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Enhancement of High-Order Harmonic Emission Using Attosecond Pulse Trains

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Abstract—We theoretically predict and experimentally confirm that enhancement of high-order harmonic generation is possible by combining an attosecond pulse train with an IR driving laser. This combination replaces the disadvantages of tunnel ionization with single-photon ionization and permits the manipulation of the time-frequency properties of high-order harmonic generation already at the single-atom level.

INTRODUCTION

High-order harmonic generation (HHG) [1, 2] provides a source of temporally and spatially coherent radiation covering a wavelength range from the fundamental wavelength of the driving laser, typically in the near-infrared range, to the water window [3, 4]. The major drawback of this source is its inefficiency and the lack of sufficient control over its properties, e.g., over the spectral resolution or spectral position at a certain wavelength range. Nevertheless, this source is very interesting for many different reasons, a commercial one being that a laserlike source emitting at 90 eV would be a great tabletop tool for the next generation of lithography. Record photon yields at 90 eV are around 10^8 photons per laser shot [5] (10^{17} photons for a TW-class laser), which are about two orders of magnitude below the requirements for soft x-ray microscopy.

In order to understand the limitations of this method and the requirements for up-scaling in intensity, many investigations have been conducted [6]; nevertheless, many parameters simply elude the experimentalist. Recent theoretical studies [7–9] have shown that enhancement and even control over this process is nevertheless feasible on the single-atom level and macroscopically, leading to an enhancement by at least two orders of magnitude. This is possible through a combination of an attosecond pulse train (APT) with an IR field: the attosecond pulse train allows one to set the time of ionization and the initial kinetic energy and emission direction of the electron wavepacket, thereby removing the disadvantages of strong-field tunneling. The linear dependence of ionization on the strength of the APT also leads to a large volume effect, since even the radial wings of the IR pulse contribute significantly to HHG, whereas in the tunneling case the intensity would be below threshold. The experiment presented in this paper confirms these predictions: it shows an enhancement by a factor of five even without proper timing control.

THEORY

We consider the harmonics generated by a macroscopic number of helium atoms exposed to a combination of a focused APT and an IR laser pulse. Our simulation [9] describes the macroscopic response (propagation) by solving the three-dimensional Maxwell wave equation (MWE) in the slowly evolving wave approximation [6, 10] and the microscopic response (single atom) through a solution of the time-dependent Schrödinger equation (TDSE) in the strong-field approximation [11].

The driving field, at the entrance of the medium, consists of the sum of an IR pulse (27-fs FWHM at a center wavelength of 810 nm) with a peak intensity of 5×10^{14} W/cm^2 and an APT (fourth power of the IR field envelope with total a FWHM of 14 fs) with a peak intensity of 10^{13} W/cm^2. The APT is synthesized from odd harmonics 11 to 19 of the IR field. Both fields are described as Gaussian beams with maximal volume overlap such that the APT focus is kept at the center of the medium and both beam waists are similar over at least 1 mm of the medium. Figure 1 shows results calculated for a 1-mm-long medium of helium atoms with a density of 2×10^{18} cm^{-3}.

The lower curve in Fig. 1 shows the medium response when only the IR field (5×10^{14} W/cm^2) is present. Adding an APT with an intensity of 10^{13} W/cm^2 results in a significant increase in the harmonic signal (by several orders of magnitude) in the plateau region, with decreasing enhancement for increasing harmonic order, for both of the time delays between the APT and the IR field, which are shown by the upper two curves. With the APT present, all harmonics are nicely resolved with increasing spectral width for increasing order; this is a strong indication that, in the much simplified picture of the simple man's model [12, 13], only the short quantum path [14, 15] is predominantly selected. A detailed investigation [9]

