Intense few-cycle light pulses in the deep ultraviolet

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Abstract: We demonstrate that nonlinear frequency upconversion of few-cycle near-infrared (NIR) laser pulses, by means of harmonic generation in noble gases, is a promising approach for extending cutting-edge, few-cycle ultrafast technology into the deep ultraviolet and beyond, without the need for UV dispersion control. In our experiment, we generate 3.7-fs pulses in the deep UV (~4.6 eV) with adjustable polarization and gigawatt-scale peak power. We demonstrate that the implementation of this concept with a quasi-monocycle driver offers the potential for advancing UV pulse generation towards the 1-fs frontier.

OCIS codes:(190.7110) Ultrafast nonlinear optics; (190.7220) Upconversion; (320.5550) Pulses

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#100781 - $15.00 USD Received 15 Sep 2008; revised 17 Oct 2008; accepted 17 Oct 2008; published 3 Nov 2008

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1. Introduction

Ultrashort pulse laser sources are important tools for gaining insight into a wide range of microscopic processes via time-resolved spectroscopy. Ultrafast processes in the microcosm include atomic motion in molecules, which can be traced [1] and controlled [2] with femtosecond laser pulses, as well as electronic motion inside and between atoms, such as electron tunneling in atoms [3] or angstrom-scale charge transport in solid matter [4], which have been made perceivable with attosecond extreme ultraviolet (XUV) pulses and waveform-controlled few-cycle NIR light [5, 6].

#100781 - $15.00 USD Received 15 Sep 2008; revised 17 Oct 2008; accepted 17 Oct 2008; published 3 Nov 2008

Electron phenomena that are relevant to chemistry involve electrons residing in valence molecular orbitals which play a key role in the formation/rupture of the chemical bond [1], as well as in ultrafast intramolecular electron transfer [7] or charge migration [8]. Real-time access to these phenomena, which may spawn new ways of molecular control via directing charge in molecules with the electric field of light [9,10], requires tools that permit both triggering and probing valence-electron dynamics with sufficient resolution. Formation of a valence electron wavepacket involves excited states of energy of a few electronvolts (typically $\hbar\omega$ > 4 eV), spaced by ΔW_{exc} ranging from a fraction of an electronvolt to several electronvolts in different molecules. Preparation of a wavepacket comprised of the coherent superposition of the excited electronic states requires a broadband UV or deep UV pulse, whose transform-limited duration $(\tau_{\mathrm{UV}} \sim \Delta W_{\mathrm{exc}}/\hbar)$ ranges from a few femtoseconds to a few hundred attoseconds. Electron wavepacket control in molecules thus requires the extension of few-cycle laser technology, which is well established in the near infrared [11], into the deep ultraviolet spectral range and beyond.

2. The approach

To date, the generation of ultrashort-pulse UV radiation has relied primarily on nonlinear upconversion processes driven by picosecond and multi-cycle femtosecond pulses [12-15] yielding conversion efficiencies on the order of 0.1%. Numerous approaches, including non-degenerate four-wave mixing in gases [16,17], supercontinuum generation [18,19] or achromatic phase matching in solids [20], have resulted in significant enhancement of the photon yield as well as the spectral bandwidth of the generated ultraviolet pulses. However, these techniques rely on precision dispersion control in the deep ultraviolet, which is a formidable technological challenge and has limited the achievable pulse duration to the multi-cycle regime and compromised efficiency and scalability to higher pulse energies so far. As a result, state-of the art-UV pulse generation has been limited to durations longer than 7 fs [16].

He we take another approach. It is based on the use of intense few-cycle light pulses in the visible (VIS) and NIR spectral range, where the relevant technologies – spectral broadening via self-phase modulation in gas-filled hollow-core fibers and precision ultrabroad-band dispersion control via chirped multilayer mirrors – are well established [21] and allow for pushing the frontiers of ultrafast technology close to its ultimate limit set by the wave cycle [22]. We directly upconvert the carrier frequency of few-cycle VIS/NIR pulses to the UV range by means of harmonic generation in a noble gas jet. As the generation medium introduces little dispersion and is accommodated in a vacuum chamber, the process, if driven with transform-limited few-fs NIR pulses, directly yields near-transform-limited deep UV pulses of unprecedented duration without the need for sophisticated UV dispersion control techniques.

