Laser Physics, Vol. 15, No. 6, 2005, pp. 799–803.
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Generation of Attosecond Pulses in a Dense Medium

V. V. Strelkov[1, 2, *], V. T. Platonenko[3], and A. Becker[1, **]

[1] Max-Planck-Institut für Physik komplexer Systeme, Nöthnitzer Str. 38, D-01187 Dresden, Germany
[2] General Physics Institute, Russian Academy of Sciences, ul. Vavilova 38, Moscow, 119991 Russia
[3] Moscow State University, Physics Department, Vorob'evy Gory, Moscow, 119992 Russia
*e-mail: v-strelkov@yandex.ru
**e-mail: abecker@mpipks-dresden.mpg.de
Received December 24, 2004

Abstract—Attosecond pulse emission via high harmonics generated in a dense medium is studied theoretically. We solve numerically the three-dimensional Schrödinger equation for a single-electron atom in the combined field of the neighboring ions and the laser and average the results over different random positions of the particles using the Monte Carlo method. The XUV spectra are calculated for different medium densities. A change in the harmonic properties is seen when the medium density exceeds a certain transition density, where the single-atom response is affected by neighboring ions of the medium. The transition density differs for the two (shorter and longer) quantum paths by about an order of magnitude. The latter effect leads, for ionic densities in the transition regime, to a shortening of the attosecond pulses. This is exemplified for the generation of attosecond pulse trains as well as of a single attosecond pulse in a few-cycle pulse.

1. INTRODUCTION

High-harmonic generation (HHG) is a fundamental nonlinear process in strong-field physics for 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. The broad 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 cutoff. 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 has been demonstrated recently (e.g., [3, 4]). Also, it has been shown that single subfemtosecond pulses can be generated from harmonics at the cutoff with a few-cycle driving pulse (e.g., [5]).

An understanding of the perspectives and limits of attosecond pulse generation from high-harmonic radiation requires a thorough analysis of HHG itself. The current 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 surrounding medium, ions and/or atoms, has been considered for the phase-matching effects and the propagation of the harmonics only. It is a widely accepted consensus that the main characteristics of HHG can be explained by the semiclassical three-step rescattering model [6, 7]. First, the atom is ionized by tunneling of an electron through the potential barrier of the combined Coulomb and laser fields, followed by the acceleration of the electron in the laser field, during which, for linear polarization of the field, the electron may return to and recombine with the parent ion with the emission of a harmonic photon. This interpretation of the process has been confirmed by a quantum-mechanical description of HHG [8, 9].

It is important to note that two different quantum paths of the electron wavepacket 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 wavepacket 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 [10–11]. In the cutoff region, both contributions usually strongly interfere and cannot be separated.

Recently, we have shown [13] in numerical calculations that, at high medium densities, the quantum paths of the wavepacket will be perturbed by the fields of other ions or atoms in the vicinity. Above a certain transition density, this leads to a suppression of the HHG signal, as has also been experimentally observed in an expanding water microdroplet [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 is interesting, since it may lead to a narrowing of the harmonic lines and a shortening of the attosecond pulses.

In this paper we analyze theoretically the generation of attosecond pulses in a partially pre-ionized Ar gas under high pressure. Under these conditions, the harmonic generation is affected by neighboring ions, as we will discuss below. The harmonic response of the Ar atom is found by numerically solving the three-dimensional Schrödinger equation for a single-electron atom in the combined fields of the laser and of the neighboring ions. The results are averaged over different random positions of the ions using the Monte Carlo method. We concentrate then on the application of the high harmonics to attosecond pulse generation. For the driving laser, both long pulse durations of up to several tens of femtoseconds as well as few-cycle pulses are considered. The results, calculated at the transition densities of the medium, are compared with those generated by the single atom (without a medium), and the shortening of the attosecond pulses in the presence of the medium is discussed.

