Parametric attosecond pulse amplification from high order harmonic generation in He$^+$ far from the ionization threshold

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Parametric amplification of attosecond coherent pulses around 100 eV at the single-atom level is demonstrated for the first time by using the 3D time-dependent Schrödinger equation in high-harmonic generation processes from excited states of He$^+$. We present the attosecond dynamics of the amplification process far from the ionization threshold and resolve the physics behind it. The amplification of a given photon energy requires the seed XUV pulses to be perfectly synchronized in time with the driving laser field for stimulated recombination to the He$^+$ ground state in agreement with the experimental measurements. The results from our numerical simulations are an important step for the understanding and realization of compact attosecond pulse intense XUV lasers.

High harmonic generation (HHG) yield enhancement is needed for the design of compact XUV and x-ray lasers fitting in hospitals and university laboratories and therefore for HHG coherent radiation to be useful in a large number of applications [1, 2]. In particular, the amplification of attosecond HHG pulses at high photon energies far from the ionization threshold in gases has been extensively investigated both for its fundamental interest and also for several straightforward applications, such as the use of parametric amplified HHG pulses as seed in full coherent plasma x-ray lasers [3, 12].

Attosecond XUV pulses of high photon energies have been successfully amplified in experiments using He gas and the fundamental physics behind the involved scattering mechanisms have been examined both experimentally and theoretically [3, 4, 5, 7, 9, 12]. Parametric processes involving stimulated recombination in HHG have been found as the dominant mechanism behind the observed XUV pulse amplification, although a description of the single-atom medium, while negative values mean recombination is dominantly produced by x-ray parametric amplification (XPA) as it was determined from the previous experimental and theoretical studies. We do this by examining the simultaneous excitation of He$^+$ ions with an intense IR pulse and a weak XUV pulse, by numerically solving the spin-free single-active electron (SAE) [13, 14] 3D time-dependent Schrödinger equation using the implementation described in [12, 15] with a hydrogen-like potential.

The main results of our simulations are shown in Fig. 1. We have considered a linearly polarized IR field resulting from a 20-cycle $\cos^2$ envelope vector potential ($\approx$20 fs duration) of 800 nm, with IR field peak intensities of $4.0 - 6.0 \times 10^{14}$ W/cm$^2$, as indicated in the subplots in Fig. 1. The IR field interacts with an He$^+$ ion together with a weak ($\approx 10^{12}$ W/cm$^2$) XUV single pulse of $\cos^2$ shape and of 1.5 fs total ($\approx 0.55$ fs intensity FWHM) duration, considering the harmonic frequencies H61, H71 and H81, as it is also shown in the figure. We note that we have considered a single attosecond XUV pulse in order to analyze the physics of the amplification process and have checked that the results that we present do not change substantially by using instead an attosecond pulse train. In Fig. 1 $t = 0$ is considered at the center of the sin–IR pulse, so that the delay between the XUV and the IR pulse is given by the central time position of the XUV pulse and hence it coincides to the values in the horizontal time-axis in Fig. 1.

To study the absorption of the XUV pulse in the medium, we compute the total probe absorption signal $S_a$ obtained in the frame of transient absorption as described in [16] (see [12] for further details). In all calculations atomic units (a.u.) are considered and the total probe absorption signal $S_a$ is given in units of area (barns). Positive values of $S_a$ mean absorption of the XUV pulse in the single-atom medium, while negative values mean amplification (gain), so that the absorption cross section...
$S_a$ multiplied by the number density of atoms results in the absorption/gain coefficient $[16]$. Our study is centered in the 90 – 130 eV XUV gain region which has been observed in recent HHG measurements in He. Importantly, this photon energy region is distant from both the ionization energy of He (24.6 eV) and He$^+$ (54.4 eV). Convergence of the numerical simulations has been verified in all cases. For the parameters configuration that we have considered, our results indicate that if the population density of highly excited states is significantly low compared with the ground state He$^+$ (1s) ion density, only absorption is observed at all delays. Thereupon, we have considered the He$^+$ ion initially in diverse excited states such as 2p, 2s, 3p, 4p and 5d.