Moreover, the generation method – few-cycle driven harmonics – may be scalable towards the vacuum ultraviolet and pulse durations approaching the 1-fs frontier. Indeed, VIS/NIR pulses (of duration τ_0), upconverted to ultraviolet frequencies via an n^{th}-order nonlinear process originating from the n^{th}-order electric susceptibility ($\chi^{(n)}$), are shorter than the driving pulse by a factor of $\sqrt{n}$: $\tau_{n\omega} = \tau_0/\sqrt{n}$. Owing to the negligible dispersion of noble gases in the UV and VUV regime (see: Measurement accuracy and resolution), the upconverted pulse duration $\tau_{n\omega}$ can be preserved during the entire buildup of the harmonic pulse in the generation medium. With the generation medium incorporated in a vacuum chamber, the harmonic pulse can be delivered to experiments with this duration. These considerations suggest that a 780-nm, $\tau_0 = 6$ fs few-cycle pulse, which is now available in many laboratories, could yield UV pulses at a photon energy of ~4.5 eV with a duration of $\tau_{3\omega} \simeq 3.5$ fs. State-of-the-art quasi-monocycles in the sub-4-fs regime [22,23] might give

#100781 - $15.00 USD Received 15 Sep 2008; revised 17 Oct 2008; accepted 17 Oct 2008; published 3 Nov 2008

rise to 8.5-eV pulses with a duration of $\tau_{5\omega} \simeq 1.5$ fs. Generating harmonic radiation with the shortest possible driver pulses also enhances the harmonic conversion efficiency, owing to the enhanced coherence length due to a reduced ionization rate, just as in the case of high-order harmonic generation [24,25,11]. Preliminary calculations based on [26] predict a third-harmonic photon flux that is enhanced by more than an order of magnitude with 5-fs as compared to that of 20-fs driver pulses of the same energy.

3. Experimental-results

In the first series of experiments we have used 780-nm, 0.25-mJ laser pulses with duration of about $\tau_0 \simeq 6$ fs. They have been focused (f=50 cm) into a quasi-static gas cell (~2.5 mm), which is accommodated in a vacuum chamber as illustrated schematically in Fig. 1. This experimental scheme is almost identical to the one employed for producing XUV attosecond pulses [5,6], and enables intensities on the order of ~4-5 10^{14} W/cm^2 at the focus.

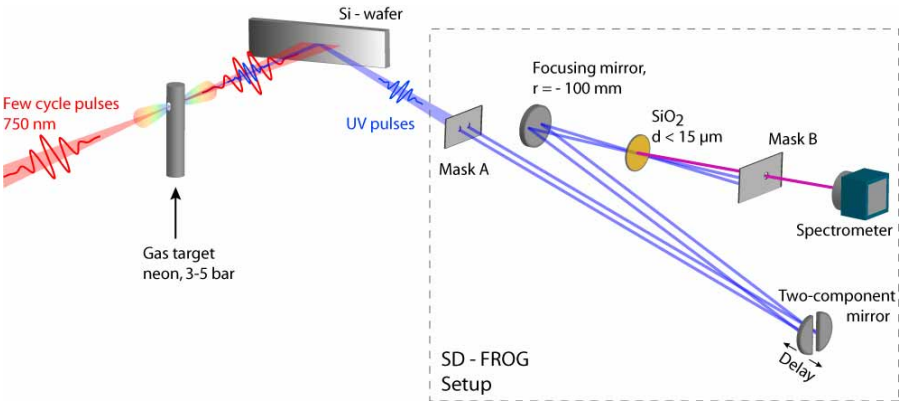

Fig. 1. Generation and measurement of sub-4-fs pulses in the deep ultraviolet. Few-cycle pulses of visible and near-infrared light with durations of 3.5 to 6 fs are focused into a quasi-static gas cell formed by a tube with an inner diameter of 2.5 mm, with two holes in its 0.3-mm-thick wall to generate harmonic pulses in the deep ultraviolet. The intensity of the fundamental field is suppressed by more than two orders of magnitude upon a Brewster reflection from a silicon wafer, reflecting >42% of the generated ultraviolet radiation. The resulting pulses are temporally characterized with a dispersion-minimized SD-FROG apparatus (dashed frame) [29], employing exclusively reflective optics and an ultrathin (<15 µm) UV-grade fused silica plate as a nonlinear medium (see: Measurement accuracy and resolution) The entire apparatus shown is incorporated in a vacuum chamber.