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 three-dimensional time-dependent Schrödinger equation for a single-electron atom in the superposition of the external fields of the ions and a linearly polarized laser (Hartree atomic units are used throughout; $e = m_e = \hbar = 1$):

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

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 $\mathbf{r}$ 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 position of the parent ion):

$$V_{\text{parent}}(\mathbf{r}) = V_{\text{parent}}(z, \rho) = V_{\text{ion}}(\sqrt{z^2 + \rho^2}) \quad (2)$$

and

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

where $\{z_j, \rho_j\}$ is a set of positions of the ions in the medium and M is the number of particles with $M = n\nu$, where n and ν 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 surely have no effect on the HHG. Note that such an "axially symmetric" medium potential (Eq. (3)) certainly does not correctly reproduce the effect of the medium on the electron motion in the radial direction (considered in [12]). It can be shown [13] that for the generation conditions used below this effect is negligible in comparison with the effect on the longitudinal motion, which is reproduced adequately in Eq. (3).

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

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

with $A = 5.4$ and $a = 2.125$. The potential has properties similar to the potential suggested in [15]; e.g., 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 wavefunction $\psi(z, \rho, t = 0)$ represents an electron in the lowest bound state of potential (4). A set of random positions $\{z_j, \rho_j\}_k$ of ions (or atoms) is chosen for a given medium density (k is the number of the Monte Carlo attempt), and medium potential (3) is calculated. Then, Schrödinger Eq. (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:

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

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

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

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

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

3. RESULTS AND DISCUSSION

Before discussing the influence of an ionic medium on the generation of attosecond pulses, we first present results of our numerical simulations for HHG. We have considered the interaction of an Ar atom with a short laser pulse at the typical Ti:sapphire wavelength,

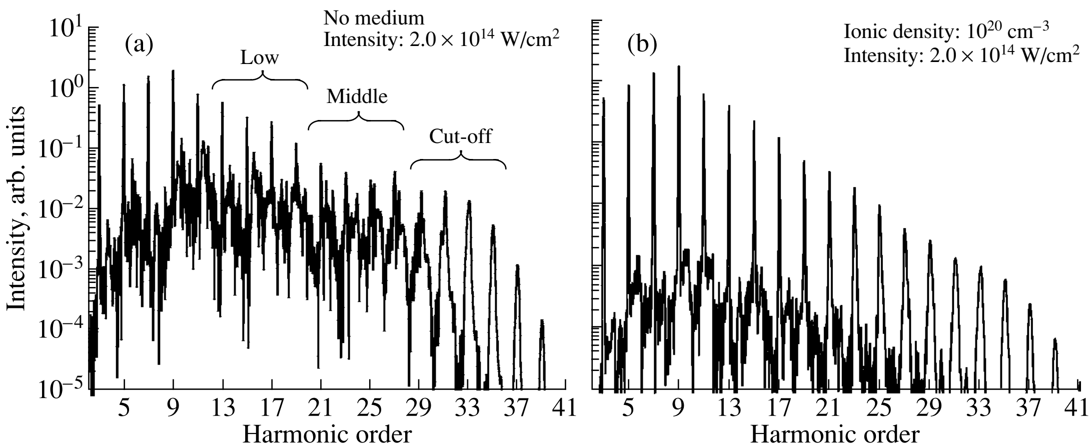

Fig. 1. Harmonic spectra generated under the fundamental peak intensity 2×10^{14} W/cm^2 for a single atom without a medium (a) and in a fully ionized medium of density 10^{20} cm^{-3} (b). Groups of harmonics, used in the analysis below, are indicated.

namely, $\lambda = 800$ 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 a shorter pulse, the absolute phase is specified below.

3.1. High-Harmonic Spectra

In Fig. 1 we show a comparison of the harmonic spectra generated in the presence of an ionized background medium with a density of 10^{20} cm^{-3} (right-hand panel) with those obtained without a 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 in incoherence in the harmonic response by the variation in the harmonic phase, the intensities of the harmonic lines are reduced. It appears that the higher the harmonic number, the stronger the reduction, which results in a change in the envelope of the spectrum. It is also seen from Fig. 1 that the harmonic lines appear to be much sharper in the presence of the ionic medium than for the single-atom response.

