# High-harmonic generation in a dense medium

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The high-harmonic generation in a plasma or gas under conditions when the single-atom response is affected by neighboring ions or atoms of the medium is studied theoretically. We solve numerically the three-dimensional Schrödinger equation for a single-electron atom in the combined fields of the neighboring particles and the laser, and average the results over different random positions of the particles using the Monte Carlo method. Harmonic spectra are calculated for different medium densities and laser intensities. 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 two (shorter and longer) quantum paths. The latter effect leads for ionic densities in the transition regime to a narrowing of the harmonic lines and a shortening of the attosecond pulses generated using a group of harmonics.

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## I. INTRODUCTION

High-harmonic generation (HHG) is one of the fundamental nonlinear processes in strong-field physics. Due to the interaction with an intense short laser pulse a gas of atoms or molecules emits coherent radiation at odd multiples of the laser frequency. The harmonic spectrum has a universal characteristic shape with a slow decrease for the first few harmonics, followed by a plateau region of harmonics having similar intensity, which ends with a sharp cutoff. Harmonic frequencies can exceed $300\omega$ (e.g., [1]), where $\omega$ is the fundamental laser frequency. Therefore, HHG provides an efficient source of ultrashort coherent radiation in the vacuum ultraviolet and soft x-ray spectral range. Applications of such light sources are time-resolved x-ray spectroscopy and microscopy as well as nanoscale lithography and holography. Further, subfemtosecond pulse trains can be obtained by a superposition of several harmonics from the plateau region (e.g., [2,3]) or single subfemtosecond pulses can be generated from harmonics at the cutoff (e.g., [4]).

The current understanding of HHG has been established from theoretical research assuming the interaction of a laser pulse with gases at low pressures. In this case the process can be described as a purely single-atom (or single-molecule) phenomenon, neglecting the effects of the other ions or atoms in the neighborhood. The surrounding medium is considered for the phase-matching effects and the propagation of the harmonics only. For the isolated atom the main characteristics of HHG can be explained by the semiclassical three-step (or rescattering) model [5,6]. According to this model, first, the atom gets ionized by tunneling of an electron through the potential barrier of the combined Coulomb and laser fields, then the electron is accelerated in the laser field, and finally for linear polarization of the field it may return to the parent ion and recombine under the emission of a harmonic photon. An important feature of the model is that it predicts correctly the universal cutoff of the harmonic spectrum at $I_p+3.17U_p$ where $I_p$ is the ionization potential of the atom and $U_p=E^2/4\omega^2$ is the quiver energy (in hartree atomic units) of a free electron in the laser field of an amplitude $E$. A quantum-dynamical description of HHG [7,8] has confirmed the interpretation provided by the semiclassical three-step model. Moreover, the quantum model revealed the role of two different quantum paths of the electron wave packet for the generation of the harmonics in the plateau region. The two paths differ in the time interval between the moment of creation of the wave packet in the continuum (tunneling) and the time instant of recombination. They are usually referred to as short and long quantum paths (or shorter and longer electron trajectories in the semiclassical three-step model). In the plateau region of the spectrum the contributions of the two paths can be distinguished: the central peak of a harmonic is generated by the contribution of the short quantum path while the sidebands appear due to the long quantum path [8–10]. On the other hand, in the cutoff region the two contributions usually strongly interfere and cannot be separated.

Thus, the HHG process is essentially determined by the evolution of the electron wave packet in the continuum. In the usual single-atom approximation this evolution is governed by the oscillating field of the laser and the field of the parent ion only. One may expect, however, that at sufficiently high intensities of the laser and 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. This impact of the surrounding medium on the high-

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harmonic spectrum is not well studied up to now, although it is interesting in many respects. First, the efficiency of HHG depends on the number of generating particles, and thus in particular on the medium density. Therefore, a high medium density is desirable, but naturally the unique characteristics of the radiation should not be affected by the influence of the ionic (atomic) neighborhood. It is therefore essential to get knowledge about a maximum tolerable medium density to achieve highest efficiencies; some estimations of this density were done in [11]. Next, emission of radiation from an extended dense medium, namely, an expanding water microdroplet, has been studied in an experiment recently [12]. A transition from pure incoherent plasma emission to coherent HHG was observed, when the particle density in the microdroplet decreased. These observations indicated the existence of an optimum medium density for HHG. Finally, in a recent numerical study using a one-dimensional model [13] it has been found that in a partially ionized medium harmonic energies well beyond the single-atom cutoff can be generated. These harmonics are identified to be generated when an electron emitted by one atom is captured by a neighboring ion. Such an extension of the harmonic spectrum would be certainly of much interest in view of its applications.

