High harmonic generation and attosecond pulse production in dense medium

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ABSTRACT

We have studied the high harmonic generation and attosecond pulse production in a plasma or gas under conditions when the single-atom response is affected by neighboring ions of the medium. We solve numerically the three-dimensional Schrödinger equation for a single-electron atom in the combined fields of the parent ion, the neighboring particles and the laser, and average the results over different random positions of the particles using the Monte-Carlo method. We observe a change of the harmonic properties due to a random variation of the harmonic phase induced by the field of the medium, when the medium density exceeds a certain transition density. The transition density is found to depend on the harmonic order, but it is almost independent of the fundamental intensity. It also differs for the shorter and longer quantum paths. The latter effect leads to a narrowing of the harmonic lines and a shortening of the attosecond pulses generated using a group of harmonics. The effect of the medium might be important even for much lower densities in the case of XUV generation using radiation in the micron wavelength regime.

Keywords: high-order harmonic generation, attosecond pulses, intense laser field

1. INTRODUCTION

High harmonic generation (HHG) is a fundamental nonlinear process in strong field physics for laser frequency conversion, the generation of coherent intense light at short wavelengths and the generation of attosecond pulses[1] (for a recent review, see[2]). This is due to the fact that coherent radiation is generated at odd multiples of the laser frequency over a broad spectral range. This range is characterized by the universal shape of a high harmonic spectrum with a slow decrease for the first few harmonics, followed by a long plateau region of harmonics having similar intensity, which ends with a sharp cut-off. Assuming equal phases among the harmonics in the plateau region, trains of subfemtosecond pulses can be obtained by the coherent superposition of several harmonics, as it has been demonstrated recently (see, e.g.[3,4]). Also it has been shown, that single subfemtosecond pulses can be generated from harmonics at the cut-off with a few-cycle driving pulse (see, e.g.[5]).

The perspectives and limits of attosecond pulse generation from high harmonic radiation can be understood via an analysis of HHG itself. The standard picture of HHG has been established from theoretical research assuming the interaction of a laser pulse with gases at low pressures, at which the process can be described as a purely single-atom phenomenon. In this approach the effect of the ions and/or atoms in the medium has been considered for the effects of phase-matching and propagation of the harmonics. The main characteristics of HHG can be explained by the semi-classical three-step rescattering model[6,7]. According to this model, the atom gets first ionized by tunneling of an electron through the potential barrier of the combined Coulomb and laser fields. The electron is then accelerated in the field and,

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for linear polarization of the field, may return to and recombine with the parent ion under the emission of a harmonic photon. This interpretation of the process has been confirmed by a quantum mechanical description of HHG[8,9].

Recently, we have studied numerically and theoretically the effect of an ionic or atomic medium on the single atom response in high harmonic generation[10,11]. We have shown that at high densities of the medium the quantum paths of the wave packet will be perturbed by the fields of other ions or atoms in the vicinity. In this respect it is important to note that two different quantum paths of the electron wave packet have been identified for the generation of the harmonics in the plateau. The two paths differ in the time interval between the creation of the wave packet in the continuum and the moment of recombination, and are, hence, usually referred to as short and long quantum paths. In the plateau region of a HHG spectrum the central peak of a harmonic is generated by the contribution of the short quantum path while sidebands appear due to the long quantum path[12, 13]. In the cut-off region both contributions usually strongly interfere and cannot be separated. In a medium the random variation of the harmonic phase induced by the field of the medium leads to a suppression of the HHG signal, when the medium density exceeds a certain transition density. This effect has been observed before in an expanding water micro droplet experimentally[14]. The results of our calculations have shown that the transition density differs by almost an order of magnitude for the two (shorter and longer) quantum paths. This leads to a narrowing of the harmonic lines and a shortening of the attosecond pulses. Below we will review the main results of our studies.

