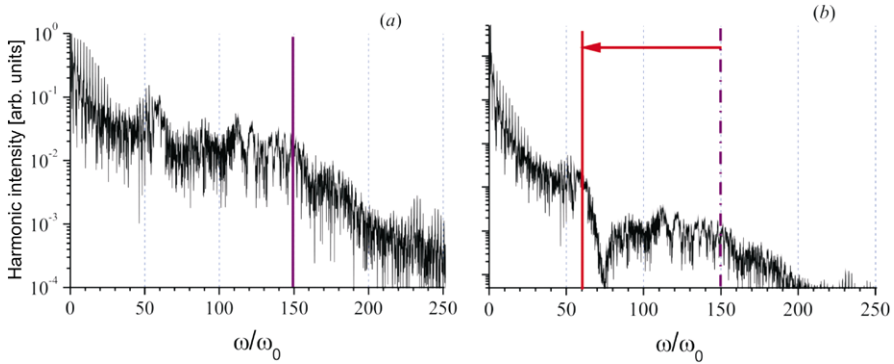
where

$$\omega_{p\alpha(e)} = \sqrt{\frac{4\pi e^2 N_{\alpha(e)}}{mV}}$$

is the plasma frequency corresponding to neutral atoms ( $\alpha = I$ ), ions ( $\alpha = II$ ), and free electrons (e);  $N_{\alpha}$  is the number of neutral atoms and ions in the irradiated volume  $V$ , and  $N_e$  is the number of free electrons.

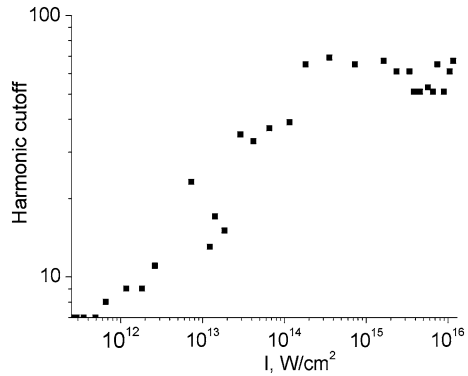
In the case of the Gaussian incident beam the effective density of responding atoms can be approximated as follows

$$\rho_\omega(\vec{r}) = \frac{N_0}{V} \left[ \exp \left( -\frac{z^2}{L^2} - \frac{\rho^2}{d^2} \right) \right]^{\omega/\omega_0},$$



**Fig. 2** The comparison of photoemission spectra generated in silver by a single atom (a) and an ensemble of atoms (b) at the laser pulse intensity  $I = 2.37 \cdot 10^{15} \text{ W/cm}^2$

**Fig. 3** The CF of an ensemble of atoms as a function of a laser pulse intensity



where  $d$  is the focal waist of a driving laser beam, and  $L$  depends both on the confocal parameter of driving laser beam and a spatial distribution of an atomic density in the laser plume produced by the pump laser pulse. We have assumed that the laser beam is propagated along the  $z$  axis.

By taking into account the typical experimental conditions we have calculated the function  $|f(\vec{k}, \vec{k}_0)|^2$  defined by Eq. (5). Figure 2 shows in comparison the single atom response spectrum (Fig. 2a) and spatially distributed ensemble of atoms (Fig. 2b). One can see that the CF in atomic ensemble spectrum is significantly shifted in the long wave region in comparison with the CF in spectrum of single atom response. Figure 3 shows the integral dependency of the CF as function of laser pulse intensity which is calculated for spatially distributed ensemble of atoms. In these calculations we have taken into account that the ionization probability depends on the peak pulse intensity [9].

## 5 Conclusions

In this paper, the saturation of the CF of the photoemission spectra generated in a silver by a femtosecond laser pulses has been demonstrated theoretically. The theoretical analysis is based on the non-perturbative theory of nonlinear atomic response, which enables us to vary the laser field strength in wide region from a subatomic to an over-atomic values. The results of computer simulations have shown that the CF of single atom response is saturated. However, the dispersion effects in spatially distributed ensemble of atoms reduce CF significantly. Hence, the development of the dispersion-suppressed schemes of HHG can play the decisive role in the practical applications of this effect.

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