In this paper we study theoretically HHG in a gas or a plasma of relatively high pressure, namely, under conditions when the harmonic generation due to the rescattering mechanism is affected by neighboring ions (or atoms). The harmonic response is found by solving numerically the three-dimensional Schrödinger equation for a single-electron atom in the combined fields of the laser and of the neighboring ions or atoms, and the results are averaged over different random positions of the ions or atoms using the Monte Carlo method. We concentrate here on the effects of the ionic (atomic) medium on the atomic response and do not consider other macroscopic effects, such as phase matching, etc. Harmonic spectra are calculated for different medium densities and are compared with those generated by the single atom (without medium). As will be seen below, there is a *transition* medium density above which the coherent harmonic response differs in the shape of its envelope and shows a decrease in intensity compared to the low-density single-atom predictions. The numerical results for the transition density will be compared with simple analytical estimations and the observations in a recent experiment. An investigation of its dependence on the fundamental intensity and the harmonic order as well as on the different quantum paths will allow us to obtain further insight into the physics of HHG in a dense medium. Finally, we will discuss the impact of our findings on applications of high harmonics in spectroscopy and attosecond pulse train generation.

## II. NUMERICAL MODEL

The harmonic response of an atom in a medium, 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 medium and a linearly polarized laser (hartree atomic units are used throughout; $e=m_e=\hbar=1$):

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

where $F$ is the instantaneous value of the laser field:

$$F=E(t)\cos(\omega t), \tag{2}$$

with

$$E(t)=E_0\sin\left(\frac{t\pi}{2T}\right). \tag{3}$$

$\omega$ is the laser frequency, $T$ is the laser pulse duration (full width at half maximum of the intensity), and $z$ is the projection of $\mathbf{r}$ on the polarization direction of the laser field. $V_{\text{parent}}$ and $V_{\text{medium}}$ are the potentials of the parent ion and of the medium, which are given in cylindrical coordinates $(z,\rho)$, as (the origin of the coordinate system coincides with the parent ion)

$$V_{\text{parent}}(\mathbf{r})=V_{\text{parent}}(z,\rho)=V_{\text{ion}}(\sqrt{z^2+\rho^2}) \tag{4}$$

and

$$V_{\text{medium}}(\mathbf{r})=V_{\text{medium}}(z,\rho)=\sum_{j=1}^{M}\frac{1}{2\pi}\int_0^{2\pi}d\phi\times V_{\text{ion,atom}}(\sqrt{(z-z_j)^2+\rho^2+\rho_j^2-2\rho\rho_j\cos\phi}) \tag{5}$$

where $\{z_j,\rho_j\}$ is a set of positions of the ions or atoms in the medium, and $M$ is the number of particles with $M=nv$, where $n$ is the medium density and $v$ is the volume of the medium. In the simulations this volume is chosen to be much larger than the computational box. It has been tested that the volume is large enough that particles outside of it have no effect on the HHG. Note that such an "axially symmetric" medium potential, Eq. (5), certainly does not reproduce correctly the effect of the medium on the electron motion in the radial direction; however, as will be discussed below, this effect is negligible in comparison with the effect on the longitudinal motion, which is reproduced adequately in Eq. (5).

As gas medium we have considered an ensemble of argon atoms or ions. For the ionic and atomic potentials in Eqs. (4) and (5) we have, therefore, used approximate potentials of the form

$$V_{\text{ion}}(r)=-\frac{1+A\exp(-r)}{\sqrt{a^2+r^2}}, \tag{6}$$

with $A=5.4$ and $a=2.125$, and

$$V_{\text{atom}}(r)=\begin{cases}-B\exp[-(r/b_1)-(r/b_2)^2], & r\leqslant 1.7,\\ -\dfrac{D}{(d^2+r^2)^2}, & r>1.7,\end{cases} \tag{7}$$

with $B=3.012$, $b_1=1.11$, $b_2=2.09$, $D=5.357$, and $d=1.05$. The ionic potential Eq. (6) has similar properties to the one suggested in [14]. The ground-state eigenenergy of an electron bound in this potential reproduces correctly the ionization energy of Ar and the binding energies of the two lowest

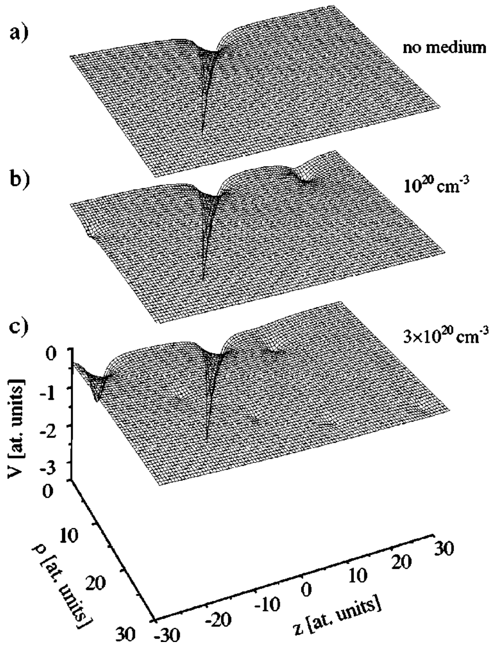


FIG. 1. Samples of the potential $V_{\text{parent}}(z,\rho)+V_{\text{medium}}(z,\rho)$ for certain random realizations of the ion positions, generated for the ionic medium density equal to zero (a), $10^{20}$ cm$^{-3}$ (b), and $3\times10^{20}$ cm$^{-3}$ (c).