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 [13]. This leads to an effective narrowing of the harmonic line and might be interesting for spectroscopic applications, where 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 seen from the results presented in Fig. 2. We have equally divided the plateau harmonics into three groups, which we denote as the lower, middle, and cutoff 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 cutoff 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

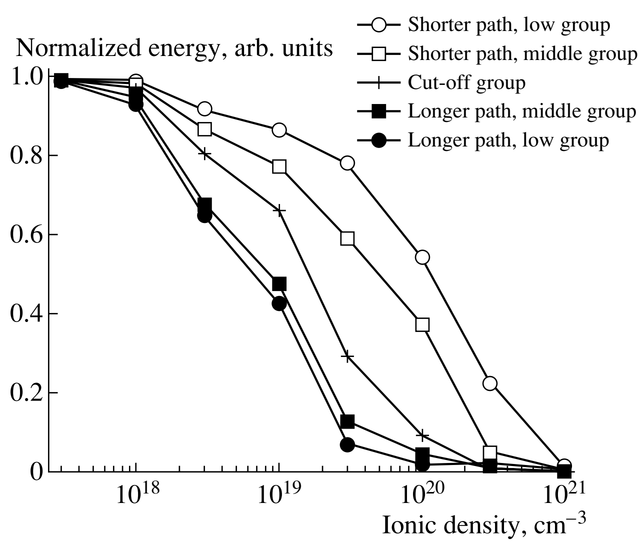

Fig. 2. Total emitted energy of groups of harmonics generated under the peak intensity 2.0×10^{14} W/cm^2 vs. density of the completely ionized medium. Contributions of shorter and longer quantum path are presented separately; groups of harmonics are as indicated in Fig. 1. The energies are normalized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

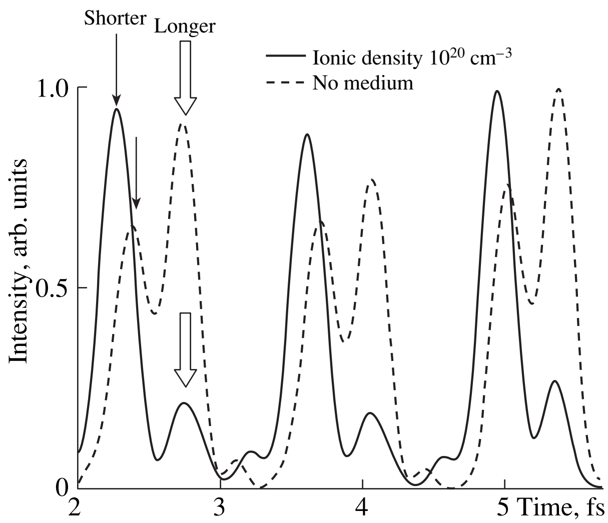

Fig. 3. Intensity of the 27th to 37th harmonics vs. time generated without a medium (dotted line) and in the ionized medium (solid line). Peak intensity of the fundamental is 2×10^{14} W/cm^2. Arrows mark parts of the attosecond pulse mainly generated due to the shorter and the longer path contributions,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

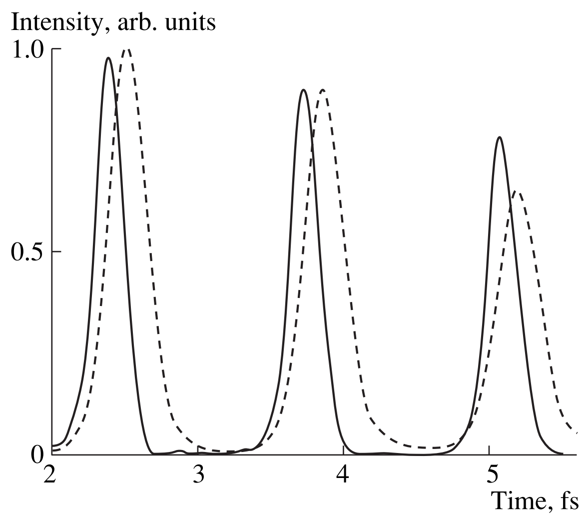

Fig. 4. Intensity of the highest harmonics (31st and higher) vs. time. Other parameters are the same as in Fig. 3. Every attosecond train is renormalized to its maximum.

the single-atom result (without a medium) is set to 1 in each case.

