Observation of Attosecond Light Localization in Higher Order Harmonic Generation

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The superposition of selected higher order harmonics and continuous background, produced in Ar gas through two time delayed 60 fs pulses at 800 nm, is measured as a function of their delay time. An isolated less than 100 as sharp feature, indicative for pulse production in the attosecond regime, followed by a subfemtosecond beating, indicative for the production of trains of sub-fs XUV pulses, is clearly observable in the resulting temporal trace. The trace Fourier transforms to a frequency spectrum composed of harmonics from the selected ensemble and a continuous background that correlates with the observed ultrashort feature. Numerical simulation spectra encompass similar temporal localization.

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It is well known that short duration pulses can be generated through the coherent superposition of phase related electromagnetic waves at regularly spaced wavelengths. The beating pattern that results from such a superposition exhibits a temporal localization, the extent of which is inversely proportional to the frequency content of the contributing fields. Higher order harmonics generated from intense laser pulses has been proposed a few years ago as a promising candidate for the formation of ultrashort pulses breaking the fs barrier [1–3]. Several alternative ways of attosecond (1 as $= 1 \times 10^{-18}$ s) pulse generation using harmonics have been proposed [2,4–13]. The spectrum of a short-pulse, strong-field harmonic generation consists of a rapid decreasing in intensity low order harmonics followed up by a region of effectively constant intensity peaks known as the "plateau" and a sharp decreasing in the intensity region known as the "cutoff." The main features of this behavior have been successfully explained through both classical and quantum-mechanical treatments [5,14–16]. The central proposition of a short pulse generation from higher order harmonics is that the superposition of comparable intensity and phase related harmonics may result in trains of ultrashort light pulses of attosecond duration in a manner similar to the mode-locking techniques. A large number of early investigations followed up these first ideas addressing several issues of the problem, such as the phase relation between the harmonics [6,17], attaining phase locking through phase matching [18,19], the chirp of the harmonics [20], the creation and separation of attosecond pulses, and time resolved attosecond spectroscopy.

The present work reports the observation of a subfemtosecond train and an even shorter isolated structure within the "higher order" autocorrelation spectrum recorded, detecting the extreme ultraviolet (XUV) light of a selected frequency region comprising a superposition of higher order harmonics and continuum radiation, generated by two time delayed laser pulses of short duration. Higher order refers to the order of the contributing harmonics and not of the perturbation, since in the region of interest the harmonic generation is a nonperturbative process. The observed train of temporal localizations to a few hundreds of attoseconds and the even narrower, less than 100 as, isolated structure at the leading edge of the measured trace are indicative of the temporal profile of the XUV radiation produced. It is, to our knowledge, the first observation of temporal localization in the attosecond regime produced by electromagnetic radiation.

The titanium sapphire laser employed outputs 1 mJ pulses at ~800 nm and a 1 kHz repetition rate, with a measured pulse duration of ~60 fs. In the experiment 200 μJ has been used in total. The laser beam was passed through an autocorrelator creating two collinear time delayed pulses of about 100 μJ, with the second pulse being weaker by 5%. The two pulses were focused via a 35 cm focal length lens at the 50 μm diameter exit pinhole of an Ar gas laminar flow cell, kept at constant pressure. The resulting intensity was measured to 1.7×10^{14} W/cm^2 for the first pulse. The density of Ar in the cell was of the order of 10^{17} atoms/cm^3. Under such experimental conditions ionization of the Ar ground state is less than 10% and the produced electron density less than 10^{16} electrons/cm^3. The corresponding plasma pulsation is of the order of 10^{13} rad/s which is 3 orders of magnitude less than the 29th harmonic frequency 7×10^{16} rad/s. Thus abrupt cutoff of the incident radiation that may introduce additional Fourier components can be excluded.

The linearity of the delay introduced by the Michelson interferometer has been interferometrically confirmed by recording the single photon first order autocorrelation of the fundamental with a linear Si photodiode. The total trace shown in Fig. 1(a) depicts an interference pattern that occurs when the two coherent pulses overlap in time. An expanded area of 8 fs that allows monitoring of irregularities at the time scale of the observed beating is shown in Fig. 1(b). Only a random amplitude fluctuation due to the instability of the laser beam is observed in addition to the modulation at the laser frequency. Slight fluctuations in the phase are also present. The Fourier transform of this interference pattern as is illustrated in Fig. 1(c) confirms the above. The inserted portion shows the spectrum

