



PhD Thesis Defense: High-peak-power mid-infrared OPCPAs for extreme nonlinear photonics

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May 03, 2022

10:00

ICFO Auditorium

In the last decades, intense carrier-envelope-phase-stable (CEP-stable) and near-single-cycle, coherent mid-infrared sources have become charming for a variety of applications in physics, chemistry and biology. In particular, those mid-infrared sources are of tremendous interest for broadband spectroscopic applications, solid-state light-matter studies, strong-field physics research, and attosecond science. On the one hand, broadband coherent mid-infrared sources are capable of replacing time-consuming scanning techniques to classify organic structures or detect hazardous chemical compounds. On the other hand, high-energy, CEP-stable, near single-cycle mid-infrared sources are key in strong-field physics and attoscience due to the wavelength scaling nature of strong-field electron re-collision-based processes.

Nevertheless, implementing such mid-infrared sources remains challenging due to the lack of user-friendly temporal, spectral and spatial characterisation instruments, efficient and affordable reflection/transmission coatings, and commercially accessible low-loss dispersion compensation optics. Moreover, the absence of suitable laser gain materials reinforces nonlinear down conversion and amplification methods. One approach to overcoming the current limitations and developing intense ultrafast mid-infrared systems is to use a commercially available high-power near-infrared laser combined with second-order nonlinear processes such as the optical parametric amplification (OPA) process or the optical parametric chirped-pulse amplification (OPCPA) process. OPCPA can be essential to avoid damage to the nonlinear crystals or tailor the amplified spectrum. OPCPAs are also used when femtosecond pulses are required to be amplified using picosecond pump lasers. As a result, OPCPA systems offer novel opportunities for producing high-intensity, broadband mid-infrared femtosecond pulses.

Here the 160 kHz high-power mid-infrared OPCPA system is developed to overcome the existing limitations in the high-repetition-rate mid-infrared regime. This thesis demonstrates the generation of unique 3.2 μm pulses with a single-cycle duration and delivering up to 3.9 GW of peak power. The combination of the CEP stability with the single-cycle duration and the high energies demonstrated makes this system suitable to produce ultrafast radiation in the kilo-electron-volt X-ray regime.

A newly developed mid-infrared nonlinear crystal named BGGSe is proposed for efficient broadband infrared radiation generation. The ultra-broadband source is produced using the BGGSe crystal combined with a unique anti-resonant-reflection photonic crystal fibre (ARR-PCF) that enables tailoring the compression of our 3.2 μm pulses at 160 kHz. Using the BGGSe crystal and the ARR-PCF, we demonstrate the generation of coherent light expanding up to seven octaves, from UV to the THz regime.

The second mid-infrared system presented in this thesis is the high-energy 7 μm OPCPA operated at a 100 Hz repetition rate and developed to generate hard X-rays in the multi-kilo-electron-volt regime. The development of this second OPCPA centred at 7 μm overcomes the considerable challenges in the mid-infrared regime. This thesis demonstrates the amplification of those mid-infrared pulses to 750 μJ and the efficient back-compression to 188 fs.

Moreover, high harmonic generation in solids driven by 7 μm pulses at 100 Hz and 3.2 μm pulses at 160 kHz has been exploited for solid-state studies using the developed OPCPA systems. This thesis highlights the results achieved in the high-temperature YBCO superconductor, where exponential enhancement of harmonics is demonstrated below the critical temperature.

All these demonstrations make those systems a key-enabling technology for the next generation of studies in solid-state physics, extreme nonlinear photonics, strong-field physics and coherent X-ray science.

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Hosted by: Prof. Dr. Jens Biegert