In Fig. 2 one can clearly 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 half that of the single-atom response. It can be seen from the results in Fig. 2 that this density 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 paths should be large enough to allow for experimental observations of the effects discussed below.

We may note that our results for the decrease in the coherent harmonic response towards incoherent emission are in agreement with the observations in a recent experiment [14] on HHG in water microdroplets. The numerical value for the transition density is in 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.

3.2. Attosecond Pulses

As was mentioned 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 was 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 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 cutoff harmonics, and few-cycle pulses are also considered. In all cases, we observe a shortening of the attosecond pulses due to the suppression of the contribution from the longer quantum path [13]; examples are shown in Figs. 3–5.

First, we consider the signal from a group of harmonics, namely, $\Omega_{low} = 26\omega$ and $\Omega_{high} = 38\omega$. The results of the calculations for a medium density of 10^{20} cm^{-3}, a laser peak intensity of 2×10^{14} W/cm^2, and a 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 a 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.

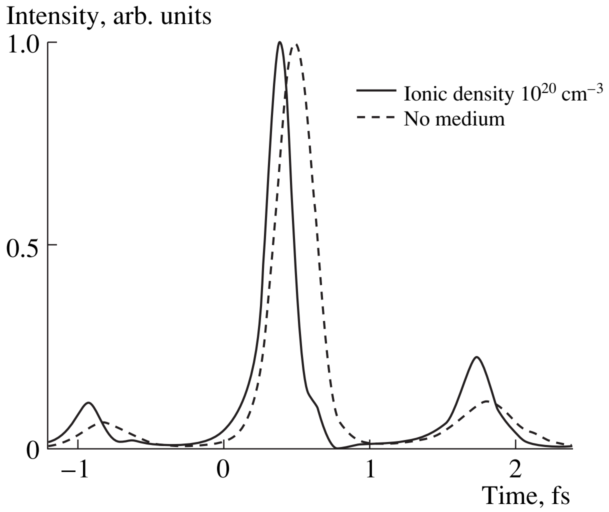

Fig. 5. The same as in Fig. 4 but using a 5-fs-long "cos-like" fundamental pulse with peak intensity 2×10^{14} W/cm^2.

The train of attosecond pulses obtained from the cutoff harmonics (namely, using $\Omega_{low} = 30\omega$ and $\Omega_{high} = 48\omega$) is presented in Fig. 4; the laser parameters are the same as in Fig. 3. We 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 path contributions can not be strictly separated for the cutoff 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 cutoff harmonics is especially important, since a *single* attopulse has been obtained via these harmonics using a few-cycle fundamental laser pulse [5]. Therefore, we also have performed calculations with a 5-fs-long fundamental having an absolute phase of $\varphi = \pi/2$ ("cos-like" pulse). Results are presented in Fig. 5. One can see that a single attopulse is generated both in the presence and the absence of the medium; however, in the presence of the medium, the satellite attopulses are more pronounced. The attopulse generated in the presence of the medium is again shorter than without a medium, as in the previous figures.

Thus, under certain conditions and using medium densities higher than about 3×10^{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 by calculating the Monte Carlo average for sets of randomly located ions. Significant changes in the harmonic response from the single-atom result (without a 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 wavepacket, 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 cutoff group is shortened by about 80 as to 230 as in the presence of an ionic medium with a density of 10^{20} cm^{-3}.

ACKNOWLEDGMENTS

Financial support from the Max-Planck Society and the Russian Foundation for Basic Research (grant no. 02-02-16563) is greatly acknowledged.

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