excited states are close to the corresponding energies in the argon atom. The atomic potential Eq. (7) coincides with the ionic potential in the vicinity of the origin and reproduces [15] the attraction of the electron to the argon atom for large $r$ due to polarization of the atom.

In order to calculate the harmonic response in the medium we have used the Monte Carlo method. A set of random positions $\{z_j,\rho_j\}_k$ of ions (or atoms) is generated for a given medium density ($k$ is the number of the Monte Carlo attempt) and the medium potential (5) is calculated. In Fig. 1 we present samples of the potential $V_{\text{parent}}(z,\rho)+V_{\text{medium}}(z,\rho)$ in the vicinity of the origin for certain random sets of positions of the ions. The initial wave function $\psi(z,\rho,t=0)$ is chosen to represent an electron in the lowest bound state of the potential (6). Then, the Schrödinger equation (1) is solved and the second derivative of the atomic dipole moment (which is proportional to the force acting on the electron) is obtained as

$$f_k(t) = F(t) - \langle \psi(z,\rho,t)| \frac{\partial}{\partial z}[V_{\text{parent}}(z,\rho) + V_{\text{medium}}(z,\rho,\{z_j,\rho_j\}_k)]|\psi(z,\rho,t)\rangle, \quad (8)$$

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

$$\overline{f(\Omega)} = \frac{1}{N}\sum_{k=1}^{N} f_k(\Omega) \quad (9)$$

converges. The required number of runs, $N$, in our calculations ranged from several tens to several hundreds depending on the medium density, for instance, $N=20$ for the density $10^{18}$ cm$^{-3}$ and $N=320$ for the density $10^{20}$ cm$^{-3}$. The harmonic spectrum is then given by the coherent response $|\overline{f(\Omega)}|^2$ of this Monte Carlo simulation.

## III. RESULTS AND DISCUSSION

### A. Estimation of the transition density

Before proceeding with the results of the numerical calculations, we estimate analytically the density of a fully ionized medium, which is sufficient to affect the high-harmonic spectrum significantly. To this end, we consider the phase of a harmonic with frequency $\Omega$, generated due to the propagation of a wave packet in the continuum, which is given by ([16]; see also [17] and references therein)

$$\varphi(E,E_{\text{ext}}) = -\frac{1}{2}\int_{t_i}^{t_r} p^2(t)dt - I_p\tau + \Omega t_r, \quad (10)$$

where $t_i$ and $t_r$ are the time instants of ionization and recombination, respectively, and $p$ is the mechanical momentum of the electron in the combined fields of the laser, $E$, and an ionic background (medium), $E_{\text{ext}}$,

$$p(t) = \frac{E}{\omega}(\sin\omega t - \sin\omega t_i) - (t-t_i)E_{\text{ext}}. \quad (11)$$

$I_p$ is the atomic ionization potential and $\tau=t_r-t_i$ is the time of free motion of the electron. Note that we have considered in Eq. (11) the component of the external field due to the ion attraction along the polarization of the laser field only. We may further stress that $p(t)$ as well as $t_i$ and $t_r$ depend on the strengths of both fields.

Random distribution of the positions of the ions leads to a random variation of the phase, which will be as larger as stronger is the external field $E_{\text{ext}}$ and, hence, as larger is the density of the medium. Therefore, an increase of the density of the medium will result in a transition from a coherent harmonic response to an incoherent plasma emission. We may define a transition density by the concentration of ions, at which the coherent harmonic response is reduced approximately by a factor of 2, compared to the case of single-atom response without ionic background ($E_{\text{ext}}=0$). This is satisfied if

$$|\varphi(E,E_{\text{ext}}) - \varphi(E,E_{\text{ext}}=0)| = \pi/4. \quad (12)$$

Approximating the attractive field of the ionic background by a homogeneous field

$$E_{\text{ext}} = 1/R^2, \quad (13)$$

where $R=n^{-1/3}$ is the interparticle distance and $n$ is the density of the medium, we obtain from Eq. (12), after lengthy but otherwise elementary mathematics, the transition density $n_{\text{tr}}$ as