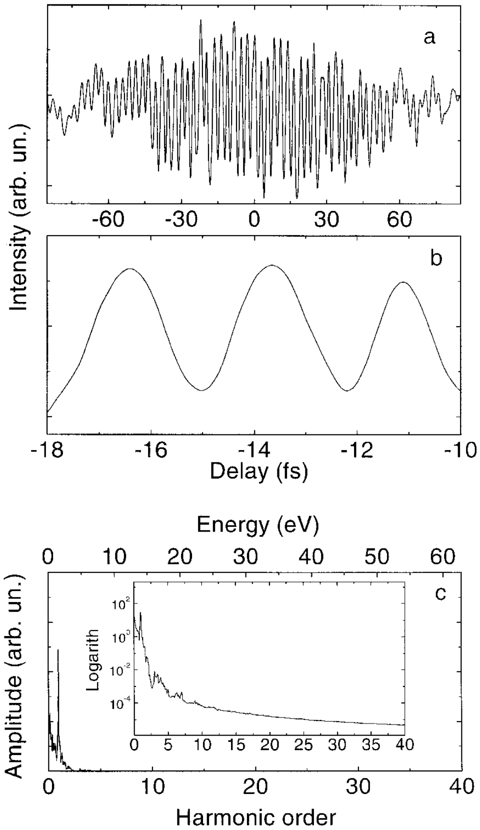

FIG. 1. First order interferometric autocorrelation trace of the laser field. The total trace (a); an expanded region (b); the Fourier transform of the total trace (c).

in logarithmic scale for better visibility of small contributions. One single dominant frequency is observable, the frequency of the laser radiation. Its observed amplitude is more than 2 orders of magnitude larger compared to all other contributions in the spectrum. In fact, the amplitude ratio of the fundamental to other higher frequencies is even larger ($\sim 10^5$), due to the strong suppression of the fundamental frequency during acquisition as described in the next paragraph. What is important here is the absence of any additional frequencies in the region of 10–100 PHz (1 PHz $= 10^{15}$ Hz) in which the higher harmonic structure is observed.

The harmonics generated at the pinhole entered collinearly with the fundamental in a vacuum chamber attached windowless to the quasistatic cell, passed through a 150-nm-thick Al-Si(1%) filter, and were detected with a microchannel plate (MCP) detector. The MCP (and the Si diode) signal was monitored through a lock-in amplifier locked at the laser repetition rate and recorded by a digital oscilloscope. Acquisition occurs in such a mode that the recorded trace is the derivative of the real trace and thus it reflects the field amplitude rather than the intensity. Moreover low frequencies are suppressed and "seen" as part of the dc component. Oscillations at the fundamental frequency are suppressed by a factor of 30, at the third harmonic by a factor of 3, and at higher harmonics by only 30%. In this way the strong contribution of the optical interference, occurring when the two laser pulses overlap [21], can be practically eliminated from the recorded data. The filter is used in order to cut the intense lower order harmonics and the fundamental, selecting harmonics in the energy region up to the cutoff value $E_{\text{cutoff}} = \text{IP} + 3.1U_P$, with IP being the ionization potential of Ar and U_P the ponderomotive shift of 10.1 eV at the laser intensity 1.7×10^{14} W/cm^2. The selection of harmonics through filters has been previously suggested [5,8,11,12]. In the region of the cutoff the harmonics produced by a single atom have locked phases [6]. Filtering is complemented by the photon energy dependent efficiency of the detector. Its response is constant within 15% in the wavelength region 80–25 nm and drops off towards the fundamental frequency.

The total intensity transmitted through the filter and detected by the MCP detector assembly was measured in a continuous mode as a function of the time delay between the two subsequent laser pulses. A recorded autocorrelation trace is shown in Fig. 2(a). Figures 2(b) and 2(c) show expanded regions of this trace corresponding to the temporal overlap of the leading and trailing edges of the two pulses. This part of the trace clearly depicts a train of subfemtosecond bunching of fast oscillations. The full width at half maximum of the beating is about 400 as. The repetition rate of the beating is $\approx(1.4\text{ fs})^{-1}$ which equals twice the optical cycle frequency of the fundamental. Figures 2(d) and 2(e) show two other expanded parts of the trace of Fig. 2(a). While the spectrum of Fig. 2(d) shows qualitatively the same behavior as that of Fig. 2(b) in 2(e), the temporal localization of the beating is not as clear as before. Note that the spectrum of Fig. 2(e) results from almost complete overlap of the two pulses.

The notable feature of the spectrum of Fig. 2(a) is the narrow structure, indicated by the arrow at the leading edge of the recorded trace. Since its width is at the limits of the temporal resolution, the measured height of the feature appears to strongly vary in different runs obtained under the same experimental conditions. The inset of Fig. 2(a) shows this feature in an expanded scale. The existence of additional ultranarrow spikes in the region of strong overlap of the two laser pulses cannot be excluded, since they may be hidden within the dense fast oscillations of the trace, which are of the same temporal scale. The width of the narrow feature is on the average of the order of 60 as and correlates with coherent continuum radiation as discussed in the next paragraph.