3.1 Upconversion efficiency

We investigated the upconversion efficiency of our few-cycle laser pulses into ultraviolet radiation in the range of 3.8-5.6 eV (third harmonic) as a function of the atom density of the quasi static cell. Figure 2(a) summarizes results for xenon, argon, neon and helium. The photon yield in the case of xenon and argon is found to reach its maximum at a pressure of ~0.3 bar and ~1 bar, respectively, and is followed by a roll-off at higher pressure. Neon yields the highest photon flux at a pressure of ~7 bar, which is almost a factor of four higher than that of Argon at the corresponding optimal pressure. Insufficient pumping of helium has prevented us from exploring the optimum pressure range for this gas. The highest harmonic photon flux emerges from neon, yielding pulse energies > 1.4 µJ and a conversion efficiency of ~0.6 %. This deep-UV photon yield is to our knowledge the highest demonstrated to date by direct nonlinear conversion of a femtosecond pulse in a gaseous nonlinear medium. The high efficiency of neon can be attributed to its atomic properties which -at the intensity range of this study- afford a reasonable degree of nonlinear polarizability at a reduced ionization rate. The latter is essential to achieving low degrees of ionization-induced phase mismatch.

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Our intuitive calculations are supportive of this picture [26]. A representative spectrum of the ultraviolet emission from the neon source is shown in Fig. 2(b).

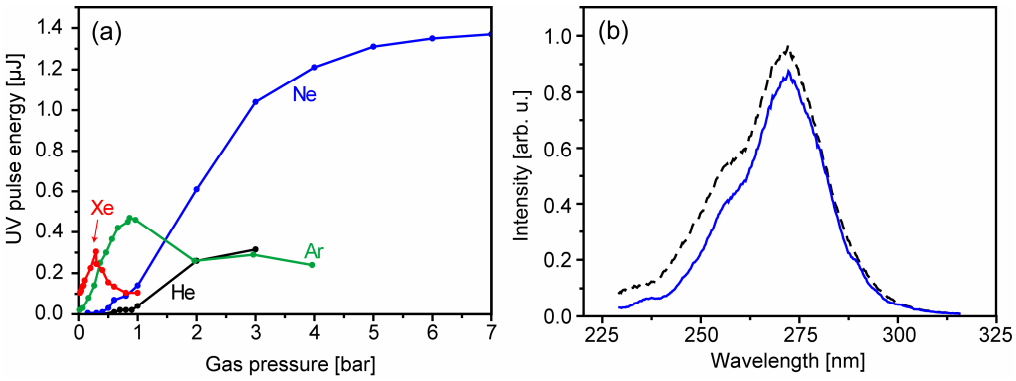

Fig. 2. UV pulse characteristics. (a) Energy of UV pulses generated by gently focusing (f=50 cm, 1/e beam diameter at beam waist ~100 μm, intensity profile) 6-fs, 780-nm pulses with an energy of 0.25 mJ into a neon target (effective thickness ~2.5 mm) versus target gas pressure for xenon, argon, neon and helium. The maximum energy is obtained with neon and its value is in good agreement with simple quasi-phase matching calculations. The optimal pressure range for helium could not be reached because of insufficient vacuum pumping power for this type of gas. (b) Deep ultraviolet spectra emerging directly from the neon source at a pressure of 5 bar (black curve) and arriving at the nonlinear medium of the SD-FROG apparatus (blue curve) following a reflection off a Si-wafer at Brewster's angle, and two reflections from UV-enhanced Al-mirrors (see Fig. 1).

Brewster angle reflection-based suppression of the fundamental is a technique broadly employed in VUV experiments [27]. A four orders of magnitude suppression of the fundamental (experimentally 2-3 orders) at 800 nm at the expense of approximately 60% of photons in the ultraviolet range permits a dispersionless isolation of the third harmonic in lieu of advanced UV optics or grating-based broadband ultraviolet compressors.

3.2 Temporal characterization

For temporal characterization of the generated deep UV pulses, we have developed a dispersion-minimized SD-FROG (self-diffraction frequency-resolved optical gating) [28,29] apparatus (see Fig. 1), enclosed in a vacuum chamber that also incorporates the harmonic source.

