Single attosecond pulse generation in the multicycle-driver regime by adding a weak second-harmonic field

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We present a method of producing single attosecond pulses by high-harmonic generation with multicycle driver laser pulses. This can be achieved by tailoring the driving pulse so that attosecond pulses are produced only every full cycle of the oscillating laser field rather than every half-cycle. It is shown by classical and quantum-mechanical model calculations that even a minor addition (1%) of phase-locked second-harmonic light to the 800 nm fundamental driver pulse for high-harmonic generation leads to a major (15%) difference in the maximum kinetic energies of the recombining electrons in adjacent half-cycles. © 2006 Optical Society of America

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The availability of attosecond flashes of soft-x-ray–extreme-ultraviolet light[1–3] is on the verge of opening up a new field of science. Just as the femtosecond laser permitted observation of coherent molecular dynamics and Rydberg orbital motions in real time, attosecond pulses will be used in the future to study the coherent dynamics of electrons on their natural time scales. Attosecond pulses are produced in the process of high-harmonic generation[4] (HHG): an intense laser field interacts with an atom and generates coherent high-frequency radiation by ionizing, accelerating, and recombining part of the electronic wave function of the atom. All of this occurs every optical half-cycle when multicycle duration driving pulses are employed.

Trains of attosecond pulses have thus been observed in experiments[2] for the case of multicycle (40 fs) 800 nm driver pulses. However, for a conventional pump–probe-type experiment it is necessary to use one clearly defined pump and one clearly defined probe pulse.[3,5] It has been shown experimentally that with few-cycle drivers single attosecond pulses can be generated.[1] The requirement on the pulse duration is rather stringent: it has to be shorter than ~6 fs (for the commonly used Ti:sapphire 800 nm lasers), which is achievable only using state-of-the-art laser technology.

This Letter presents a method of using a very small amount (1%) of collinearly polarized phase-locked second-harmonic (SH) light to double the maximum allowable pulse duration. In the regime of few-cycle pulses, such an increase in pulse duration is a tremendous simplification with regard to the level of sophistication of the laser setup, and weak SH light can be generated easily from the main pulse. Earlier experiments using SH light to control the HHG process focused on the generation of even harmonics, controlling the polarization state of the harmonics,[6] and on enhancing the harmonic output.[7] Variation of the ellipticity of the fundamental field in time, produced by two different nearby frequencies, was considered as a method for single attosecond pulse production from long driving pulses.[8] For the same purpose another theoretical study proposed incommensurate two-color fields of comparable field strengths in an effort to control mainly the ionization step of the HHG process.[9] An interesting experiment with ac radio waves showed half-cycle symmetry breaking of field ionization of electrons if the second harmonic is added.[10] To date no experiment has produced single attosecond pulses from multicycle drivers.

In our approach, which should be experimentally straightforward to implement, the addition of a very small fraction (1%) of the phase-locked SH intensity to the fundamental is enough to significantly modulate the maximum kinetic energy that the electronic wave packet will have at the point of recombination with the atom. Instead of producing the same kinetic energy of electrons every half-cycle of the laser field, the addition of SH light leads to the highest kinetic energy being produced only every full cycle of the fundamental field (see Fig. 1). By applying the same filtering methods that have been used in conventional few-cycle attosecond pulse generation (i.e., using light only in the cutoff region of the harmonic spectrum), it is then possible to generate single attosecond pulses with multicycle drivers that are about a factor of 2 longer than the few-cycle ~6 fs pulses conventionally used. In the following, we first use a classical calculation based on the three-step model[4] to obtain an intuitive picture of the processes at work. Afterwards, we perform a quantum-mechanical simulation for 800 nm pulses with a model one-dimensional argon atom to compare our method with the traditional approach. The results are obtained by numerically solving the time-dependent Schrödinger equation.

In the classical approach, we start by considering the classical motion of an electron in a linearly polarized continuous-wave (cw) laser field with a small amount of phase-locked SH light: $E(t)=\cos(\omega t)+a\cos(2\omega t+\varphi)$, in which ω is the fundamental laser frequency, φ is the relative phase between the SH

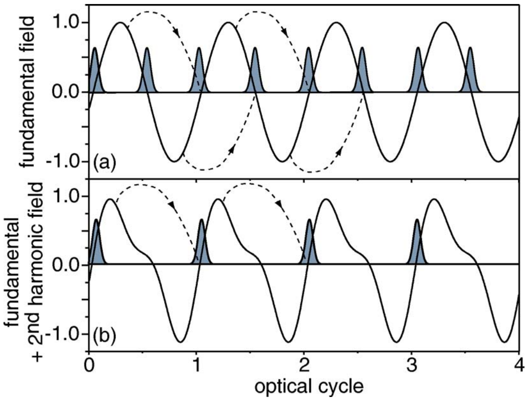

Fig. 1. (Color online) Illustration of the idea of full-cycle single attosecond pulse generation (solid curves, laser fields; shaded regions, generated attosecond pulses; dashed curves, time of birth and recombination of electrons). If the SH frequency is added to the driving laser field, the symmetry between first and second half-cycles is broken in particular for the fastest returning electrons, leading to emission of attosecond pulses in the cutoff photon energy region only once every full cycle of the laser field.

