Towards parametric synthesis of millijoule-level two-octave-wide optical waveforms for strong-field experiments

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Abstract: We discuss a carrier-envelope phase-stable, 3-channel parametric synthesizer for creating a 2-octave-wide spectrum $(0.5-2.2\mu m)$. The synthesizer output is straightforwardly scalable to the millijoule level with ~1.8-fs pulse duration, rendering it a versatile tool for attosecond science.

The coherent synthesis of custom-tailored, intense, few-cycle optical waveforms is currently one of the most intriguing and promising frontiers of attosecond science and strong-field physics, opening up unprecedented prospects, e.g., for precision control of strong-field interactions in atoms, molecules and solids, for the generation of intense isolated attosecond pulses, and for attosecond pump-probe spectroscopy employing ultrashort pulses in the VIS/IR and XUV/soft-X-ray regions. The key ingredients of coherent pulse synthesis are (i) the creation of an ultrabroad spectrum, (ii) extremely precise dispersion control over the whole bandwidth, (iii) sub-cycle timing synchronization as well as tight stabilization of the relative phases between the different pulses, and (iv) carrier-envelope phase (CEP) stability required for creating reproducible electric-field transients.

Recently, coherent pulse synthesis based on supercontinuum generation in a neon-filled hollow-core fiber compressor allowed the generation of sub-cycle ~ 300 - μ J optical pulses ($\sim 250~\mu$ J energy in the NIR channel, $\sim 35~\mu$ J in the VIS, and $\sim 15~\mu$ J in the VIS-UV). However, the energy throughput of such hollow-core fiber compressors is limited by ionization losses in the gas medium, thus preventing further energy upscaling to the (multi-)mJ level, which is required for many interesting applications in attosecond science. In contrast, parametric synthesizers do not face an energy-scaling bottleneck, and spectral extension into the MIR region is particularly appealing for the realization of bright coherent tabletop high-harmonic sources in the water-window and keV X-ray region.

In our earlier works, we already demonstrated coherent pulse synthesis between ultrabroadband 870-nm and 2.15- μ m pulses based on optical parametric chirped-pulse amplification (OPCPA) resulting in a 15- μ J sub-cycle waveform, and also between two optical parametric amplifiers (OPAs) with 1-2 μ J energy each. Here, we present our ongoing development of a novel 3-channel parametric synthesizer for creating a 2-octave-wide spectrum, which can straightforwardly be upscaled in energy to the millijoule-level with ~1.8-fs FWHM duration.

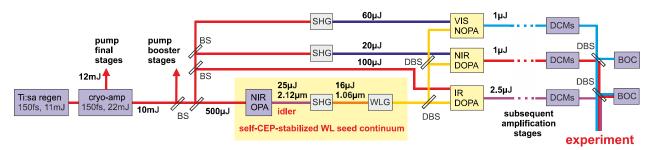


Fig. 1. Scheme of the 2-octave-wide parametric synthesizer. BS, beam splitter; SHG, second-harmonic generation; WLG, white-light generation; DBS, dichroic beam splitter; NOPA, noncollinear OPA; DOPA, degenerate OPA; DCMs, double-chirped mirrors; BOC, balanced optical cross-correlator. Energy upscaling in subsequent amplification stages is only indicated for the sake of clarity.

The scheme of the parametric synthesizer is depicted in Fig. 1. Starting point is a cryogenically cooled 1-kHz Ti:sapphire chirped-pulse amplification system delivering compressed 150-fs ~10-mJ 0.8-μm pulses from its main output port, and ~12-mJ pulses from its second output port (that eventually will be used for pumping the final booster OPAs). A CEP-stable seed continuum⁵ covering the bandwidth of all three OPAs (0.5-2.2μm) is obtained by white-light generation in a 3-mm-long YAG crystal from the second harmonic (1.06 μm) of the passively CEP-stable idler (2.12 μm) of a NIR OPA pumped by the fundamental pulses. Using custom-designed dichroic beam splitters, the CEP-stable white-light continuum is diverted to seed the VIS noncollinear OPA (NOPA) as well as the NIR and IR degenerate OPAs (DOPAs). These OPAs are pumped by the 400-nm second-harmonic and 800-nm fundamental pulses, respectively, as shown in Fig. 1. After parametric amplification, the three OPA outputs can individually be recompressed using custom-tailored double-chirped mirrors (DCMs). Finally, in order to synthesize a coherent ultrashort pulse from these three OPAs, the relative timing of the pulses will be tightly locked using balanced optical cross-correlators (BOCs), which have been demonstrated to achieve sub-cycle synchronization with <30-as RMS timing jitter.^{2,3}

Fig. 2(a) shows the measured output spectra from the first amplification stages operating in parallel. All three OPAs employ 1-mm-thick type-I BBO crystals (the phase-matching angle θ is 31°, 29°, and 20.1° for the VIS NOPA, NIR DOPA and IR DOPA, respectively). The VIS NOPA and NIR DOPA continuously cover the region from 490 nm to 1000 nm. The IR DOPA yields a spectrum from 1.1 μ m to 2.2 μ m, the peak centered at 1.06 μ m stems from the residual idler-SHG (creating the white-light seed) and is not amplified in the IR DOPA. The transform-limited FWHM pulse duration from the synthesis of these three spectra is ~1.8 fs [see Fig. 2(b)].

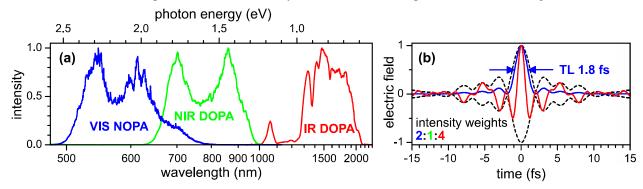


Fig. 2. (a) Output spectra from the first OPA stages. The energies of the amplified signal pulses are 1 μ J for the VIS NOPA and NIR DOPA, and 2.5 μ J for the IR DOPA. (b) The transform-limited (TL) pulse synthesized from the weighted spectra in (a) corresponds to a ~1.8-fs FWHM pulse duration. The red curve is the electric field E(t), the dashed curves indicate the field envelope, the temporal intensity profile I(t) is the blue trace.

Our current work aims to scale up the energies (by two subsequent amplification stages) to ~ 0.5 mJ for the VIS NOPA/NIR DOPA and ~ 2 mJ for the IR DOPA using the remaining 8.5 mJ of pump from the main and 12 mJ from the second output. The enabling technology for the final pulse compression to a ~ 1.8 -fs duration and successful beam recombining (avoiding *B*-integral issues) are state-of-the-art DCM pairs with ultralow residual ripple in the resulting total group-delay dispersion over the full bandwidth (0.5-2.2 μ m), which are at present in the process of being fabricated. Temporal characterization of the synthesized two-octave-spanning optical waveforms will be performed by two-dimensional spectral shearing interferometry (2DSI).^{2,3}

We foresee that our 3-channel synthesizer, when scaled in energy to the millijoule level, will become a versatile tool for controlling strong-field interactions in atoms, molecules and solids and for attosecond pump-probe spectroscopy employing ultrashort pulses in the VIS/IR and XUV/soft-X-ray regions.

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