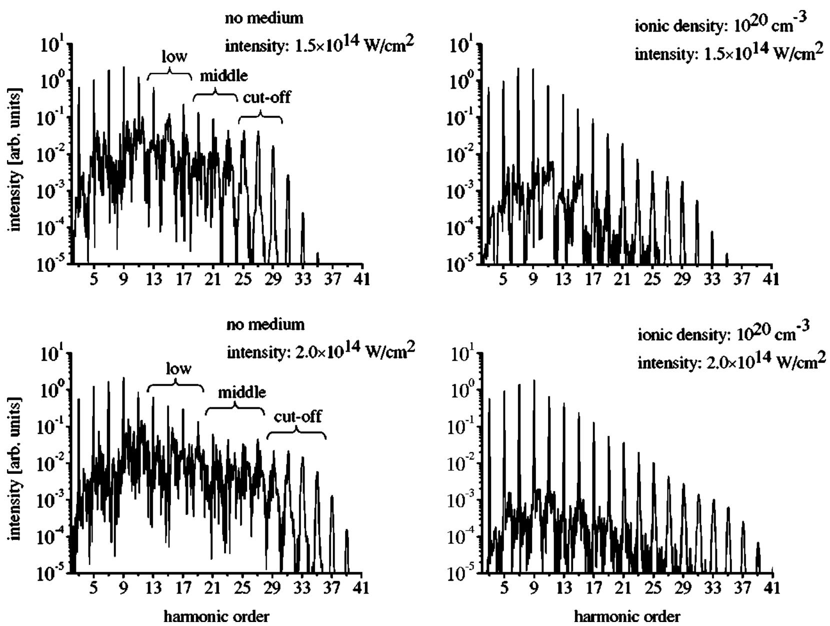


FIG. 2. Harmonic spectra generated from an Ar atom at intensities $1.5\times10^{14}$ (upper row) and $2\times10^{14}$ (lower row) $\mathrm{W/cm^2}$ without medium (left column) and in a fully ionized medium of density $10^{20}\ \mathrm{cm^{-3}}$ (right column) are compared. Groups of harmonics used in the further analysis below are marked.

$$n_{\mathrm{tr}} = \left[ \left| \frac{4}{\pi} \left( \frac{I_p \tau[2p(t_r) + E\tau \cos \omega t_r]}{2E \cos \omega t_i [p(t_r) + E\tau \cos \omega t_r]} + \frac{2p(t_r) + E\tau(\cos \omega t_i + \cos \omega t_r)}{2\omega^2} \right) \right| \right]^{-3/2} \qquad (14)$$

where the instants $t_i$ and $t_r$ are those in the absence of the ionic background field. Estimations calculated using Eq. (14) will be presented and discussed along with the results of the numerical calculations in the next subsection.

Equation (14) can be considered as an *upper limit* for the transition density, since we have approximated the ionic background field as homogeneous. This ansatz is valid, if the mean distance between the ions, $R_{\mathrm{tr}}$, is much larger than the excursion length of the electronic wave packet, $R_{\mathrm{excursion}}$. Thus, the effect of the neighboring ion on the motion of the wave packet is underestimated by Eq. (13) and the real transition density is expected to be somewhat smaller than the value estimated above.

We may finally note that the transition density Eq. (14) has been obtained by taking into account the component of the ionic attraction along the polarization direction only. We have estimated the effect of the transverse component of the external field on the wave-packet motion, following the same steps as above, and found that the effect on the transverse component is essentially weaker than the effect on the longitudinal component; thus the former can be omitted.

### B. Numerical results

We present the results of the numerical simulations for HHG of Ar obtained from Eqs. (8) and (9). We have considered the interaction with a pulse having a typical wavelength of a Ti:sapphire laser, $\lambda = 800$ nm, and the duration was 50 fs in most of the calculations; otherwise it is stated explicitly.

In Fig. 2 we show a comparison of the harmonic spectra generated in the presence of an ionized background medium having a density of $10^{20}\ \mathrm{cm^{-3}}$ (panels on the right-hand side) with those obtained without medium (panels on the left-hand side) at two intensities of the laser field. In all cases a typical harmonic spectrum is seen with peaks at the odd multiples of the fundamental frequency. But, from the comparison we observe that in the presence of the medium the envelope of the spectrum is strongly modified. The intensities of the harmonic lines are reduced, which is due to the increase of incoherence in the harmonic response caused by the varia-