and the fundamental field, and a denotes the relative amplitude of the SH field compared with the fundamental. We evaluate the classical kinetic energy of the electronic wave packet that returns to the ion core where it can potentially recombine to emit a high-energy photon (Fig. 2). We see that a small amount of SH field ($a=0.1$, intensity 1% of fundamental) generates a significant change (±7%) in the maximum kinetic energy [Fig. 2(a)] compared with the classical cutoff[4] of 3.17 U_p (U_p is the ponderomotive potential). Depending on the relative phase φ between the SH and the fundamental, the highest kinetic energies are obtained in the first or the second half-cycle of the laser field. If we choose φ correctly ($\sim 0.6\pi$ rad, $\sim 1.6\pi$ rad) and consider the application of the same high-pass filtering method close to the cutoff region of the harmonic spectrum as is done in conventional single attosecond pulse generation,[1,3,5] we thus transmit only light that has been generated at one particular half-cycle of the strong fundamental field. This finding implies that to obtain isolated attosecond pulses it is now sufficient to make the laser pulse short enough to consist of a small number of full cycles as opposed to a small number of half-cycles as is the traditional case. For this classical calculation, the short and long trajectories[11] were considered. The short trajectory is easy to select experimentally and is typically achieved in attosecond pulse generation experiments by focusing the laser pulse in front of the gas jet or cell.[11]

To evaluate the validity of this result in a more realistic manner, we performed a simulation of high-harmonic attosecond pulse generation by numerical integration of the time-dependent Schrödinger equation. Our model system is a one-dimensional argon atom with a single active electron (SAE approximation).[12] The high-harmonic spectrum is calculated by Fourier transforming the time-dependent dipole-acceleration expectation value, applying Ehrenfest's theorem.[13,14] The spectrum is then high-pass filtered (as is done in single attosecond pulse generation experiments by applying metal filters and multilayer mirrors[1]) and transformed back into the time domain to yield the temporal structure of the attosecond pulse. The fundamental peak electric field strength of the 800 nm laser pulse is chosen as 0.85 a.u. (atomic units), corresponding to an intensity of $\sim 2.5\times 10^{14}$ W/cm^2, leading to a classical cutoff harmonic order of 41. The high-pass filter was chosen to transmit light above the 43rd harmonic order (which is still below the quantum mechanical cutoff of the 45th order observed in our simulations).

When a 6 fs driver pulse is used with a cosine carrier wave (meaning the electric field peaks at the maximum of the envelope), a single attosecond pulse (700 as duration, ~6 eV bandwidth) is produced [Fig. 3(a)] in qualitative agreement with experiments conducted in Ne.[15] If the pulse duration is increased to 12 fs, a series of attosecond pulses (pulse train) is generated instead [Fig. 3(b)], as expected.

However, if 1% of the fundamental intensity is added as a SH field at a phase difference of 1.5π rad, the attosecond pulse train is reduced to a single attosecond pulse (550 as, ~7 eV bandwidth) with only small satellite pulses [Fig. 3(c)]. The optimal phase is slightly different from the classically estimated 1.6π rad because of the use of a short pulse (time-

