Generation of sub-two-cycle CEP-stable optical pulses at 3.5 µm by multiple-plate pulse compression for high-harmonic generation in crystals

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Abstract. Multiple-plate pulse compression of femtosecond mid-infrared pulses is demonstrated using YAG and Si windows. With this robust compression scheme, we produce sub-two-cycle, CEP-stable optical pulses and observe CEP-dependent high harmonic generation in crystals.

1 Introduction

A carrier-envelope phase(CEP)-stable, few-cycle mid-IR source is an important tool to upscale attosecond science. Since the cutoff energy in high harmonic generation (HHG) scales with $\lambda^2$ for gaseous media, CEP-stable, few-cycle, long-wavelength optical pulses are required to produce isolated attosecond pulses in the keV spectral range. In addition, HHG in crystals is becoming an emerging topic in attosecond science. For either gaseous or crystalline media, generated high harmonics become dependent on the CEP of few-cycle optical pulses, which can be relevant to field-driven carrier dynamics on the sub-cycle time scale, such as the sub-cycle chirp of interband harmonics [1].

So far, various optical parametric amplifiers (OPAs) are developed to produce CEP-stable, high-energy, mid-IR ($\lambda=3-10$ µm) pulses [2, 3]. However, their pulse durations are longer than 3 optical cycles, mainly due to limited phase-matching bandwidth in OPAs with customary used nonlinear crystals [4]. In order to demonstrate a compact and robust scheme to produce sub-mJ, few-cycle pulses in the mid-IR range, our approach is to employ a multiple-plate pulse compression technique to combine spectral-broadening in bulk materials and free-space propagation in a discrete manner.

We apply this technique to compress CEP-stabilized, 10-cycle optical pulses at 3.5 µm generated from a KTA-based OPA (Fig. 1(a) and (b)) down to the sub-two-cycle regime [5]. This technique is inspired by the multiple-thin-plate supercontinuum generation in the visible and near-infrared regions using the output from a Ti:Sapphire chirp-pulse amplifier [6]. We utilize spectral broadening of mid-IR pulses in a YAG plate [7] and a Si plate and empirically find that a combination of these plates with a thickness of a few millimeters efficiently broadens the spectrum of femtosecond mid-IR pulses without deterioration of

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beam quality. This multiple-plate scheme allows to compress the output pulses from the KTA-based OPA (120 µJ, 120 fs, 3.5 µm) down to 21 fs (1.8-optical-cycle at 3.5 µm) with high total throughput of 38%. CEP preservation during this pulse compression is confirmed by a single-shot \( f \)-to-\( 2f \) interferometry. The output pulses from the multiple-plate compressor are used to produce CEP-dependent high harmonics in a MgO single crystal.

### 2 Experiment

The KTA-based OPA and the multiple-plate compressor are illustrated in Fig. 1(a) and (c), respectively. The output pulses from the OPA are focused to three plates (2-mm AR-coat Si, 2-mm non-coat YAG, and 1-mm AR-coat Si), all at the normal incident angle. The focal lengths, the number and the thicknesses of plates, and the distances between adjacent plates are empirically optimized using off-the-shelf optics to achieve spectral broadening without deterioration of beam quality and drastic decrease of transmitted energy. Preservation of excellent beam quality with a throughput of 83.5% including the Fresnel losses is achieved by placing a YAG plate near the focus, Si plates several centimeters before and after the focus as shown in Fig. 1(c). Figure 2(a) shows the result of the spectral broadening when one (YAG alone), two (Si, YAG) and three (Si, YAG, Si) plates are inserted. The single YAG plate broadens the spectrum efficiently as reported in [7]. Further spectral broadenings are observed by adding two Si plates, resulting in the octave-spanning supercontinuum from 2.2 µm to 4.9 µm at the -20 dB intensity level, while the center wavelength is maintained to be 3.5 µm. After the spectral broadening, we select out a spectrally uniform part of the transmitted beam by a diaphragm with a transmission of 45%, resulting in 38% total throughput in the compressor. The final output pulses after the three plates acquire only a small amount of positive chirp due to nonlinear self-compression mechanism in a YAG plate [7]. Therefore, to compensate the residual dispersion, we insert 4-mm propagation in CaF\(_2\) to compress the output pulses down to 21-fs duration by a single-shot \( f \)-to-\( 2f \) interferometry, resulting in a CEP stability of 283 mrad (rms) for 500 seconds. To demonstrate the capability in a strong-field experiment, we conduct HHG by...
loosely focusing the sub-two-cycle, 45-μJ pulses at 3.5 μm (~0.35 eV) onto a 200-μm-thick MgO single crystal with different CEPs by changing insertion of CaF₂ wedges. High harmonics up to 7th (~2.5 eV) are easily obtained, and their CEP dependence is clearly seen as shown in Fig. 3. This result confirms the capability of our sub-two-cycle mid-IR source to investigate field-driven sub-cycle carrier dynamics in solids.

3 Conclusion

We demonstrate a compact and robust multiple-plate pulse compression scheme of sub-mJ, femtosecond mid-IR pulses from a KTA-based OPA. Octave-spanning optical pulses from 2.2 μm to 4.9 μm at the -20 dB intensity level are generated by spectral broadening in YAG and Si plates. Combined with self-compression in the YAG plate, the residual dispersion is further compensated by CaF₂ plates down to 21 fs (1.8 optical cycle). The compressed pulses are focused onto an MgO single crystal to produce high harmonics up to 7th (~ 2.5 eV) with clear CEP dependence. Our demonstrated multiple-plate pulse compression allows to access CEP-stable, few-cycle, intense mid-IR pulses that are indispensable to explore attosecond electronic dynamics.

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