Tailoring of XUV supercontinua through coherent control of high-order harmonic generation

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Abstract

We present observations of the emission of XUV supercontinua in the 20-37 eV region by high harmonic generation (HHG) with 4-7 fs pulses focused onto a Kr gas jet. The underlying mechanism relies on coherent control of the relative delays and phases between individually generated attosecond pulses, achievable by adjusting the chirp of the driving pulses and the interaction geometry. Under adequate chirp and phase matching conditions the resulting interference will yield a supercontinuum XUV spectrum. This technique opens the route for modifying the phase of individual attosecond pulses and for the coherent synthesis of XUV supercontinua without the need of an isolated attosecond burst. ©2015 Optical Society of America

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Since the advent of high-order harmonic generation (HHG) and subsequently of attosecond science, the generation of supercontinua in the XUV range has been of high interest, mainly since this is a necessary condition for the emission of a single attosecond light pulse [1]. It is also an attractive coherent source for spectroscopic applications and material characterization.

To date, most of the strategies to obtain XUV supercontinua rely on isolating an attosecond pulse at the microscopic level (i.e. single atom). This implies the occurrence of a single ionization-flight-recombination event, which is only possible if the driving field pulse is as short as the period of the central frequency or, in the case of few-cycle pulses, if HHG is confined by gating techniques. Different strategies have been proposed, based on carrier-envelope phase stabilized few-cycle driving fields, namely spectral selection of the cutoff [1] or polarization gating [2], among others. The drawback of such techniques is the decrease of the generation efficiency, making it difficult to apply them to low-energy driving fields, such as the new high repetition µJ-level OPCPA sources [3]. Macroscopic phenomena can be also relevant to isolate attosecond pulses, and different techniques that make use of phase-matching conditions have been proposed, such as tilted pulse beams [4] or time-gated phase-matching [5].

However, all mentioned techniques generate XUV supercontinua through the emission of an isolated attosecond pulse. In contrast, in this work we explore the generation of XUV supercontinua driven by multi-cycle laser pulses, by altering the relative phase of a train of attosecond bursts, without going through the complexities of generating an isolated pulse. We present experimental results, corroborated with state-of-the-art theory, that demonstrate the emission of XUV supercontinua ranging from 20 eV to 37 eV as a result of the combination of microscopic and macroscopic physics. In the first part of this letter we present the experimental setup and results. In the second part, theoretical simulations are discussed and compared to experiments, inferring the nature of the obtained XUV supercontinua.

In the experiments we use a 1-kHz Ti:Sapphire CPA amplifier (Femtolasers FemtoPower CompactPRO CEP) delivering pulses with a Fourier-transform limit (FL) duration of 25 fs (FWHM) and energy up to 0.9 mJ. The output pulses are spectrally broadened in a hollow-core fiber (HCF) with an inner diameter of 250 µm and 1 m length. The HCF is filled with argon at 1 bar pressure. The carrier-envelope phase (CEP) is stabilized in the oscillator, and well-preserved during the amplification and post-compression processes.
The resulting broadband pulses are then sent through a chirped mirror and glass wedge compressor that forms part of a dispersion-scan (d-scan) pulse measurement and compression system. By compensating the spectral phase with 10 bounces off double-angle chirped mirrors (Ultrafast Innovations GmbH; nominal GDD: $-40 \text{ fs}^2$ per pair at 800 nm, minimum reflectance: 99%) and addition of normal dispersive material (BK7 wedge pair), few-cycle pulses are obtained. Using this setup we can routinely achieve pulse durations down to sub-4 fs, with an estimated intensity on target of $2 \times 10^{14} \text{ W/cm}^2$. Depending on the gas pressure, pulse duration can be tuned in a certain range. In our experiments we work with pulse durations from 4 to 7 fs (FWHM), determined with d-scan measurements.