A mask with two openings generates two beams, each less than 1 mm in size, as required for the realization of the SD-FROG scheme. A mirror segmented into two parts is used to delay the pulses delivered in the two beams with respect to each other. The two segments can be accurately delayed with nanometer precision using a piezo-controlled translational stage. The beams are in turn focused into a <15 μm UV-grade fused silica nonlinear medium by means of a focusing (focal length = 5 cm) Al-coated mirror. One of the diffracted beams steered only by broadband reflective optics is spatially isolated and focused into the entrance of our spectrometer. Less than 50 nJ of UV pulse energy was sufficient to generate a well discernible diffracted beam resulting in an excellent signal-to-noise ratio.

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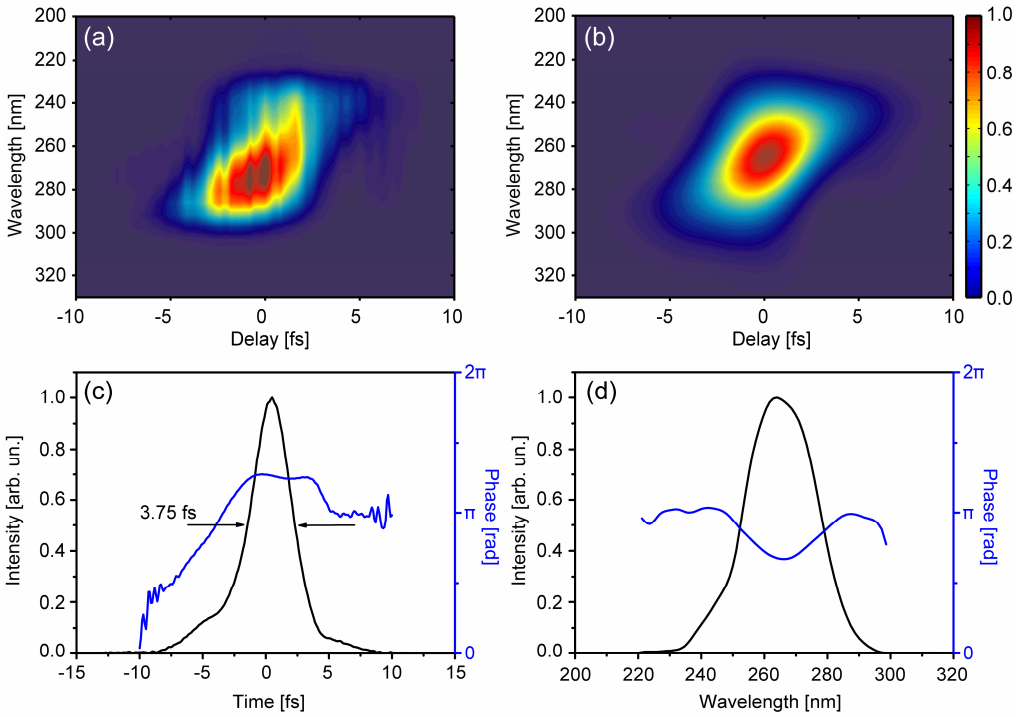

Fig. 3. Temporal characterization of few-cycle pulses in the deep ultraviolet. (a) Measured SD-FROG trace over a delay range of 20 fs in steps of 0.2 fs. (b) Spectrogram reconstructed with commercial software (Femtosoft FROG 3.2.2). The retrieval error in our analysis was 0.010 using a grid size of 256x256. (c) Retrieved temporal intensity profile and phase of the ultraviolet pulses yielding a FWHM duration of $\tau_p = 3.7$ fs. (d) Retrieved spectral intensity and phase of the deep ultraviolet emission.

Figures 3(a) and 3(b) depict the measured and the reconstructed SD-FROG spectrogram of third harmonic pulses generated from the neon source, respectively. The retrieved pulse has a duration of $\tau_{3\omega} \simeq 3.7$ fs and a carrier photon energy of ~4.6 eV implies ~4 field oscillations within the full width at half maximum of the temporal intensity profile of the pulse. The pulse carries a small positive chirp of ~5 fs^2, which can be attributed to various factors including dispersion introduced by the Al-mirror protective coatings employed in the FROG setup as well as by the thin (<15 μm) nonlinear medium. Nevertheless, the retrieved duration is close to the bandwidth limit which is calculated as 3.4 fs via a Fourier transform of the spectrum shown in Fig. 2(b).

