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Generation of sub-two-cycle pulses tunable around 1.8 µm with passively stabilized carrier-envelope phase at 100 kHz repetition rate

C. Homann[1], M. Bradler[1], M. Förster[2], P. Hommelhoff[2], and E. Riedle[1]

[1]LS für BioMolekulare Optik, Ludwig-Maximilians-Universität, 80538 München, Germany
[2]Ultrafast Quantum Optics Group, MPI für Quantenoptik, 85748 Garching, Germany

Abstract. We present an efficient concept for generating carrier-envelope phase stable pulses tunable between 1.6 and 2.0 µm with durations down to 8.2 fs based on nonlinear frequency conversion. As a first application we measure the high nonlinearity of multiphoton photoemission from a nanoscale metal tip.

1 Sub-two-cycle pulses by OPA and DFG at 100 kHz

The generation of few-cycle light pulses with stable carrier-envelope phase (CEP) at wavelengths around 2 µm is still a challenging but worthwhile task [1]. High-power pulses at these wavelengths are of interest because they allow extending the cutoff of high-order harmonic generation towards the water window. But also in the CEP-sensitive emission of electrons from metal nanostructures, where pulse energies of only 240 pJ at 800 nm sufficed to generate electrons with kinetic energies of up to 13 eV, the use of longer wavelengths is expected to greatly enhance electron energies and to give further insight into the interplay between multiphoton and tunneling ionization [2]. For investigations along these lines, CEP stable sub-two-cycle pulses with high repetition rates are a promising tool.

Here we present an efficient and simple scheme for the generation of such pulses with a duration around 10 fs and energies on the order of 100 nJ at a repetition rate of 100 kHz [3]. The pulses are tunable in the range from 1.6 to 2.0 µm while maintaining a sub-two-cycle duration (see Fig. 1). To the best of our knowledge this constitutes the shortest pulse durations in this wavelength range for repetition rates above 1 kHz, combined with excellent tunability.

Our concept is based on supercontinuum generation (SCG) in bulk material followed by noncollinear optical parametric amplification for the generation of a broadband visible spectrum, and subsequent broadband difference frequency generation (DFG) with the narrow-band pump laser to generate the passively CEP stabilized output around 1.8 µm. A commercial Yb:KYW based pump laser (Jenlas® D2.fs; JENOPTIK Laser GmbH) delivers ~ 300 fs pulses at 1025 nm with an energy of 40 µJ at 100 kHz repetition rate. Approximately 5 % of the used 32 µJ are split off for SCG in a 4 mm thick YAG crystal, producing the seed for the NOPA. The remainder of the pump pulse is frequency-doubled in a BBO crystal with an efficiency of about 30 %. The continuum seed is then noncollinearly amplified by the 512 nm pump pulses in a 3 mm thick BBO crystal. The amplified spectrum can be tuned in central wavelength between 650 nm and 700 nm while maintaining a Fourier limit below 10 fs and energies around 2 µJ per pulse. After collimation with a spherical mirror the output pulses are compressed using fused silica prisms.

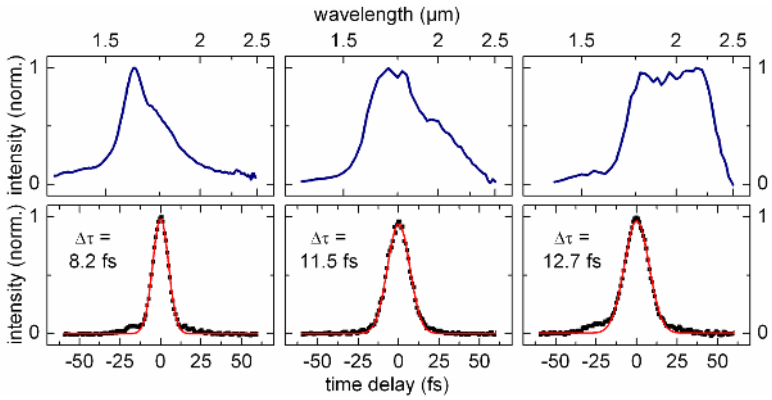

Fig. 1. Spectra and corresponding autocorrelation measurements showing the wavelength tunability of our concept while maintaining a sub-two-cycle pulse duration (Δτ: deconvolved FWHM Gaussian pulse duration).

The 1025 nm light remaining after frequency doubling is rotated in polarization by 90°. It is then focused into a 0.8 mm thick BBO crystal together with the compressed NOPA output to perform type-I DFG to the infrared. This crystal thickness is much less than typically used for an OPA stage and therefore allows a largely increased acceptance bandwidth. To avoid the need for dichroic optics, the two beams are combined under a small angle, which is kept as small as possible (~ 0.9°) to limit the associated spatial chirp of the infrared pulse. For the chosen angle the angular dispersion between the spectral components at 1.6 μm and 2.2 μm of the output pulse is 0.55° and therewith less than the natural divergence of ~ 1°.