2. NUMERICAL MODEL

The harmonic response of an atom in a medium of ions, induced by a short intense laser pulse, is obtained numerically by solving the 3D time-dependent Schrödinger equation for a single-electron atom in the superposition of the external fields of the ions and a linearly-polarized laser field (Hartree atomic units are used throughout, $e = m_e = \hbar = 1$):

$$i\frac{\partial\psi(\mathbf{r},t)}{\partial t} = \left(\frac{\hat{p}^2}{2} + zF(t) + V_{parent}(\mathbf{r}) + V_{medium}(\mathbf{r})\right)\psi(\mathbf{r},t)\,, \tag{1}$$

with $F = E(t)\sin(\omega(t-T)+\varphi)$ and $E(t) = E_0\sin\left(\frac{t\pi}{2T}\right)$. Here ω is the laser frequency, T is the laser pulse duration (FWHM of the intensity), φ is the absolute phase of the field, and z is the projection of r on the polarization direction of the laser field. V_{parent} and V_{medium} are the potentials of the parent ion and of the medium, given in cylindrical coordinates, (z,ρ), (the origin of the coordinate system is set at the place of the parent ion):

$$V_{parent}(\mathbf{r}) = V_{parent}(z,\rho) = V_{ion}\left(\sqrt{z^2+\rho^2}\right) \tag{2}$$

and

$$V_{medium}(\mathbf{r}) = V_{medium}(z,\rho) = \sum_{j=1}^{M}\frac{1}{2\pi}\int_0^{2\pi}d\phi\; V_{ion}\left(\sqrt{(z-z_j)^2+\rho^2+\rho_j^2-2\rho\rho_j\cos\phi}\right) \tag{3}$$

where $\{z_j,\rho_j\}$ is a set of positions of the ions in the medium, M is the number of particles with $M = nv$, where n and v are the medium density and the volume of the medium, respectively. In the simulations the volume is chosen to be large enough that particles outside of it for sure have no effect on the HHG. Note that such an "axially-symmetric" medium potential, Eq. (3), certainly does not reproduce correctly the effect of the medium on the electron motion in the radial direction. It can be shown[10] that this effect is negligible in comparison with the effect on the longitudinal motion, which is reproduced adequately in Eq. (3).

As gas medium we have considered an ensemble of Ar atoms/ions and used the following potentials:

$$V_{ion}(r) = -\frac{1 + A\exp(-r)}{\sqrt{a^2 + r^2}}\,, \tag{4}$$

with A=5.4 and a=2.125. The potential has similar properties as that one suggested by Muller[15], namely the ground state eigenenergy of an electron bound in the potential reproduces correctly the ionization energy of Ar and the binding energies of the two lowest exited states are close to the corresponding energies in the Ar atom.

The harmonic response in the medium is calculated using the Monte Carlo method. The initial wave-function $\psi(z,\rho,t=0)$ represents an electron in the lowest bound state of the potential (4). A set of positions, $\{ z_j, \rho_j \}_k$, of ions (or atoms) is generated randomly for a given medium density (k is the number of the Monte-Carlo attempt) and the medium potential (3) is calculated. Then, the Schrödinger equation (1) is solved for $t \in [0,2T]$ and the second derivative of the atomic dipole moment (which is proportional to the force acting on the electron) is obtained as:

$$f_k(t) = F(t) - \left\langle \psi(z,\rho,t) \right| \frac{\partial}{\partial z}\left[V_{parent}(z,\rho) + V_{medium}\left(z,\rho,\{ z_j,\rho_j \}_k\right)\right] \left| \psi(z,\rho,t) \right\rangle, \tag{5}$$

which is expanded in a Fourier series $f_k(\Omega)$. To simulate the response of many atoms in the medium we repeat these steps until the Monte-Carlo average,

$$\overline{f(\Omega)} = \frac{1}{N}\sum_{k=1}^{N} f_k(\Omega)\,, \tag{6}$$

has been converged and the harmonic spectrum is then obtained by the coherent response $\left| \overline{f(\Omega)} \right|^2$ of this Monte-Carlo simulation. The attosecond pulses are calculated from the intensity of a harmonic group as a function of time by

$$w(t) = \left| \int_{\Omega_{low}}^{\Omega_{high}} \overline{f(\Omega)} \exp(-i\Omega t) d\Omega \right|^2 . \tag{7}$$

3. RESULTS AND DISCUSSION

We have considered the interaction of an Ar atom with a short laser pulse at the typical Ti:sapphire wavelength, λ=800 nm, and a pulse duration of 50 fs in most of the calculations, otherwise it is stated explicitly. For a 50 fs laser pulse the results certainly do not depend on the absolute phase, for shorter pulse the absolute phase is specified below.