3.3 Measurement accuracy and resolution

The temporal smearing resulting from our non-collinear beam geometry was calculated to be ~0.2 fs for a crossing angle of 3°. Self diffraction is not a phase matched process and therefore particular attention has to be paid in order to minimize the phase mismatch across the spectrum of the pulse (which gives rise to spectral distortions). The <15 μm UV-grade fused silica plate used as a nonlinear medium, which is substantially shorter than the difference in coherence length L_{coh} of the nonlinear process at the two edges of our spectrum, warrants nearly uniform phase matching over the entire bandwidth. Indeed, following [28] we have calculated the coherence length at the edges of the spectrum as $L_{coh} = 58$ μm at wavelength $\lambda_{UV} = 200$ nm and $L_{coh} = 80$ μm at $\lambda_{UV} = 300$ nm.

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Material dispersion of the nonlinear medium is another key parameter that needs to be considered. For these experiments, it was found to introduce <0.26 fs of temporal smearing to the measurement of our pulses, which results in ~7% overestimation of the pulse duration. This temporal smearing becomes more important for shorter UV pulses. Propagation of a sub-3-fs pulse at a central wavelength $\lambda_{UV} = 260$ nm through the nonlinear medium (~15 μm) was estimated to yield a distortion of ~15% with respect to its original pulse duration. However, it does not comprise a strict limitation of the approach, as the effect of material dispersion may be integrated in the retrieval algorithm.

3.4 Dispersionless polarization control

In order to be able to control the polarization of our pulses without compromising their duration and intensity, we have realized an all-reflective dispersionless polarization management scheme, based on a single reflection of the ultraviolet pulses off an aluminum-coated mirror [30-32]. We have been able to generate pulses with an arbitrary state of polarization while keeping the throughput of the control unit above 95%. Results are shown in Fig. 4.

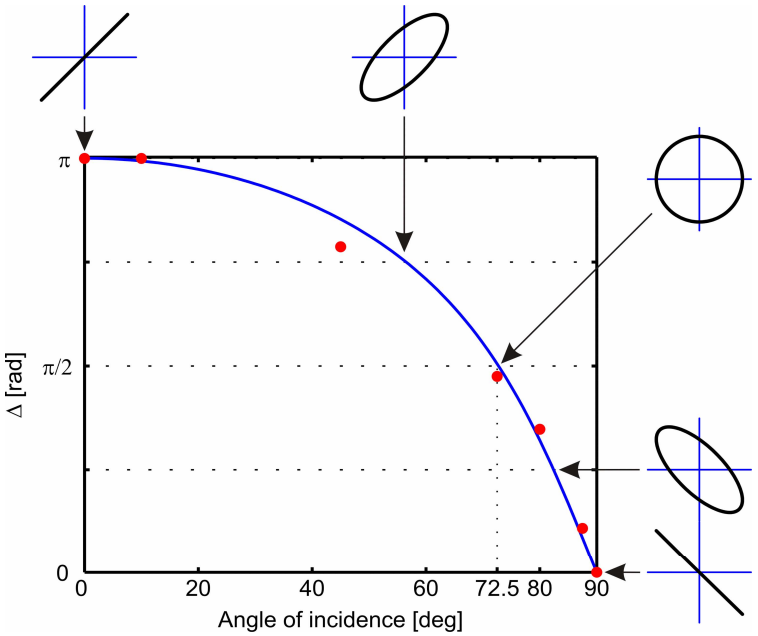

Fig. 4. Broadband dispersionless ellipticity control of ultrashort ultraviolet pulses. Calculated phase shift Δ (blue line) between parallel and perpendicular polarization components, introduced by a reflection from an aluminum surface at different angles of incidence. Measured data are shown as red dots. The corresponding polarization state is indicated by the Lissajous figures linked to each point. Circular polarization is achieved for an incidence angle of 72.5°.