The laser pulses are then focused by a spherical silver mirror ($f = 500 \text{ mm}$) into a pulsed Kr gas jet. The spherical mirror is placed on a translation stage, so the position of the focus can be controlled. The pulse enters the vacuum chamber through a 0.5 mm thick fused-silica window, situated close to the focusing mirror to avoid possible nonlinear effects. HHG is obtained in krypton (5 bar of backing pressure) using a nozzle of 500 $\mu$m diameter. The pressure reached inside the vacuum chamber where the high-order harmonics are generated is around $5 \times 10^{-3} \text{ mbar}$. A 150-nm thick aluminum foil is used to filter the IR radiation and the lower harmonics, while the higher orders can propagate through it. The HHG spectra are characterized with a grazing-incidence Rowland circle XUV spectrometer (Model 248/310G, McPherson Inc.), of 1-m radius and 133-grooves/mm spherical diffraction grating.

The gas jet is placed 4 mm and 2 mm before the focus position for 4 fs and 7 fs pulses respectively. We perform a dispersion scan on the driving pulses by finely translating one of the BK7 wedges. Therefore, HHG is studied as a function of the spectral phase of the incident beam. Fig. 1 reveals how crucial the effect of the IR spectral phase on the HHG is. As depicted, the narrower structures appear when the generating beam is positively chirped, while at negative chirping the spectra broaden, until eventually becoming supercontinua covering over 12 harmonic orders in a relatively low energy region (20-37 eV), including both HHG plateau and cutoff. The observed fringes denote the CEP dependence of the HHG (i.e. for a short range of BK7 insertion, the dispersion-scan becomes a CEP scan). The gas jet is placed before the focus so the phase-matching conditions allow for the observation of CEP-dependent HHG.

It is already known that HHG shows a spectral broadening/narrowing when negative/positive IR pulses are employed. However, supercontinua described in past works...
Fig. 1. Measured HHG dependence on BK7 insertion for 4 fs pulses (a) and 7 fs pulses (c). When
the insertion is finely varied, CEP dependence can be seen in the HHG spectra in the form of
fringes. Below each scan, in (b) and (d), XUV spectra are shown for the negative chirp BK7
insertion shown as a white dashed line in the corresponding chirp-scan.

were achieved for very high harmonic orders (in Ne, > 70 eV). Calegari et al. [10] reported
supercontinua in Xe using 5 fs pulses focused into a gas cell in a high intensity regime
\(2.5 \times 10^{15} \text{ W/cm}^2\), when selecting short quantum paths. According to the authors, this
effect is introduced by the plasma-induced chirp of the driving field as one main factor,
yielding a single attosecond pulse in the time domain. This case contrasts with ours in the
experimental conditions and, as we will see, in the origin of the supercontinuum.

We compute harmonic propagation using a method based on the electromagnetic field
propagator [11]. The dipole acceleration of each elementary source is computed using the
SFA+ method [12] (an extensi´on of SFA). Due to the lack of contrasted Coulomb potentials of
Fig. 2. Simulated HHG from the single atom response (a) and including macroscopic propagation effects (b) for different chirps of the driving IR pulse. Chirp is scanned by the insertion of BK7, in a similar way as in the experiment. Zero insertion means a FL generating pulse (4.3 fs in this case).

Krypton, we perform our HHG calculations in argon (which has similar ionization potential), using the Coulomb potential given in [13]. Propagation effects of the fundamental field, including plasma and neutral dispersion as well as time-dependent group velocity walk-off, are all taken into account. The absorption of the harmonics in the gas is modeled using Beer’s law. Agreement between the model and theory when phase matching is a relevant factor was tested in Ref. [5, 14].

The source of the behavior shown in Fig. 1 can be found on both the microscopic and macroscopic levels. In Fig. 2(a) we represent the simulated microscopic single-atom spectra driven in argon versus BK7 insertion, where the driving laser pulse is modeled by a \( \cos^2 \) envelope function of 4.3 fs FWHM, with \( 2.5 \times 10^{14} \) W/cm\(^2\) peak intensity at 720 nm. By changing the BK7 insertion, a standard harmonic spectrum structure is visible for positive and slightly negative chirp cases. However, for sufficiently negatively chirped input pulses, the HHG spectral structure changes, and spectral contributions appear out of the odd harmonic positions.