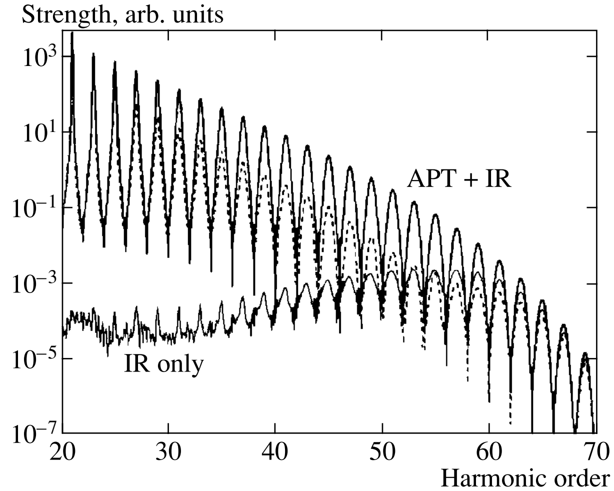

Fig. 1. Harmonic spectra generated in helium: the lower curve is calculated with only the IR field for an intensity of 5×10^{14} W/cm^2. The upper two curves show the medium response, with an additional APT (intensity of 10^{13} W/cm^2) for two different time delays of –0.095 IR cycles (solid line) and +0.185 IR cycles (dashed line).

shows that the single atom enhancement is much larger for low IR intensities than for high intensities. This means that the atoms in the wings of the IR beam profile contribute nearly as much as those in the center of the beam, leading to a large volume effect.

EXPERIMENT

The experimental setup is shown in Fig. 2: a Ti:sapphire-based laser system delivers 27-fs pulses with 0.7-mJ energy at 1 kHz and at a center wavelength of 785 nm. The collimated beam is focused with a spherical silver mirror into a 150-mm-long capillary with an inner diameter of 800 microns. The capillary is filled with xenon at pressures varying from 0 to 25 mbar and differentially pumped towards the exit. The capillary serves two purposes: it improves phase matching [16] and is expected to serve as extended spatial filter, thereby selecting predominantly the short quantum path—a prerequisite to yielding regularly spaced pulses within the APT [17, 18].

The IR intensity was determined to be 9.8×10^{13} W/cm^2, which is sufficient for generating harmonics to the 19th order in xenon, as required by the simulation. The resulting APT copropagates with the remaining IR field to another silver mirror, with which both are focused into the helium target. The helium target is a combination of a jet pulsing at 1 kHz, synchronized with the laser, and a 2-mm-long tube target to increase the interaction length [19]. Backing pressure for helium varied between 1400 mbar and 3000 mbar, with the ambient pressure being always below 10^{-2} mbar in the chamber. Without xenon present, the IR intensity was adjusted as to yield a cutoff in helium around the 69th harmonic. From the cutoff, we estimate the IR intensity to be 4.6×10^{14} W/cm^2. The generated harmonics propagate into a XUV spectrometer (McPherson), which can be read out at the full repetition rate of the laser.

The first results are shown in Fig. 3. First, we evacuated the capillary, and, with no xenon present, no APT is generated. The IR field is, however, strong enough to generate very high-order harmonics up to the 77th harmonic in helium, as shown in the left graph. Next, we

