Octave-spanning Cr:ZnS femtosecond laser with intrinsic nonlinear interferometry

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We report a few-cycle, super-octave, polycrystalline Cr:ZnS laser system with 4 W power at 78 MHz repetition rate, where all of the necessary optical signals for the measurement of the carrier–envelope offset frequency are generated intrinsically. © 2019 Optical Society of America under the terms of the OSA Open Access Publishing Agreement

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Development of new ultrafast sources in different parts of the optical spectrum remains a key topic of laser science and technology. Middle IR (MIR) femtosecond (fs) lasers in the spectral range 2–20 μm enable a host of important applications ranging from fingerprinting of molecules [1] to high harmonic generation in the x-ray regime [2]. There are numerous, both more and less established, techniques for MIR fs lasing and frequency comb generation [3]. Ultrafast sources based on Cr:ZnS and Cr:ZnSe are appealing because they allow direct generation and amplification of few-cycle pulses in the range of 2–3 μm [4].

Advanced applications of MIR fs lasers require control of the pulse repetition and carrier–envelope offset frequencies (fR, fCEO) [5]. The common approach to fCEO measurement relies on broadening the spectra of fs pulses to an octave (typically in specially designed fibers or waveguides) followed by nonlinear interferometry (which possesses common drawbacks of its linear counterpart: alignment sensitivity and susceptibility to external noise). To date, the generation of octave spanning fs pulses at high fR in bulk materials, which can be combined with direct fCEO detection schemes, has been limited to a few special cases of Ti:sapphire oscillators [6]. Here we describe a fs source at 2.5 μm central wavelength utilizing a Cr:ZnS amplifier simultaneously enabling amplification of fs pulses, nonlinear broadening to an octave, and generation of optical harmonics—all in a single pass through a 9 mm polycrystalline medium. We believe this to be the first report of a fs laser with intrinsic nonlinear interferometry.

ZnS is a wide-bandgap semiconductor with high χ(2), χ(3), and remarkable transparency. Cr:ZnS is available as a polycrystal that features random quasi-phase-matching (RQPM). Although inefficient, RQPM is well suited for three-wave mixing of fs pulses with broad spectra. Thus, the propagation of few-cycle pulses with multi-MW peak power through the Cr:ZnS polycrystal is simultaneously influenced by a number of nonlinear effects: laser interactions are combined with three-wave mixing between the pulse’s spectral components, and the conventional nonlinear index (n2 ≈ 10−15 cm2/W) is augmented by an effective index n2,CON that arises from cascaded quadratic nonlinearities [7].

Over the past several years, we have reported on incremental progress in the development of polycrystalline Cr:ZnS lasers and amplifiers [4]. Here we present the first octave-spanning system, to the best of our knowledge. We demonstrate, in a simple experiment, that this last step has enabled an important threshold: it has given us the ability to detect the fCEO of a mode-locked laser as easily and reliably as its fR.

The laser source is illustrated in Fig. 1. It is arranged as a full repetition rate single-pass fs amplifier [4]. The prerequisites for generation of an octave-spanning spectrum include: the use of a sub-octave 3-cycle master oscillator with ≈0.5 MW peak power [4], optimization of chromatic dispersion at the input of the amplifier, and optimization of the transverse size of the input beam. Obtained laser parameters are summarized in Fig. 2. The spectrum of pulses in Fig. 2(a) corresponds to 16.8 W cw erbium-doped fiber laser (EDFL) pump and 4.1 W average power of output pulses measured within the fundamental (f) MIR band at 2–3 μm. RQPM in the polycrystalline Cr:ZnS provides conversion of an octave-spanning MIR spectrum (1.6–3.2 μm) to a continuum consisting of optical harmonics from second (2f) to fourth (4f). We measured 0.3 W power in 2f band, ~10 mW power in 3f band, and ~0.1 mW of visible 4f signal.

We used a custom autocorrelator (provided by APE GmbH) with two-photon-absorption detection and relied on its control software for evaluations of the pulse duration. We separated f and 2f bands by a set of six dichroic mirrors and installed a 2 mm YAG plate for output dispersion control. The second-order interferometric autocorrelation (AC) in Fig. 2(b) corresponds to 19 fs pulses (sech2 fit) at an intermediate average power of 3.5 W. The residual 3f signal was too strong to measure AC at full 4.1 W MIR power. However, the obtained spectrum supports 16 fs (two-cycle) pulses.
APD430A2 detector is illustrated in Fig. 3(b). We then replaced this improvised prism monochromator and Thorlabs... 633 nm is separated by a band-pass filter with 3 nm bandwidth.

Fig. 2. (a) Overall spectrum obtained by the stitching of seven spectra (shown by different colors). The spectra were acquired with SP2500 and SP2150 Acton monochromators using appropriate gratings, photodetectors, and spectral filters. Error bars around the MIR spectra correspond to averaging of three sets of data acquired during several hours. (b) Autocorrelation (AC) of MIR pulses. Ripples in the AC are due to 3f background; see main text. (c) Photo of the Cr:ZnS gain element and output MIR beam profile at 4.1 W power.

The nonlinear propagation of super-octave MIR fs pulses through polycrystalline Cr:ZnS can be interpreted as a single-pass nonlinear interferometry. Such an interferometer occupies a microscopic volume inside a bulk material, is insensitive to vibrations and other fluctuations of the environment, and does not require any alignment. The photo in Fig. 3(a) shows 2f, 3f, and 4f bands of the spectrum dispersed by a prism. We steered this dispersed spectrum to a Si avalanche detector and observed heterodyne beat signal at 632.8 nm with a signal-to-noise ratio of 40 dB in a 78 MHz bandwidth. Interference beats at 633 nm are used as a reference of the entire system.

Fig. 3. (a) Photo of the visible-to-near IR part of the spectrum dispersed on a screen by a BK7 prism. Color letters show locations of the optical harmonics of the fundamental MIR. Vertical arrows show regions of spectral overlap (RSO) between the adjacent harmonics that were used to detect the interference beats at fCEO. (b) RF spectrum of the 2f-3f beating; RSO at about 875 nm is separated by a prism. (c) RF spectrum of the 3f-4f beating; RSO at 633 nm is separated by a band-pass filter with 3 nm bandwidth.

In conclusion, we have implemented a 4 W super-octave few-cycle source in the MIR range of 2–3 μm. We conservatively estimate 19 fs pulses with 2.5 MW peak power at fR = 78 MHz. Generation of optical harmonics within the laser medium provided the means for direct and robust measurement of the laser’s fCEO. To the best of our knowledge, this is the first super-octave MIR spectrum generated in a bulk material at full repetition rate of a mode-locked oscillator, and the first fCEO measurement for a Cr³⁺-based laser.

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