The Fourier transform of the total trace shown in Fig. 2(f) consists mainly of the 23rd to the 33rd odd harmonics, superimposed on a continuous background. The dashed line depicts the measured transmission of the filter. The faster decrease of the XUV intensity below the 27th harmonic (42.4 eV) could be attributed to the high degree of randomness of the relative phase of plateau harmonics (see, e.g., [11] and discussion below). Thus, besides subfemtosecond features, also broader components could be included in the temporal profile of the individual XUV

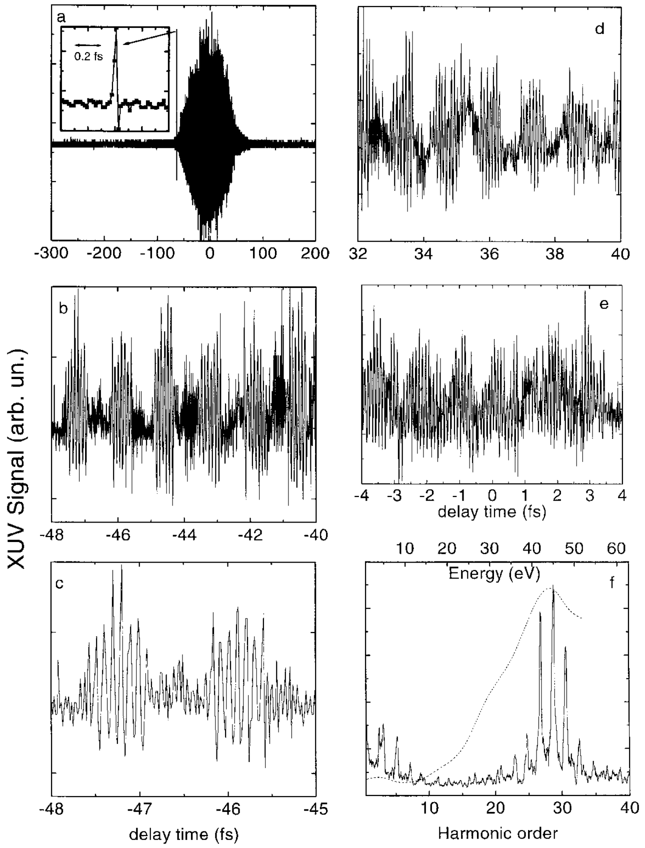

FIG. 2. Total visible-VUV-XUV signal filtered with a Al-Si filter as a function of delay of the two harmonic generating laser pulses. Total trace including an inset showing an expanded area of the ultranarrow feature at around 60 fs delay (a); an expanded region of the leading edge (b); an expanded region displaying one laser period (c); an expanded region from the trailing edge (d); an expanded region from the central part of the total trace (e); the power Fourier transform spectrum of the total trace. The dashed-line shows the measured filter transmission function (f).

pulses (not measurable in this work). The high absorption coefficient of Ar in this spectral range could also contribute to the above intensity reduction. However, absorption cannot be quantified due to the unknown absorbing atomic density distribution. The fast intensity drop above the 29th (45.5 eV) harmonic, although the filter transmits, reflects the cutoff region. Some low intensity peaks are also present in the low energy part of the spectrum. Three of them can be attributed to the 3rd, 5th, and 7th harmonics. Despite the high absorptivity of the filter in this region they are observable because of their much higher intensity (3–4 orders of magnitude) compared with the intensity of the plateau harmonics. The small peak overlapping with the 3rd harmonic may be attributed to a stimulated resonant transition and will not be further discussed as it is of no relevance to the present study. The dominant features of the frequency spectrum are astonishingly the odd harmonics produced by the atom and filtered by the response of the detection system and their relative phases. Moreover a coherent continuum background is clearly observable in the nonabsorbed region between 25 and 60 eV. Its width $\Delta\nu$ is of the order of 3×10^{15} Hz. The associated temporal width given by $\Delta\nu\Delta\tau \geq (2\pi)^{-1}$ has a minimum value of 53 as which is exactly the order of magnitude of the temporal width of the observed ultrashort spike at the leading edge of the temporal trace. The correlation is pretty good and implies that coherent continuum components of the harmonic generation spectrum are the key parameter in generating of attosecond pulses.