Having a look at Fig. 1, we observe that several gain regions are produced in different delays for the central photon energies H61, H71 and H81. Those gain regions for such high photon energies had never been predicted before by using the solution of a first principles theory at the single-atom level, as we report here. The degree of amplification depends both on the central XUV photon energies being amplified and the peak intensity and duration of the IR pulse considered. Indeed, when the He$^+$ ion is initially in one of the 2p, 2s, 3p, 4p or 5d bound states, it first interacts with the leading part of the IR pulse, which tunnel ionizes the He$^+$ ion and single and multiphoton excitations to the lower and upper bound states also occur. Therefore, when the synchronized XUV seed pulses interact with the He$^+$ ion at a particular delay, they will be absorbed or amplified depending on the balance between the probability of absorption and the one for parametric stimulated recombination to the ground state – as it will be shown in Fig. 2, which is influenced by the configuration of the electron wave function at this
particular delay. Otherwise, the details of the regions where the amplification is produced in Fig. 1 are difficult to disentangle. We observe the highest gain for H61 of about 30 b at the higher IR peak intensity that we have considered (6 × 10^{14} \text{ W/cm}^2) [see Fig. 1(c)], which occurs for the He\(^+\) ion being initially in the 3p state and at the earlier delay of all cases (≈−9.5 fs). As a general trend, the gain decreases as the central XUV pulse photon energy is increased, and it is higher for the higher IR peak intensities, which allows us to predict a relatively small gain (0.05 b) with 6 × 10^{14} \text{ W/cm}^2 at photon energies as large as H81 (≈125.5 eV). In this sense, we have to stress that although the values of the calculated gain are somewhat small at the single atom level, the probability for the parametric amplification effects is to be exponentially enhanced by propagation in the medium due to avalanche effects as described in [8]. It is also worth noting that the peak intensities typically used in several experiments (≈10^{15} \text{ W/cm}^2) can be higher than the ones that we have considered, and that the IR pulse durations are typically also longer, as it will be shown below. Higher peak IR intensities with 20-cycle or longer duration pulses are however almost prohibited for numerical studies due to computing capabilities. Nevertheless, the numerical analysis that we report indicates how the medium can produce XUV gain in precise XUV–IR delay regions for geometries that can be accomplished in the experiments, also with higher IR peak intensities and longer pulses and with both He atoms or He\(^+\) ions initially in their ground state as amplifying media.

Figure 2 (a) shows a frequency-time analysis of the high-order harmonics generated by an IR pulse alone with peak intensity I = 5 × 10^{14} \text{ W/cm}^2, in the case that the system is initially in the 3p bound state. The intensity of the generated harmonics is shown in logarithmic scale and arbitrary units. (b) Zoom of the frequency-time analysis in (a) close to the harmonic H61. The intensity of the generated harmonics is here in linear scale and arbitrary units. The calculated spectrally integrated single-atom XUV absorption signal S\(_a\) for H61 (Fig. 1(b) – red-dashed line) is plotted on top of the spectra (white line). The vertical axis of this white line has been reversed here with respect to Fig. 1(b) for a better visual comparison.

![Figure 2](image.png)

**FIG. 2.** (color online) (a) Frequency-time analysis of the high-order harmonics intensities generated by an IR pulse alone with peak intensity of 5.0 × 10^{14} \text{ W/cm}^2, in the case that the system is initially in the 3p bound state. The intensity of the generated harmonics is shown in logarithmic scale and arbitrary units. (b) Zoom of the frequency-time analysis in (a) close to the harmonic H61. The intensity of the generated harmonics is here in linear scale and arbitrary units. The calculated spectrally integrated single-atom XUV absorption signal S\(_a\) for H61 (Fig. 1(b) – red-dashed line) is plotted on top of the spectra (white line). The vertical axis of this white line has been reversed here with respect to Fig. 1(b) for a better visual comparison.
FIG. 3. (color online) (a) Experimental setup of the two-jet arrangement. (b) CCD images of the harmonic spectra measured from the jets separately (seed and unseeded) and the two jets together (seeded), and (c) the same spectra plotted with the transmission curve of the used Zr thin-film filter.