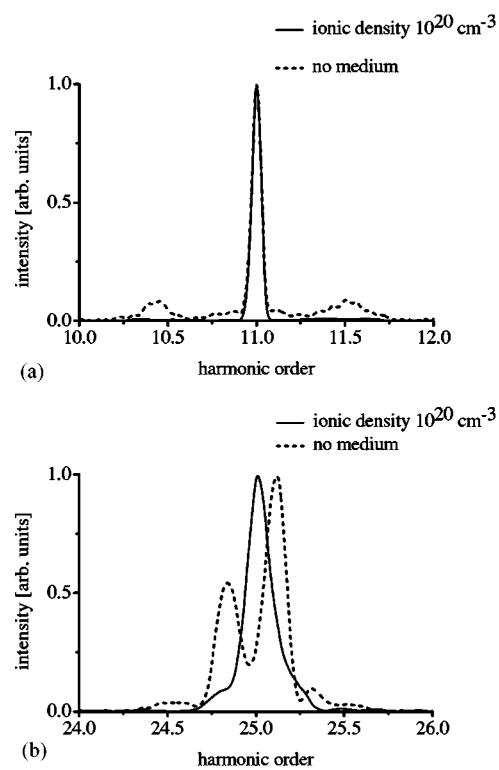


FIG. 3. Lines of the 11th (a) and the 25th (b) harmonics without medium (dotted line) and in an ionized medium (solid line). Same parameters as in Fig. 2, upper row. The spectra are renormalized to match at the maximum.

tion in the harmonic phase. Further, it is seen from the comparison that the harmonic lines appear to be much sharper in the presence of the ionic medium than for the single-atom response.

The latter feature can be analyzed in more detail from the results for the 11th [Fig. 3(a)] and the 25th [Fig. 3(b)] harmonics, generated at an intensity of $1.5\times10^{14}$ W/cm$^2$. The line from the lower part of the plateau consists in the absence of the ionic background (dotted line) of a sharp central peak and two sidebands. These sidebands disappear in the presence of the ionic medium (solid line). As has been explained at the outset, the sidebands arise due to the contribution of the longer quantum path of the electron wave packet in the continuum, while the central peak can be related to the shorter quantum path. Thus, the suppression of the sidebands indicates that the harmonic component generated due to the longer quantum path is more sensitive to the presence of the medium. For the harmonic line from the cutoff region [Fig. 3(b)] we see a double-line structure in the absence of the medium (dotted line) and again a sharp single line in the presence of the ionic background (solid line). The double-line structure appears due to the strong interference of the contributions of both the quantum paths. Its change into a single line in the presence of the medium is consistent with our interpretation that the long quantum path will be affected more strongly by the ionic background than the shorter one. For the present parameters the contribution of the short path to the high-harmonic spectrum appears to be effective alone.

Next, we have studied the harmonic spectra as a function of the medium density in order to see whether or not a transition from coherent harmonic generation to incoherent plasma emission can be identified. To this end, we have divided equally the plateau harmonics into three groups, which we denote as lower, middle, and cutoff groups (see Fig. 2, left column). Thus, at a given intensity there is the same number of harmonics in each group, but this number depends on the intensity of the laser. In addition, we have separated the contributions of the shorter and the longer quantum paths in the lower and the middle group. As mentioned above, this can be done 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 calculated and the results are shown in Fig. 4 as a function of the density of an ionic medium (a) and of a neutral atomic medium (b), respectively. For the sake of comparison the results are normalized such that the single-atom result (without medium) is set to 1 in each case.

In Fig. 4(a) 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 quasiclassical motion along the two paths is most pronounced for the lower plateau harmonics [9,16]. We may note that this difference in the decrease between contributions from the two quantum paths is a single-atom effect, and will influence the many-atom (medium) response too. It

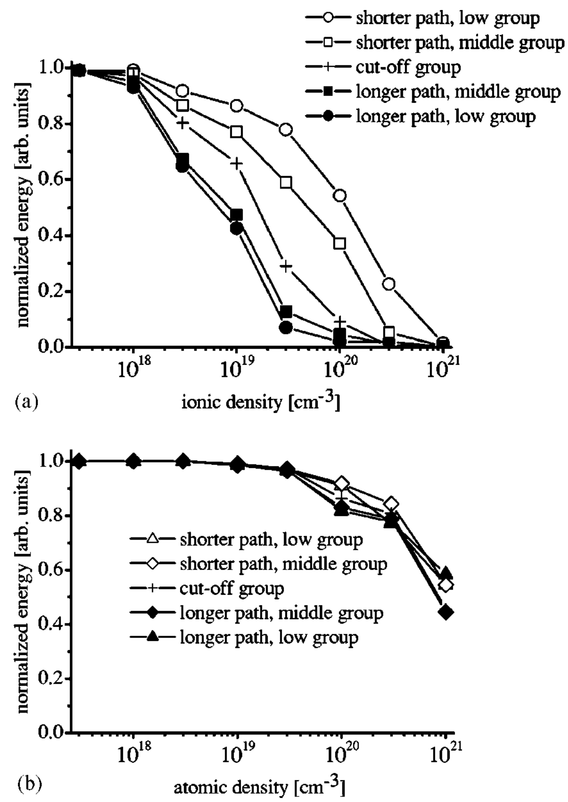


FIG. 4. Total emitted energy of different groups of harmonics generated at $2\times10^{14}$ W/cm$^2$ of (a) an ionized medium and (b) an atomic medium as a function of the density. Contributions arising from the shorter and the longer quantum paths are presented separately; groups of harmonics are selected as shown in Fig. 2, lower row. The energies are normalized to match with each other in the absence of the medium.