3.1. High harmonic spectra[10]

In Fig. 1 we show a comparison of the harmonic spectra generated in the presence of an ionized background medium having a density of $10^{20}\,\text{cm}^{-3}$ (right hand panel) with those obtained without medium (left hand panel). It is seen from the comparison that in the presence of the medium the typical harmonic spectrum with peaks at the odd multiples of the fundamental frequency is strongly modified. Due to the increase of incoherence in the harmonic response by the variation in the harmonic phase the intensities of the harmonic lines are reduced. It appears that the reduction is as stronger as higher is the harmonic number, which results in a change in the envelope of the spectrum. It is also seen from the Figure that the harmonic lines are much sharper in the presence of the ionic medium than for the single-atom response.

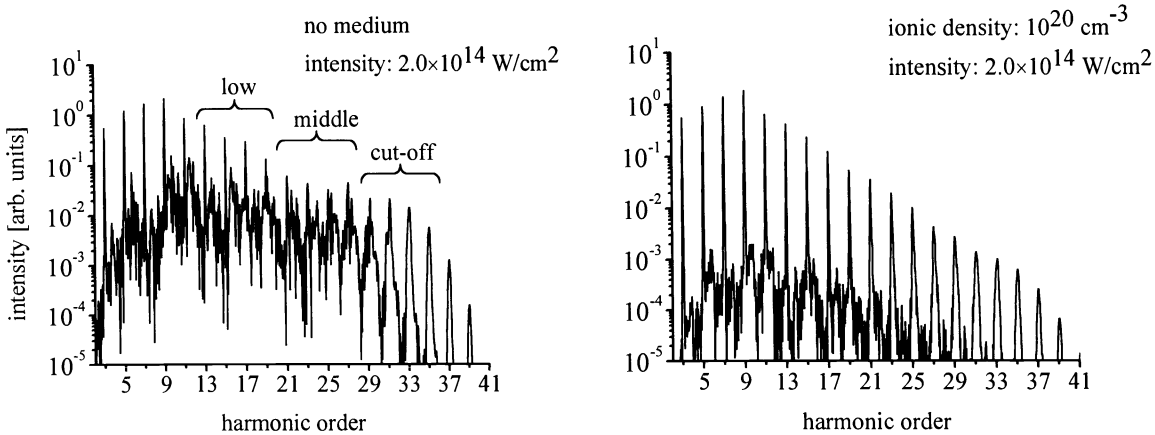

Fig. 1. Harmonic spectra generated under the laser peak intensity 2 x 10^{14} W/cm^2 for a single atom without a medium (left) and in a fully ionized medium of density 10^{20} cm^{-3} (right). Groups of harmonics, used in the analysis below, are presented[10].

We may note parenthetically that the latter effect is due a suppression of the sidebands in each harmonic line, which are generated by the contributions of the long quantum path, while the central peak remains[10]. This leads to an effective narrowing of the harmonic line, and it might be interesting for spectroscopic applications, in which the harmonic lines are often used as a pump pulse to excite the target to a specific excited state, which is then probed by ionization with a second pulse.

The change in the envelope and in the characteristics of the harmonic line is due to a different dependence of the contributions of the two quantum paths on the medium density. This can be clearly seen from the results presented in Fig. 2. We have divided equally the plateau harmonics in three groups, which we denote as lower, middle and cut-off groups (see Fig. 1, left hand panel), such that at a given intensity there is the same number of harmonics in each group. Also, we have separated the contributions of the shorter and the longer quantum paths in the lower and the middle group by identifying the sharp peak in the middle of the harmonic line as the contribution of the shorter path and the sidebands as due to the longer path (the two contributions cannot be separated in the cut-off region). The total energy due to each of the contributions in the three groups is shown in Fig. 2 as a function of the density of an ionic medium. The results are normalized such that the single-atom result (without medium) is set to 1 in each case.