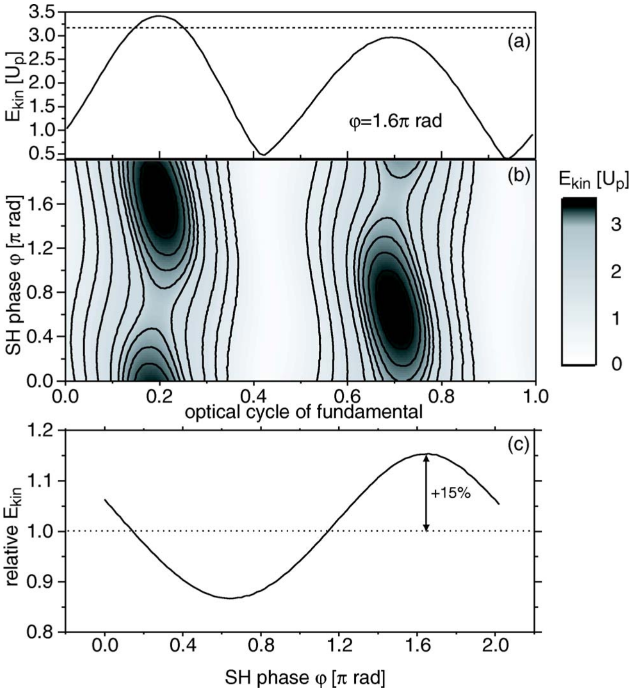

Fig. 2. (Color online) Classical analysis of the kinetic energy E_{kin} of the returning electron in the three-step model when a weak second harmonic (SH) field is added to the fundamental (1% intensity). (a) Symmetry breaking between the first and second half-cycles of the driving fundamental laser field leads to an asymmetry of the kinetic energies in the first and second half-cycles, shown here for a SH phase of $\varphi=1.6\pi$ rad (dashed line, classical cutoff energy at 3.17 U_p). (b) Depending on the relative phase φ of the SH with respect to the fundamental, the first or the second half-cycle can yield a higher kinetic energy. (c) Ratio of kinetic energies in the first versus the second half-cycle.

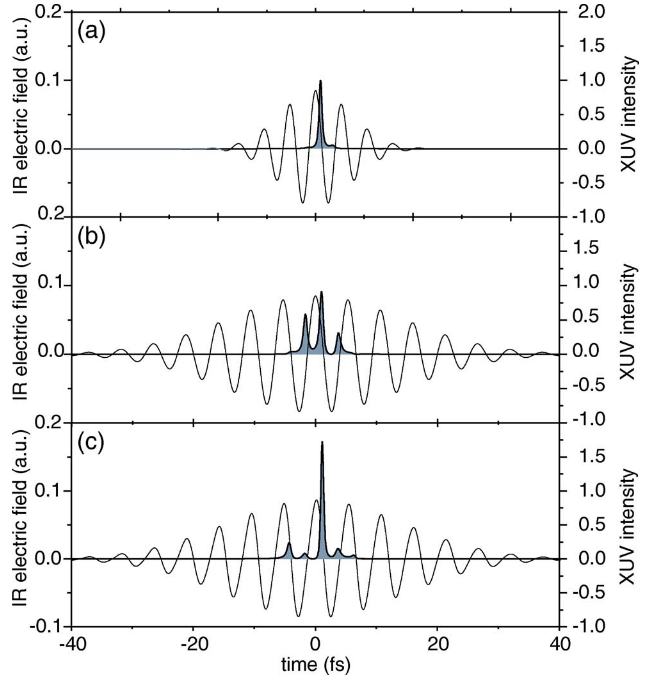

Fig. 3. (Color online) Quantum-mechanical simulation of attosecond pulse generation for different pulse durations of an 800 nm 2.5×10^{14} W/cm^2 laser pulse in a model Ar atom selecting cutoff harmonics by filtering (see text). (a) 6 fs cosine driving pulse, (b) 12 fs cosine driving pulse, (c) 12 fs cosine driving pulse with 1% intensity of its SH added at a relative phase of 1.5π rad.

varying envelope) rather than the classical cw model. The value of φ can be varied as much as $\pm0.2\pi$ to keep the satellite pulses below 20% of the main pulse intensity. The same is true even if the intensity of the SH field fluctuates by as much as ±30% (adding more SH further reduces the relative satellite intensity). This result ensures the experimental feasibility despite intensity variations and a phase slip of $\sim0.1-0.2\pi$ that will occur between the SH and the fundamental fields along a millimeter-scale interaction length in argon at typical phase-matching pressures of the order of ~0.1 bar.

Two additional benefits of this method are the reduced sensitivity of single attosecond pulse production on the carrier-envelope phase of the fundamental driver by a factor of 2 and an increase of the intensity of the single attosecond pulse (Fig. 3). A possible explanation for the latter effect is the shifting of the harmonic cutoff to higher photon energies, as is anticipated in the classical calculation [Fig. 2(a)]. Another explanation could come from the enhancement of HHG recently observed experimentally when a SH field is added to the fundamental.[7]

The intriguing fact that 1% (intensity) SH addition to the fundamental field changes the maximum kinetic energy of the electron considerably (±7%) can be easily understood: 1% intensity corresponds to 10% field strength compared with the fundamental field. Since the fundamental field strength is reduced by only 0.5% after this partial conversion into SH, a roughly 10% higher electric field is effectively available for additional electron acceleration. At the right phase φ this can lead to ~10% higher velocity and thus ~21% higher kinetic energy. The first experimental evidence for this effect should be the observation of even-order harmonics alongside the odd-order ones in (and beyond) the classical cutoff of the spectrum at the correct phase between the SH and the fundamental.

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