The spectrum of the infrared output can be tuned in a considerable wavelength range by changing the NOPA spectrum and readjusting the phase-matching angle of the DFG and the time delay between the NOPA output and the 1025 nm pulses (see Fig. 1). The deconvoluted pulse durations, obtained from autocorrelation measurements, lie between 8.2 fs and 12.7 fs, corresponding to only 1.5 to 1.9 optical cycles. Pulse energies of up to 145 nJ are achieved, however with slightly longer pulses (11.7 fs at 1.6 μm central wavelength). For the shortest pulse durations pulse energies of 40 nJ are achieved.

The compression of the infrared pulses is solely adjusted by the chirp of the NOPA output and thus the prism compressor in the visible. The shortest IR pulses are achieved when the NOPA pulses are also nearly optimally compressed. This scheme even allows compensating for material dispersion in the infrared, introduced by, e.g., an entrance window to a vacuum chamber.

2 Stability and control of the carrier-envelope phase

The NOPA output inherits the CEP fluctuations of the supercontinuum seed and hence of the 1025 nm pump. Since the NOPA output is subsequently difference frequency mixed with the 1025 nm light, the infrared output is expected to be CEP stable [1]. To verify this, we set up an f-2f interferometer. To achieve the necessary bandwidth, we coupled the infrared pulses into a highly nonlinear fiber. The broadened spectrum and the frequency-doubled light were then focused into a spectrometer with a time delay of ~ 400 fs. For proper alignment of the continuum generation, we observe high contrast interferences that directly show the CEP stability of the IR pulses. When integrating over 1 ms and acquiring 500 consecutive spectra (i.e. 0.5 s acquisition time), we measure phase fluctuations of only 78.5 mrad rms. Fig. 2 shows a measurement over 5 minutes (averaged over 100 ms). After 1.5 minutes we induce a CEP change of π by translating one of a pair of wedges located in the 1025 nm beam before the DFG stage. After 3 minutes the wedge is moved back. This clearly shows that we have full control of the CEP and can compensate for the observed slow drift. For the slower fluctuations (0.1 Hz to 5 Hz) we find a value of 135 mrad rms.

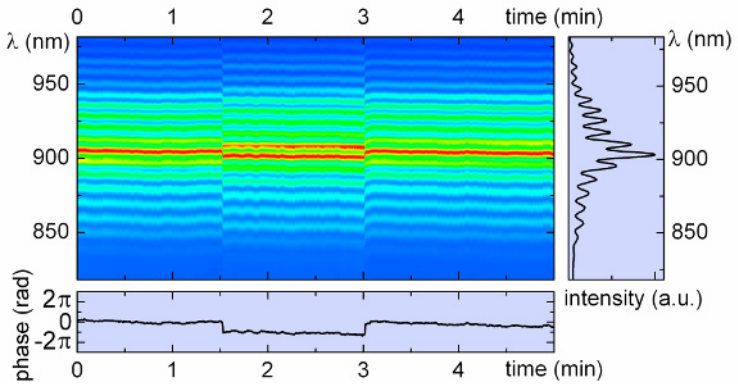

Fig. 2. f-2f interference measurement over 5 min. The right panel shows the spectrum of a single scan, the lower panel the evaluated phase. After 1.5 min. a CEP shift of π is induced by moving a wedge, which is reversed after 3 min.

3 Electron emission from nanoscale metal tips and outlook

As first experimental application of our new source, we tightly focus the IR pulses onto a nanoscale tungsten tip, with the laser polarization parallel to the tip-pointing direction (see Fig. 3(a)). This leads to photoemission of electrons from the tip, which we accelerate by an electric dc field towards a microchannel plate (MCP) detector. When measuring the electron current I on the MCP screen as a function of the incident pulse energy E, we find a $I \propto E^{3.8}$ dependence, indicating dominating multiphoton photoemission in this pulse energy range (see Fig. 3(b),(c) and [2]). The next step will be the measurement of the energy spectrum of the emitted electrons, which is expected to have a clear dependence on the CEP of the IR pulses.

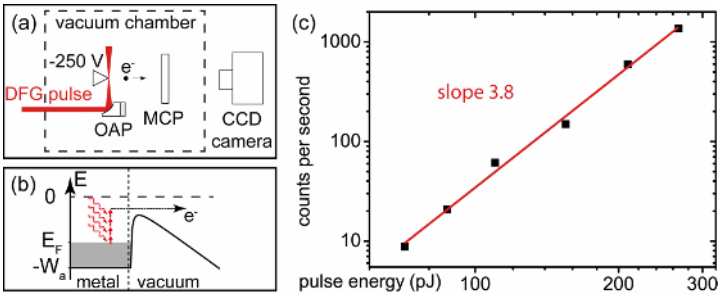

Fig. 3. (a) Schematic of the setup for the photoemission of electrons from a nanoscale metal tip. (b) Schematic of the multiphoton photoemission step. (c) Measured electron counts per second on the MCP as function of the laser pulse energy in a double logarithmic plot with a linear fit.

Our concept is also an interesting seed source for high power applications. It provides much more seed energy than the nowadays used few pJ to nJ, which will lead to a drastic improvement in pulse contrast. The combination of SCG, preamplification in the visible and subsequent DFG is furthermore expected to provide few-cycle pulses throughout a large part of the IR when used with different crystals.

References

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