In Fig. 2 one can see that the contributions due to the longer quantum path decrease at a lower ionic density than those due to the shorter quantum path. The difference between the transition densities is larger for the lower harmonic group than for the one from the middle of the plateau, which is consistent with the fact that the difference of the quasi-classical motion along the two paths is most pronounced for the lower plateau harmonics[9,16]. We may define a characteristic transition density as the value at which the efficiency of harmonic generation is decreased twice compared to the single-atom response. It can be seen from the results in Fig. 2 that this density

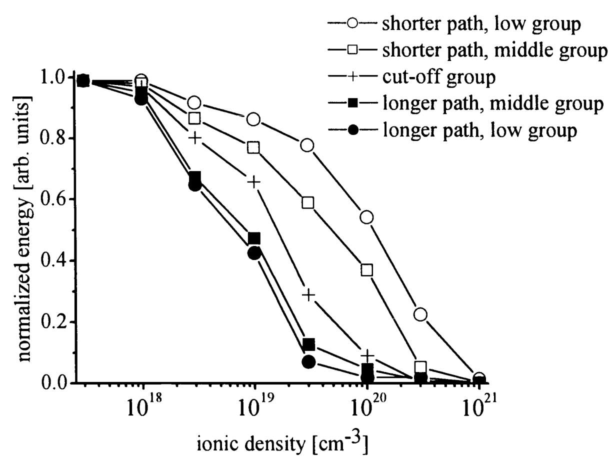

Fig. 2. Total emitted energy of groups of harmonics generated under the peak intensity 2.0 10^{14} W/cm^2 vs density of the completely ionized medium. Contributions of shorter- and longer quantum path are presented separately; groups of harmonics are shown in Fig. 1. The energies are normalized at the energy in the absence of the medium[10].

depends slightly on the harmonic group and ranges between about 10^{19} cm^{-3} (contributions of the longer path) and 4×10^{19} – 10^{20} cm^{-3} (contributions of the shorter path). Note that this difference of about an order of magnitude in the transition densities for the two quantum path should be large enough to allow for experimental observations of the effects discussed below.

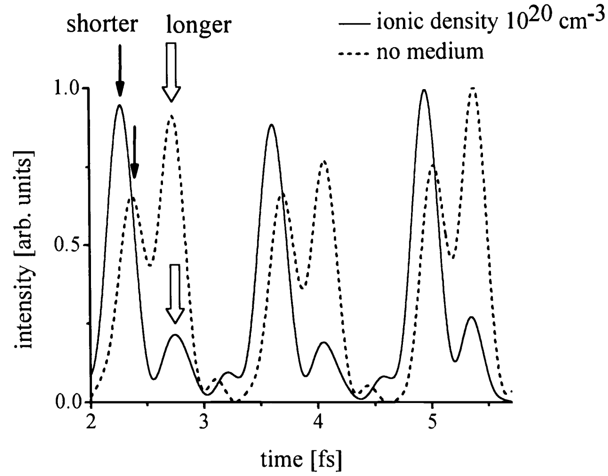

Fig. 3. The XUV intensity of 27th to 37th harmonic-orders vs time generated without medium (dotted line) and in the ionized medium (solid line). Peak intensity of the fundamental is 2×10^{14} W/cm^2. The arrows mark parts of the attosecond pulse mainly generated due to the shorter- and the longer-path contribution, respectively. Every curve is renormalized to its maximum. Origin of the time axis corresponds to the center of the pulse[10].

We may note that our results for the decrease of the coherent harmonic response towards an incoherent emission are in agreement with the observations in a recent experiment[14] on HHG in water micro droplets. The value for the transition density is in a good order-of magnitude agreement with the experimental results. A more detailed comparison would require primarily the theoretical investigation of HHG in a medium of water molecules as well as a higher accuracy of the measurements.

The transition density found in our studies is very sensitive to the fundamental frequency[10], namely it decreases rapidly with the frequency decrease. Thus, the effects of the medium on HHG considered in this paper can be important even under moderate medium densities for harmonic generation using several-micron wavelength fundamental, used recently by several groups[17,18].

3.2. Attosecond pulses[10,11]

As we mention at the outset it has been shown that attosecond pulse trains as well as single attosecond pulses can be obtained using the radiation of a group of harmonics. Usually two pulses (pulse trains) are generated due to the contributions from the shorter and the longer quantum path, respectively (see e.g.[16]). As shown above, at a medium density in the transition regime the two contributions are influenced differently, while the contribution of the long quantum path is suppressed strongly, that one of the shorter path is nearly unaffected. Thus, the attosecond pulses should be also modified. We have calculated the attosecond signal from groups of harmonics in the plateau as well as from the cut-off harmonics, also few-cycle pulses are considered. In all cases we observe a shortening of the attosecond pulses due to the suppression of the contribution from the longer quantum path[10,11], examples are shown in Fig. 3, 4.