might be expected that the relatively slow decrease in the single-atom response might be compensated by the $n^2$ increase in the many-atom response ($n$ is the number of atoms). However, this macroscopic effect would affect the harmonic spectrum at the whole and not the contributions from the individual quantum paths separately. Furthermore, since the experimentally observed harmonic signal is limited by either absorption or phase-matching effects, the number of particles contributing to the observed coherent signal is usually nearly independent on the medium density. Thus, the decrease of the harmonic signal discussed here should be real.

As discussed above, 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. 4(a) that this density depends slightly on the harmonic group and ranges between about $10^{19}$ cm$^{-3}$ (contributions of the longer path) and $4\times10^{19}$–$10^{20}$ cm$^{-3}$ (contributions of the shorter path). Interestingly, the transition density is found to be nearly independent of the laser intensity, as it is exemplified for the results for the harmonic group in the middle of the plateau in Fig. 5.

In Fig. 6 we present the transition densities estimated analytically using Eq. (14) for the low and middle harmonic

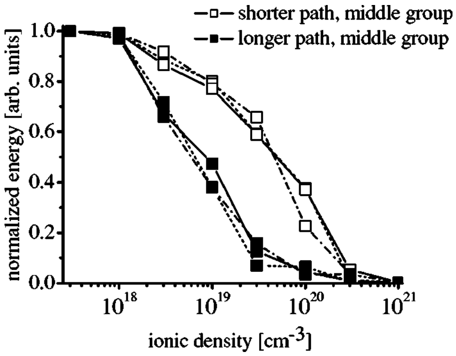


FIG. 5. Contributions to the total energy of the middle group of harmonics arising due to the different quantum paths as a function of the ionic density at different intensities: $2\times10^{14}$ (solid lines), $1.5\times10^{14}$ (dash-dotted lines), and $10^{14}$ (dashed lines) $\mathrm{W/cm^2}$. The energies are normalized to match with each other in the absence of the medium.

groups (more accurately, we present the transition density for radiation with energy $I_p+0.5U_p$ and $I_p+1.5U_p$ for the low and middle groups, respectively). The analytically estimated results for the transition density are, as expected, up to one order of magnitude higher than numerical ones. The deviation is smaller for the longer path (about $4\times10^{19}$ cm$^{-3}$ and $10^{19}$ cm$^{-3}$ for the middle group from the estimation and numerical calculations, respectively) than for the shorter one (about $8\times10^{20}$ cm$^{-3}$ and $6\times10^{19}$ cm$^{-3}$). However, the analytical approach predicts correctly the qualitative behavior of the transition density, namely, the ratio of the transition densities for different harmonic groups and the weak dependence of these densities on the fundamental intensity. Detailed analysis of Eq. (14) shows that the parameter that characterizes the effect of the ionic medium on the harmonic generation is rather the time of the free motion of the wave packet, $\tau$ (which is independent of the laser intensity), and not the excursion length of the packet (which depends on the laser intensity). The weak dependence of the transition density on the fundamental intensity for a given time $\tau$ (which is a characteristic value for each harmonic group) is clearly illustrated by the results in Fig. 6.

One may argue that in a typical experiment on HHG the medium is usually partially ionized by the generating pulse only and near the peak of the pulse the medium consists of ions as well as atoms. Therefore, we have calculated harmonic spectra for a neutral atomic background as well. The results are shown as a function of the density of the atoms in Fig. 4(b). The comparison of the results in Fig. 4(a) (ionized medium) and Fig. 4(b) (neutral medium) shows that the effect of the neighborhood on the harmonic spectrum is much stronger in the case of an ionic background than for the neutral medium. The transition density is found to be about an order of magnitude higher for the neutral medium than for the ionic background and, moreover, in the former case the effect of the medium appears to be the same for each harmonic group as well as for both the contributions of the short and the long quantum paths. The higher transition density for the neutrals is due to the short-range character of the atomic potential (7) in contrast to the long-ranged ionic Coulomb potential (6); thus a higher concentration of particles is required to induce a significant effect on the harmonic phase.