Propagation effects alter further these spectral structures, as shown in Fig. 2(b), obtained
for similar experimental focusing conditions. We use a Gaussian beam, whose waist at the entrance plane of the 40 cm focal length lens is 2.5 mm, focused into a gas jet placed 2 mm before the focus position. The gas jet is modeled by a Gaussian distribution along the gas propagation direction with 1 mm FWHM and peak density of 10 mbar. We show the harmonics as detected on-axis. Cutoff harmonics are lost during propagation and also odd harmonics show less chirp-dependent modulation than the single atom response in the positive chirp region. In the case of negative chirp, variations in the odd harmonic structure are still observed, but smoother than in the single atom case. The modulated spectrum obtained in the single-atom response is filled when propagation is taken into account. For a further insight on the nature of the supercontinua, we have analyzed the numerical simulations. For the sake of clarity, we compare negatively ($-100 \mu m$, Fig. 3) and positively (94 \mu m, Fig. 4) chirped IR driving pulses. In agreement with experiments, the negative chirp case yields a continuous spectrum [Fig. 3(a)] and spectra driven by unchirped or positively chirped IR fields show the usual HHG comb-like structure composed of odd harmonics [Fig. 4(a)].

Fig. 3. (a) Theoretical XUV spectra when 100 \mu m of BK7 have been removed from the FL condition (4.3 fs). Thin line denotes single atom response while thick line shows collective spectrum. Temporal profile is shown as inset for both cases, denoting at different colors the main individual attosecond pulses. (b) Corresponding spectral phase differences between first and second attosecond pulses (thick violet line for propagated, thin dashed violet line for single atom), and second and third pulses (green lines).
Fig. 4. (a) XUV spectra simulated theoretically for positively chirped pulses (94 µm of BK7 added from the FL condition (4.3 fs). Thin line denotes single atom response while thick line shows collective spectrum. Time evolution is shown as inset for both cases. (b) Corresponding spectral phase differences between attosecond pulses (violet lines for first and second pulses, green lines for second and third pulses).

insets show the temporal structure for both the single-atom and the propagated cases. The role of negative chirp in inducing changes over the harmonic structure at the single atom response level has been analyzed in HHG from solid targets \[15\], observing a non-constant time separation between the attosecond pulses. This may lead to a Moiré pattern in the spectral domain, merging some harmonics and changing their wavelength but, in general, maintaining a peaked structure. In Ref. \[15\], negative chirp in the IR driving field was identified as a major factor, since it enhances time delay emission differences. In our case, this phenomenon is observed (see insets in Figs. 3 and 4), but notably the supercontinuum is also associated to a particular phase difference among the individual attosecond pulses [Figs. 3(b) and 4(b)]. The XUV spectra corresponding to the different chirp cases have been analyzed as the result of spectral interference of the single bursts, as suggested in Ref. \[3\].

For a negatively chirped driving field (Fig. 3) the spectral phase difference between consecutive attosecond pulses within the pulse train yields a supercontinuum. Note that the supercontinuum structure appears when the phase difference between pulses 1 and 2 differs from the phase difference between pulses 2 and 3. In other words, at wavelengths were
dephasing of a pair of pulses is $\pi + 2m\pi$ ($m$ being natural), a minimum is found in the spectrum due to destructive interference, and maxima arise from constructive interference when the dephasing is $2m\pi$. When the dephasings among the pulse pairs are similar (as it happens when using a positively chirped driving field, see Fig. 4), a sharp odd harmonic structure arises. Contrarily, when dephasings do not agree and even have a difference of $\pi$ rad [Fig. 3(b)], a more continuous structure emerges from interference [Fig. 3(a)]. Please note that the dephasing is established at the single atom level and is conserved when propagated. Propagation therefore completes the spectrum shape by filling the structure.

To conclude, spectral supercontinua have been observed in the medium energy range of high harmonic spectra, for particular focusing and IR pulse conditions. Our simulations reveal that the origin of the supercontinua can be understood not as the emission of an isolated attosecond pulse, but as the interference of several pulses in a train with different time delays and relative phases. The time delay among pulses and the particular phase differences yielding the supercontinuum structure can be obtained through adjustment of the focus position, IR pulse chirp and CEP, allowing a fine control of the resulting spectral interference pattern. Effects due to microscopic and macroscopic contributions are described and compared. From a practical point of view, this method enables obtaining supercontinua without the need of pulse gating techniques, high intensities or single-cycle driving IR pulses.

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