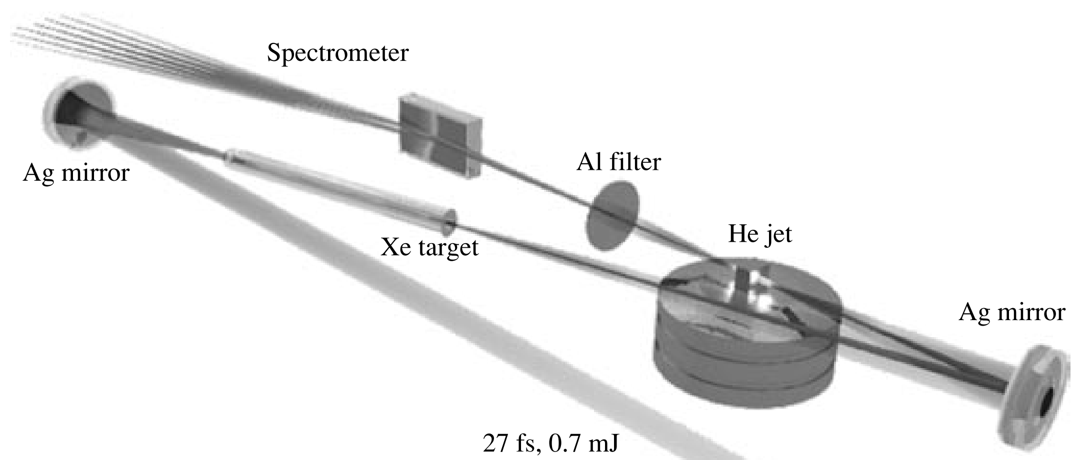

Fig. 2. Setup of the experiment: 27-fs, 0.7-mJ pulses at a center wavelength of 785 nm and at a repetition rate of 1 kHz are focused by a silver mirror into a 15-cm long and 800-micron diameter capillary filled with xenon. The capillary is differentially pumped towards the exit port. The low-order harmonics propagate together with the leftover IR pulse to another silver mirror, which focuses the radiation onto a pulsed helium target. The emerging high-order harmonics encounter the entrance slit of a commercial XUV spectrometer after the filtering out of the remaining IR with an aluminum foil. The spectra are recorded with a CCD with a fiber-coupled multichannel plate.

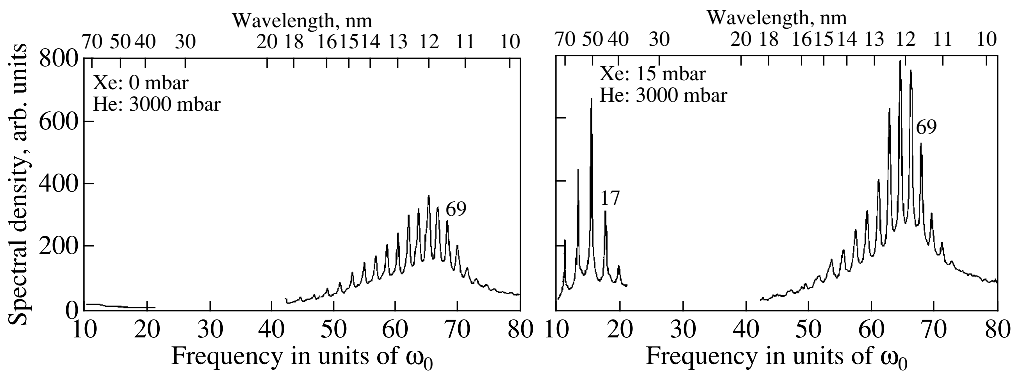

Fig. 3. Measured harmonic spectra for identical pressures of helium for two distinct pressures in xenon. Left: without xenon, no APT is generated, and shown are the very high-order harmonics generated in helium by the strong IR field. Right: with xenon at 15 mbar, the presence of the APT clearly influences the harmonics at the very highest orders.