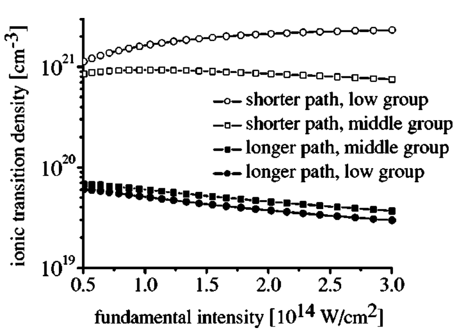


FIG. 6. The transition densities for an ionic background medium estimated analytically using Eq. (14) for the low and the middle harmonic groups vs the intensity of the fundamental pulse. The results for the shorter and the longer quantum paths are presented separately.

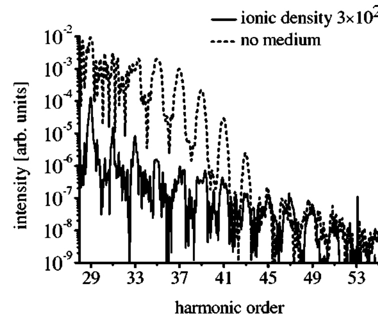


FIG. 7. Comparison of the spectra in the cutoff region at intensity $2\times10^{14}$ $\mathrm{W/cm^2}$ generated without medium (dotted line) and in a fully ionized medium of density $3\times10^{20}$ cm$^{-3}$ (solid line).

Our results for the decrease of the coherent harmonic response toward an incoherent emission, as the density of the medium is increased, are in agreement with the observations in a recent experiment [12] on HHG in water microdroplets. Due to the specific pump-probe method used in that experiment the density of the medium, in which the harmonics were generated, could be changed and controlled. A transition from harmonic generation to plasma emission was observed in this experiment, the transition density in the experiment is in a good order-of-magnitude agreement with the density at which we observe a suppression of harmonic generation in the calculations. 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.

It might be expected, that in a dense medium the harmonic spectrum is extended *beyond* the single-atom cutoff due to the possibility of a recombination of the electron with a neighboring ion. Such an extension of the spectrum has been predicted, e.g., for HHG in molecular ions [18–20]. We have therefore investigated the harmonic spectrum beyond the single-atom cutoff at a rather large density of $3\times10^{20}$ cm$^{-3}$ and a laser intensity of $2\times10^{14}$ $\mathrm{W/cm^2}$. At this medium density the plateau harmonics are strongly suppressed already [cf. Fig. 4(a)]. For this study we have reduced the pulse duration to 25 fs due to the large number of Monte Carlo attempts needed to achieve convergence of the results under such extreme parameters. From the comparison between the results in the dense medium (solid line) and without medium (dotted line), presented in Fig. 7, one sees that there is no increase of the harmonics beyond the cutoff. This is consistent with the fact that the coherence of the contributions due to recombination with a neighboring ion should be very low for randomly distributed ions in the medium position. Coherent harmonic radiation from a plasma was predicted theoretically [21], but its intensity for our conditions should be very low [22]. Our calculations show that the efficiency of harmonic generation beyond the cutoff is still lower that that of the plateau harmonics and is, if present at all, lower than the noise level in our calculations, at least for the range of parameters used here. We may note that in an earlier publication [13] a significant extension of the harmonic spectra in a partially ionized medium was predicted. We attribute this difference to the fact that in the earlier publication a one-dimensional model was used, which strongly restricts the range of positions of the ions in the simulations.

Finally, we have studied the dependence of the harmonic phase on the medium density. It varies for different harmonics and intensities, but remains always weak; namely, the change of phase is much smaller than $\pi$ when the density varies from zero to the transition regime. Thus one can conclude that the dependence of the harmonic phase on the medium density, in contrast to the dependence of the harmonic intensity on the density, should not lead to any additional phenomena in comparison with the low-density limit.

### C. Applications

High-harmonic generation has a number of important applications; we therefore discuss the impact of our findings on harmonic generation in a dense medium for spectroscopic applications and subfemtosecond pulse generation.

It was predicted theoretically [23,24] and shown experimentally [2,3] that a train of attosecond pulses can be obtained using the radiation of a group of harmonics. Usually two pulse trains are generated due to the harmonic contributions from the shorter and the longer quantum paths, respectively (see, e.g., [25]). Since we have found that the two contributions are influenced differently in the transition regime, we expect that the attosecond pulse train can be changed and partially controlled at certain densities of the medium. In order to substantiate our expectations we have calculated the intensity of a harmonic group as a function of time by

$$w(t) = \left| \int_{\Omega_{\mathrm{low}}}^{\Omega_{\mathrm{high}}} \overline{f(\Omega)} \exp(-i\Omega t) d\Omega \right|^2. \tag{15}$$

