A CEP-stable, femtosecond 8.5 μm source based on intrapulse DFG of 2.1 μm pulses

Ondřej Novák¹, Peter R. Krogen², Tobias Kroh², Tomáš Mocek¹,
Franz X. Kärtner^{2,3,4}, and Kyung-Han Hong²

1. HiLASE Centre, Institute of Physics AS CR, Za Radnici 828, 252 41 Dolní Brezany, Czech Republic

2. Department of Electrical Engineering and Computer Science and Research Laboratory of Electronics, Massachusetts Institute of Technology (MIT), Cambridge, Massachusetts 02139, USA

3. Center for Free-Electron Laser Science and Deutsches Elektronen-Synchrotron (DESY), Notkestraße 85, 22607 Hamburg, Germany

4. Department of Physics and Center for Ultrafast Imaging, University of Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany

Author e-mail address: kyunghan@mit.edu

Few-cycle carrier-envelope phase (CEP) stable laser pulses enable the precise control of strong-field electron dynamics, such as high-harmonic generation in gases and solids or electron emission in nano-structures [1]. The intrapulse difference-frequency generation (DFG) pumped by a broadband pulse is a reliable method of producing passively CEP-stable pulses. Mid-infrared (mid-IR) pulse generation via intrapulse DFG has been demonstrated with near-IR pump pulses. For example, the intrapulse DFG of Ti:sapphire laser pulses can generate $\sim 2\ \mu\text{m}$ [2] or $\sim 5\ \mu\text{m}$ pulses [3], while spectrally broadened and compressed pulses of Yb:YAG laser were used to cover the region from 6.8 to 16.4 μm [4]. Since the efficiency and output pulse energy is rather low for intrapulse DFG, these pulses typically seed further optical parametric amplifiers. If the pump wavelength is shifted to $\sim 2\ \mu\text{m}$ region, the mid-IR intrapulse DFG at $>5\ \mu\text{m}$ becomes more efficient due to a lower quantum defect and the excellent phase matching characteristics of mid-IR nonlinear crystals based on non-oxide materials.

Here, we demonstrate intrapulse DFG in an AgGaSe₂ (AGSe) crystal pumped by a few-cycle 2 μm beam. We believe this is the first intrapulse DFG with a $\sim 2\ \mu\text{m}$ beam. The pump pulses stem from a kHz, 26 fs, 2.1 μm OPCPA [2]. Figs. 1(a) and (b) show the optical spectrum and temporal profile, respectively. An energy of 250 μJ was used to pump a 2-mm-long AGSe crystal. The estimated pump peak intensity was $\sim 300\ \text{GW}/\text{cm}^2$. Polarization of the beam was adjusted using a waveplate mainly to maximize the type II phase-matching (Fig. 1(c)).

Fig. 1 (a) Spectrum and (b) temporal profile of the 2.1 μm beam measured by FROG. The black dotted line in (a) is independently measured spectrum and the red dotted line in (b) is the transform-limited pulse shape. (c) Phase matching curve of type II AGSe with $\theta = 56.5^\circ$.

We obtained the mid-IR pulses of $\sim 2\ \mu\text{J}$ pulse energy with $\sim 1\%$ conversion efficiency. The measured spectrum (Fig. 2(a)) covers the range of 7–11 μm with a peak at $\sim 8.5\ \mu\text{m}$. The intrapulse DFG process is supported by numerical calculations [5], as shown by Fig. 2(b). The pulse duration of $\sim 85\ \text{fs}$ (FWHM) or 3 cycles is expected (Fig. 2(c)). The CEP stability can be measured using cross-referencing spectral interferometry to the CEP-stable 2.1 μm pump pulse. This pulse is particularly useful for strong-field experiments in nanostructures as well as seeding OPCPA/OPA in the long-wavelength IR.

Fig. 2 (a) The measured and (b) calculated spectra of the intrapulse DFG pulse. (c) Calculated temporal profile of 85 fs pulse.

References

- [1] W. Putnam, R. Hobbs, P. D. Keathley, K.K. Berggren, and F. Kärtner, "Optical-field-controlled photoemission from plasmonic nanoparticles," *Nat. Physics* (2016), doi:10.1038/nphys3978.
- [2] K.-H. Hong, C.-J. Lai, J.P. Siqueira, P. Krogen, J. Moses, C.-L. Chang, G.J. Stein, L.E. Zapata, and F.X. Kärtner, "Multi-mJ, kHz, 2.1 μm optical parametric chirped-pulse amplifier and high-flux soft x-ray high harmonic generation," *Opt. Lett.* **39**, 3145 (2014).
- [3] T. Zentgraf, R. Huber, N.C. Nielsen, D.S. Chemla, and R.A. Kaindl, "Ultrabroadband 50-130 THz pulses generated via phase-matched difference frequency mixing in LiIO₃," *Opt. Express* **15**, 5775 (2007).
- [4] I. Pupeza, *et al.*, "High-power sub-two-cycle mid-infrared pulses at 100 MHz repetition rate," *Nat. Photon.* **9**, 721 (2015).
- [5] T. Lang, *et al.*, "Impact of temporal, spatial and cascaded effects on the pulse formation in ultra-broadband parametric amplifiers," *Opt. Express* **21**, 949 (2013).