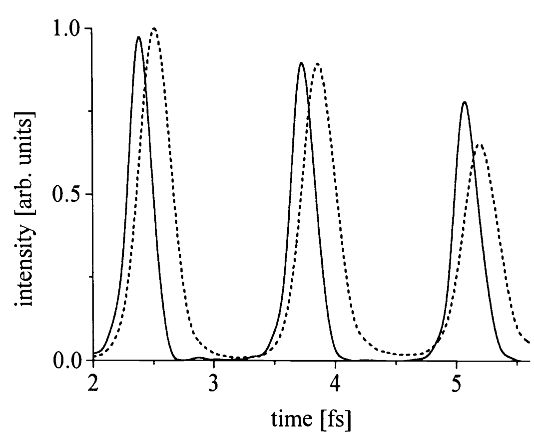

Fig. 4. The XUV intensity of the highest harmonic-orders (31-st and higher) vs time. Other parameters are the same as in Fig. 3. Every attosecond train is renormalized to its maximum[11].

First, we consider the signal from a group of harmonics, namely using harmonics from $\Omega_{low} = 26\omega$ to $\Omega_{high} = 38\omega$. The results of the calculations for a medium density of $10^{20}\,\text{cm}^{-3}$, peak laser intensity of 2×10^{14} W/cm² and laser pulse duration of 50 fs are presented in Fig. 3 (solid line). In the absence of the medium one can recognize the two trains of attosecond pulses (dotted line) generated due to the contributions from the two quantum paths. The two trains are partially superimposed, resulting in a train of pulses with duration of more than a femtosecond. As expected, in the presence of the ionized medium with a density in the transition region one of the trains is strongly suppressed and, indeed, a single attosecond pulse train, generated by the shorter path contributions, arises.

The train of the attosecond pulses obtained from the cut-off harmonics (namely using radiation in the frequency rande from $\Omega_{low} = 30\omega$ to $\Omega_{high} = 48\omega$) is presented in Fig. 4, the laser parameters are the same as in Fig. 3. One can see that in the presence of the medium mainly the front of the attopulse "survives". This leads again to the shortening of the pulse: in the absence of the medium the pulse duration is about 310 as, while in its presence it is about 230 as. Although shorter and longer quantum paths contributions can not be strictly separated for the cut-off harmonics, most likely the origin of this shortening is the same as in the previous figure, namely, the difference of the medium effect on the different quantum paths. Attopulse production using the cut-off harmonics is especially important, since a *single* attopulse has been obtained via these harmonics using a few-cycle fundamental laser pulse[5].

So we see that under certain conditions using medium densities higher than about $3\text{x}10^{18}$ cm^{-3} a shortening of attosecond pulses seems to be feasible. In this paper we did not consider the effect of the phase-matching on the XUV generation, which also can favor a contribution of a certain quantum path. Depending on the experimental parameters, either the effect of the phase-matching or the phenomena considered in this paper may play a dominant role in the influence of a medium on the XUV generation.

4. CONCLUSIONS

We have investigated high harmonic and attosecond pulse generation from an argon atom in a dense medium. Results are obtained by solving the Schrödinger equation for an atom in the combined fields of the laser and of the ionic background medium numerically and calculating the Monte-Carlo average for sets of randomly located ions. Significant changes in the harmonic response from the single-atom result (without medium) are found in a transition regime between 10^{19} and 10^{20} cm^{-3}. Due to the random variation of the harmonic phase induced by the external field of the neighboring particles on the free motion of the electron wave packet the harmonic lines are suppressed. Most interestingly, the contributions of the shorter quantum path to the harmonic lines are found to be affected at higher densities than those of the longer quantum path. This leads to a shortening of the attosecond pulses in a train as well as of a single attosecond pulse at certain transition densities. For instance, the attosecond pulse duration obtained from the cut-off group is shortened by about 80 as to 230 as in the presence of the ionic medium with the density 10^{20} cm^{-3}. The considered phenomena can be important even under much lower densities for XUV generation using several-micron wave-length fundamental.

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