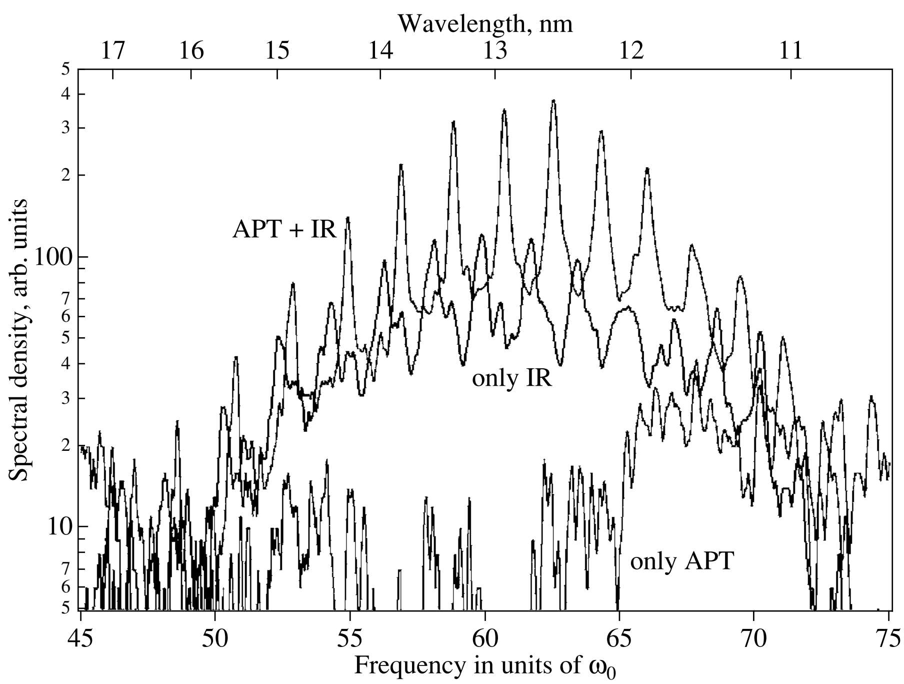

Fig. 4. Measured highest order harmonics for three cases: in "only APT," only xenon (no helium) is present and the generated harmonics (11 to 19), synthesizing the APT are of too low order to be visible in the graph. Furthermore, due to the low saturation intensity for xenon atoms, it is impossible to generate such high orders in xenon atoms. In "only IR," no xenon and, consequently, no APT is present. The IR field is, however, strong enough to generate highest order harmonics in helium. In "APTs + IR," a clear enhancement in amplitude is observed.

filled the capillary with xenon at a pressure of 15 mbar. The graph on the right clearly shows that the desired low-order harmonics (11 to 19) are generated, resulting in the desired APT. As predicted by theory, the amplitude of the highest order harmonics increases significantly, as shown on the right.

Figure 4 confirms the results shown in Fig. 3: shown are the highest order harmonics, between orders 45 and 75, generated only by the APT, the IR field alone, and by a combination of IR and APT. Since the APT is synthesized from the low-order harmonics 11 to 19 from xenon, the lowest trace (labeled "only APT") does not exhibit any harmonic spectrum in the shown wavelength range. Furthermore, due to the saturation intensities of xenon, such high orders would be impossible to generate from atoms. Without xenon, no APT is present, but, as described above, the IR field is strong enough to generate highest order harmonics in helium alone, as shown by the curve labeled "only IR." Combining the IR field and the APT results in the uppermost curve, labeled "APT + IR," which exhibits an enhancement in amplitude by a factor of five over the "only IR" case.

Two more features are clearly present in Fig. 4: with an APT and an IR field present, the harmonics are blueshifted, and their spectral widths are smaller compared to the "only IR" case. The blueshift can be explained by the fact that the IR field propagates through an ionized medium in the capillary, which causes a significant blueshift [20]. We estimated the blueshift from ADK theory [21], and it is in qualitative agreement with the observed values. Since the IR field, which is the determining factor for the photon energy gain of the high-order harmonics, is blueshifted, the resulting harmonics are blueshifted as well. The fact that, with the APT present, the harmonics are spectrally better resolved in the plateau region, i.e., narrower, is predicted by theory and attributed to the fact that only the short quantum path contributes to harmonic generation.

CONCLUSIONS

Enhancement of high-order harmonic generation was observed by combining an APT with a driving IR field, the first experimental results qualitatively confirm our earlier theoretical predictions [8, 9]. This not only shows that APTs are natural tools for influencing harmonic generation on a single-atom level, an effect that not only persists but even smoothes out the macroscopic medium response, thereby positively contributing to an increase in harmonic yield. Decoupling the ionization from the recombination steps allows one to tailor the spectral and spatial properties as well as the enhancement. We are convinced that large improvements over the demonstrated results are possible by choosing an improved interaction geometry, optimum time delay, and interaction volumes that should increase photon yields such that they might reach the levels necessary even for soft x-ray microscopy.

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