The results, calculated using $\Omega_{\mathrm{low}}=26\omega$ and $\Omega_{\mathrm{high}}=38\omega$ for a medium density of $10^{20}\ \mathrm{cm}^{-3}$ at fundamental intensity $2\times10^{14}\ \mathrm{W/cm}^2$, are presented in Fig. 8. In the absence of the medium one can recognize the two trains of attosecond pulses (dotted lines) 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 close to the transition one of the trains is strongly suppressed and, indeed, a single attosecond pulse train generated by the shorter path contributions arises. Thus, under certain conditions using medium densities higher than about $3\times10^{18}\ \mathrm{cm}^{-3}$ a shortening of attosecond pulses seems to be feasible. We may note that is has been found that phase matching also can favor the contribution of one of the two quantum paths in the experiments and lead to a suppression of one of the attosecond pulse trains. Inclusion of the macroscopic phase-matching effect is however beyond the scope of the present paper.

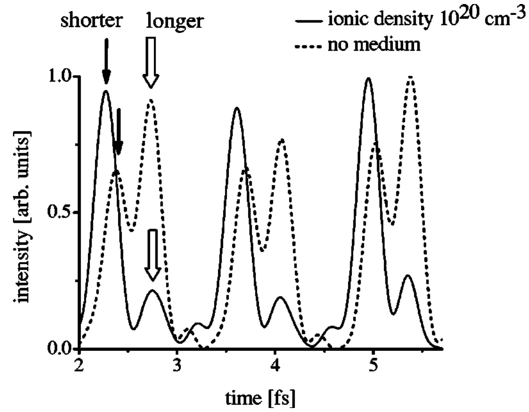


FIG. 8. Intensity of the radiation from the 27th to 37th harmonics vs time, generated without medium (dotted line) and in an ionized medium (solid line). Intensity of the fundamental pulse was $2\times10^{14}\ \mathrm{W/cm}^2$. The arrows mark parts of the attosecond pulse that are mainly generated by the shorter and the longer quantum paths, respectively. The curves are renormalized to match at the maximum.

In spectroscopy, 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 accuracy of such measurements is limited by the bandwidth of the harmonic line (see, e.g., [26–28]). As shown above (cf. Fig. 3), in an ionic medium having a density in the transition regime the sidebands of the harmonic lines in the plateau regime are strongly suppressed while the central peak still remains, which leads to an effective narrowing of the harmonic line. In order to minimize the Fourier-limited line-

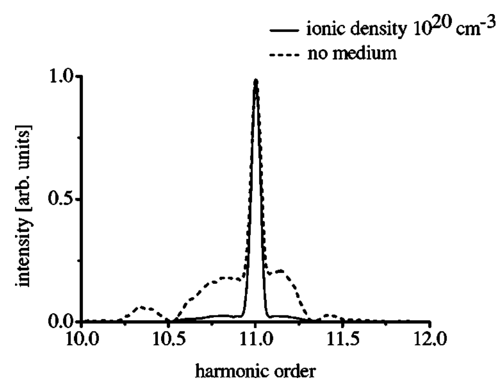


FIG. 9. The line of the 11th harmonic generated at $10^{14}\ \mathrm{W/cm}^2$ without medium (dotted line) and in an ionized medium (solid line). Spectra are matched at the maximum.

width in spectroscopic applications relatively long fundamental pulses and low intensities are used. It is interesting to note that under such modest intensities the sidebands are even more pronounced [cf. Figs. 3(a) and 9] and their suppression in the presence of the medium is even more important. Note, however, that we did not take into account a broadening of the harmonic line due to the plasma-induced blueshift of the fundamental. This broadening can be smaller or larger than the line narrowing discussed above, depending on the experimental parameters such as target material and laser pulse. It would be therefore of much interest to see if and which kind of change of the linewidth can be observed in experiment.

## IV. CONCLUSIONS

We have investigated high-harmonic generation from an argon atom in a dense medium. The results are obtained by solving the Schrödinger equation for an atom in the combined fields of the laser and of the ionic (or atomic) background medium numerically and calculating the Monte Carlo average for sets of randomly located ions or atoms. Significant changes in the harmonic response from the single-atom result (without medium), namely, a narrowing and a suppression of the harmonic lines, are found in a transition regime between $10^{19}$ and $10^{20}$ $cm^{-3}$. This is interpreted as due to the random variation of the harmonic phase induced by the external field of the neighboring particles during the time of free motion of the electron wave packet. In agreement with this interpretation, 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, and the transition density is almost independent of the laser intensity. According to our results the coherent response beyond the semiclassical cutoff is negligible due to the random distribution of the particles in the medium. Finally, it has been shown that at densities in the transition regime a significant narrowing of the harmonic lines and a shortening of the attosecond pulses in a train can be achieved in the single-atom response.

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