The observed features in the time and frequency spectra discussed above are compatible with the dynamics of the rescattering model for the production of the harmonics. The first pulse excites near to the atom core a coherent superposition of states that evolves as a function of time in the presence of the oscillating laser field and results into an oscillatory motion of the corresponding electronic wave packet. This wave packet may decay through electronic recombination in the vicinity of the core thus emitting harmonics and continuum radiation. The second laser pulse excites a second wave packet that, depending on the time delay between the two pulses, may interfere with the first one, and harmonic emission occurs from the recombination of the superposition of the two wave packets. In addition, the second laser pulse may stimulate the recombination of the electron excited by the first pulse. In both cases the second time delayed pulse is probing the dynamics of the initial coherent superposition of states and thus the dynamics of the oscillating dipoles and consequently the temporal profile of the emitted total radiation. Because of the used filter it essentially probes the transmitted phase locked higher harmonic components. The rescattered excited electronic wave packet formed appears twice per optical cycle in the vicinity of the nucleus where it can recombine and produce harmonics [8,11] exactly as observed in the present experiment. Continuum generation originating from sharply localized electronic wave packets of ultrashort temporal spread in the vicinity of the nucleus is also compatible with the rescattering model [7]. Propagation is further expected to affect the observed temporal localization, as it is observable only in a restricted Ar pressure range (2–4 mbar).

In order to further examine the validity of the argument that the above time delayed measurements probe the temporal profile of the emitted radiation, simulations have been performed that qualitatively reproduce the observed behavior. The simulation is based on solving the one dimensional damped classical equation of motion of a single electron interacting with the laser field and the atomic potential, which has been approximated with the quartic confining anharmonic potential as in previous works [5]. Field and potential parameters are chosen as to correspond to the experimental conditions. The acceleration is evaluated from the numerical solution of the Duffing equation and is further frequency filtered so as to simulate the experimental conditions. It is worth noting that only for the harmonics in the vicinity of the cutoff correlated relative phases have been obtained as in the previous studies [6,11]. The total

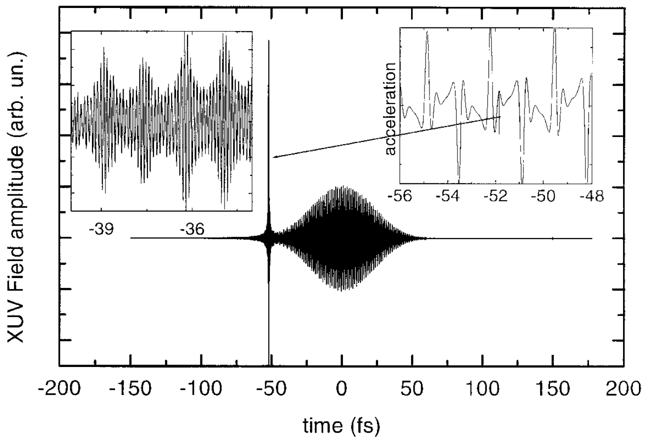

FIG. 3. Numerical simulation spectra (see text). Electric field amplitude of the 27th–33rd harmonics region (42–52 eV) as a function of time. The right inset illustrates the acceleration of the single electron. The left inset is an expanded area of the leading edge of the total trace.

frequency filtered field amplitude as a function of time shown in Fig. 3(a) consists of a train of subfemtosecond pulses similar to the observed beating in the experiment.

One possible reason for the observed reduced temporal localization in Fig. 2(e) could be the proportional to the intensity chirp of the harmonic fields [20,22]. This can be confirmed numerically, however, it needs further experimental clarification.

An ultranarrow structure, as observed in the experiment, can also be found in the simulation spectra of Fig. 3(a) at the leading edge of the calculated trace. It should be noted that this structure is reproduced in the calculation only for laser intensities so as to produce harmonics up to the experimentally measured cutoff. Decreasing the laser intensity first reduces the height of the structure; it further produces small amplitude narrow structures in the central part of the trace, while for even lower values it disappears. The simulations show that at the time this structure appears there is a very rapid variation of the acceleration [right inset of Fig. 3(a)]. This abrupt variation is compatible with the generation of very short duration pulses of coherent continuum radiation as observed in the measured spectra. Since the second laser pulse is slightly weaker than the first one, an ultrashort feature produced by the second one may primarily interfere with and probe the temporal behavior of the products of the first one. This could be a reason for the absence of the spike at the trailing edge.

In summary, temporal order of subfemtosecond duration is demonstrated in time domain correlation studies of a selected superposition of higher order harmonics and their associated continuum background. The produced and observed XUV radiation includes evidently coherent components with ultrashort temporal characteristics. They are in the form of an extremely narrow structure of less than 0.1 fs width correlating well with continuumlike coherent radiation and subfemtosecond trains of pulses correlating with the superposition of the selected phase locked harmonics. The present work provides the experimental evidence for the attosecond scale temporal order that governs the production process and products of the strong-field short-pulse higher order harmonic generation.

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