Fig. 3(b) for Jet1 alone (seed), for Jet2 alone (unseeded) and the two jets together (seeded). The intensities of the spectra can be compared in Fig. 3(c). The spectrum of the two jets together is much more intense than the seed and unseeded spectra and cannot be obtained by a coherent superposition of them. Moreover, a bright central part of the beam appeared in the seeded case with less divergence compared to the beams of the jets alone, what is a further indication of the appearance of amplification and gain.

To reveal the gain dynamics in He gas, the delay between the laser pulse and the HHG seed (or probe) pulse can indeed be scanned by changing the distance between the two jets. The delay has two possible origins. One is the difference of the Gouy phases at the positions of the gas jets within a focused laser beam. In the measurements in Figs. 4(b)-(d) the scanning was made in 20 µm steps within a range of 2 mm. Because this range is much smaller than the ~50–60 mm Rayleigh length of the laser beam used, the contribution of the Gouy phase
represents a negligible delay (< 30 as). A much larger effect is given by the contribution of the free electrons to the refractive index during the propagation of the laser beam between the jets. The delay \( \tau \) given by the distance is \( \tau \approx (e^2d_0n_e[1-\exp(-d/d_0)]/(2\epsilon_0m_ec\omega^2) \), where \( n_e = n_{e1} + n_{e2} \) is the sum of the free electron densities of the contributing two gas jets, \( d \) is the distance between the jets, \( d_0 \) is the distance from the gas jet, the gas density decreases to 1/e, and \( \omega \) is the laser frequency, with the rest of parameters being the common SI constants. The measured spectrum-series of the delay scan can be seen in Fig. 3(a). The delay is scaled in Fig. 3 as in the calculations in Fig. 1 by using the value \( d_0 = 3.5 \text{ mm} \) and 26% free electron density at 0.5 bar gas pressure. Beyond the spectral shape, the beam profiles of the harmonic beams were also evaluated from the CCD images of the spectra similar to Fig. 3(b). The gray dashed line in Fig. 3(c) shows the beam waist for the most intense harmonic order that was measured (H61). It is clear that the XPA feature appears as an almost abruptly decrease of the beam waist at about −9 fs delay. A Gaussian shape has been fitted in Fig. 3(b) (dashed lines) to serve as a reference for a case without parametric gain. In Fig. 3(b), we observe how, additionally to the decrease of the beam waist shown in Fig. 3(c), the harmonic intensity increases substantially, and intensity peaks can be identified as the indication of the appearance of gain at specific XUV–IR delays. Dividing the measured pulse by the reference Gaussian shape, the observed gains for three harmonic orders are plotted in Fig. 3(d). The strongest peak at about −7 fs delay and a somewhat weaker one at about −8 fs delay can be clearly attributed to the 3p and 4p orbitals of the excited \( \text{He}^+ \) ions by comparing the results with the calculations in Fig. 1(b) and Fig. 2(c). The calculated and measured gains are plotted together in Fig. 3(c) for an easy comparison. As it can be expected, the two gain peaks are separated by half an IR cycle. The peaks at about −5 fs and −4 fs delays that appear in Fig. 3(d) are only observed for higher harmonic orders (H71) in Fig. 3(e). They might therefore correspond to the 2s initial state, as noted in Fig. 3(d). The remaining peak at −7 fs in the case of the higher harmonic order (H77) in Fig. 3(d) (gray dashed line) might also be due to propagation effects that can couple neighboring harmonic orders, which is not considered in the present simulations.

In conclusion we have shown for the first time that amplification of high photon energy XUV pulses far from the ionization threshold (around 100 eV) can be described at the single-atom level by the 3D time-dependent Schrödinger equation in \( \text{He}^+ \), which allows us to doubtless reveal both in the time and the frequency domains how the physics behind these amplification processes have a parametric character. In complete accordance with the experiments, we conclude that the XUV pulses need to be perfectly synchronized in time with the driving laser field to produce stimulated recombination. We have demonstrated that transitions from the continuum to the ground state of \( \text{He}^+ \) ions are essential for efficient high photon energy amplification. Our study reveals how the gain and photon energy for XPA is entirely influenced by the particular experimental geometry, and it also indicates the basic physics for future experiments to achieve the desired attosecond XUV gain needed for applications, such as intense coherent seeds for plasma x-ray lasers.

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