3.5 Towards sub-2-fs pulses

Finally, we have investigated the potential of our approach for generating even shorter UV pulses, by studying the influence of the duration of driving pulses on the UV spectral bandwidth. The duration τ_0 of the VIS/NIR driving pulse is adjusted by varying the spectral broadening in the hollow fiber, whilst the pulse energy is reduced by less than 10%. The spectra of the VIS/NIR driver pulses of durations $\tau_0 = 4.1$ fs, and $\tau_0 = 3.5$ fs and the corresponding spectra of the deep-UV pulses produced with these pulses are shown in Fig. 5. Instrumental limits of our intensity calibrated spectrometer (lack of VUV detection) have prevented the measurement of spectral components beyond ~5.75 eV, and the temporal

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characterization of the broadband UV/VUV pulses. Nevertheless, both the clear trend of UV bandwidth broadening with decreasing driver pulse duration τ_0, as well as the shape of the UV spectra provide clear indication that they substantially extend into the vacuum ultraviolet for $\tau_0 \leq 4$ fs. These measurements suggest that microjoule-energy, sub-2-fs ultraviolet pulses may indeed be feasible by implementing our concept with quasi-monocycle driving light. Further studies of the underpinning phenomena in this regime will be necessary in the future in order to explore the full potential offered by quasimonocycle pulses.

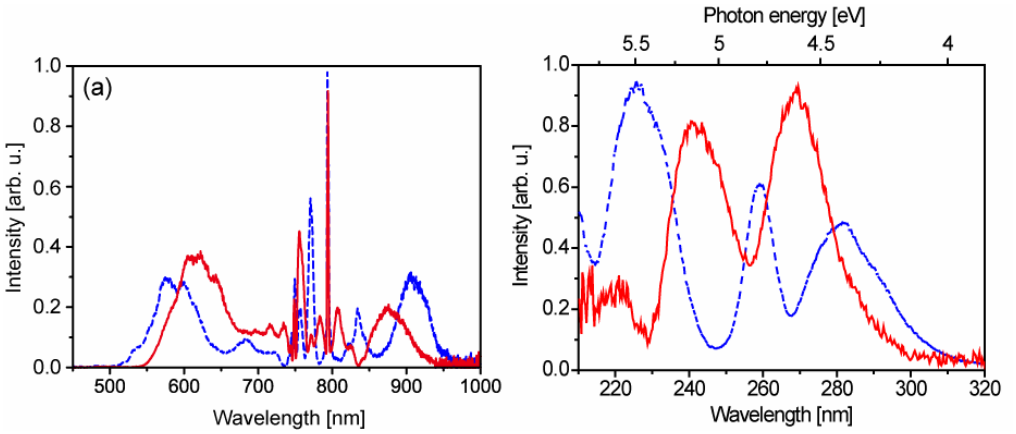

Fig. 5. Ultrawide-band deep-UV emission driven by quasi-monocycle laser pulses. (a) Spectra corresponding to driving pulses of $\tau_0 = 4.1$ fs (red), and $\tau_0 = 3.5$ fs (blue) generated in a hollow-fiber/chirped-mirror compressor. (b) Emitted spectra of ultraviolet pulses generated by focusing these driving pulses into the neon target. The experimental conditions are the same as described in Fig. 2. Same color coding as in (a) is applied.

4. Conclusions

With powerful, polarization-controlled few-femtosecond pulses readily available in the deep ultraviolet, the door is now open to a wide range of applications that include the ultrafast electron-controlled reactivity of molecules [10] to the generation of giant magnetic fields by driving electron ring currents in molecules with circularly-polarized few-cycle UV light [33]. At intensities > 10^{14} W/cm^2, reachable with moderate focusing of our > 0.4-GW pulses, field-control of electronic processes [34] sensitive to the carrier-envelope phase might also become possible. Indeed, carrier-envelope phase control is best transferred to UV pulses [35] in the absence of compression modules [36]. The presented UV generation scheme may be scalable to the VUV and can, thanks to its simplicity, be readily combined with attosecond XUV pulse generation schemes based on the same VIS/NIR driver pulse [37]. Intense, waveform-controlled few-cycle pulses with spectral components extending from the NIR to the VUV, in combination with synchronized isolated attosecond XUV pulses, will afford unprecedented capabilities for controlling and exploring electron processes in molecules and solids relevant to physical and chemical technologies as well as biological function.

Acknowledgments

We would like to thank H. Schroeder and W. Fuss for fruitful discussions, W. Schweinberger and V.S. Yakovlev for support. We acknowledge financial support from the XTRA European training network and the Munich Centre for Advanced Photonics, www.munich-photonics.de). E.G. acknowledges a Marie-Curie fellowship (MEIF-CT-2005-02440) and a Marie-Curie Reintegration grant (MERG-CT-2007-208643). R.K. acknowledges funding from the Sofja Kovalevskaja Award of the Alexander von Humboldt Foundation.

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