



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

SERGEY VASILYEV,¹ IGOR MOSKALEV,¹ VIKTOR SMOLSKI,¹ JEREMY PEPPERS,¹ MIKE MIROV,¹ VLADIMIR FEDOROV,^{1,3} DMITRY MARTYSHKIN,^{1,3} SERGEY MIROV,^{1,3,4,*} AND VALENTIN GAPONTSEV²

¹IPG Photonics—Southeast Technology Center, 100 Lucerne Ln., Birmingham, Alabama 35211, USA

²IPG Photonics Corporation, 50 Old Webster Rd, Oxford, Massachusetts 01540, USA

³Department of Physics, University of Alabama at Birmingham, 1530 3rd Ave S, Birmingham, Alabama 35294, USA

⁴e-mail: mirov@uab.edu

*Corresponding author: smirov@ipgphotonics.com

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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 (f_R , f_{CEO}) [5]. The common approach to f_{CEO} 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 f_R in bulk materials, which can be combined with direct f_{CEO} 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 $\chi^{(2)}$, $\chi^{(3)}$, 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 ($n_2 \approx 10^{-15} \text{ cm}^2/\text{W}$) is augmented by an effective index $n_{2,\text{CQN}}$ 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 f_{CEO} of a mode-locked laser as easily and reliably as its f_R .

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 at -40 dB) 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.

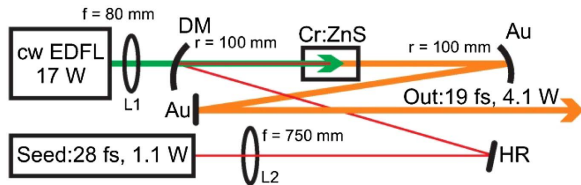


Fig. 1. Laser setup. EDFL pump beam is focused in 9 mm Cr:ZnS sample with a lens L1 ($\approx 60 \mu\text{m}$ $1/e^2$ intensity diameter). Seed pulses are pre-shaped with a lens L2, pre-chirped with GDD $\approx -800 \text{ fs}^2$, superimposed with the pump beam and focused with a curved mirror DM into $\approx 170 \mu\text{m}$ spot. Output pulses are collimated and steered with gold-coated mirrors (Au). The setup is enclosed in a dust-tight case and purged with dry air. The seed laser is equipped with a low-noise single-frequency EDFL pump.

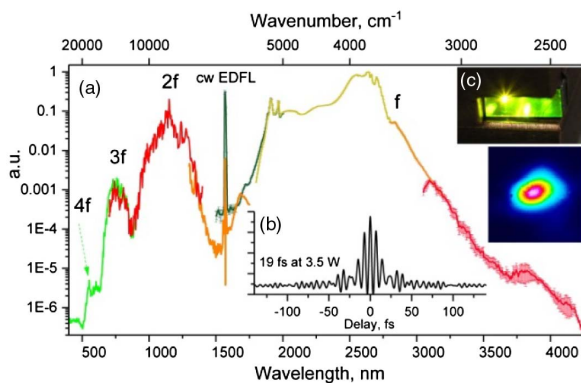


Fig. 2. (a) Overall spectrum obtained by the stitching of seven spectra (shown by different colors). The spectra were acquired with SP2500 and SP2150 Acton monochromators using appropriate gratings, photodetectors, and spectral filters. Error bars around the MIR spectra correspond to averaging of three 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 beatings in the regions of spectral overlap between the optical harmonics. The $2f$ - $3f$ interference beat signal measured with this improvised prism monochromator and Thorlabs APD430A2 detector is illustrated in Fig. 3(b). We then replaced the prism with off-the-shelf optical filters and detected the $3f$ - $4f$ beat signal at 632.8 nm with a signal-to-noise ratio of 40 dB in a 10 kHz resolution bandwidth [Fig. 3(c)]. The f - $2f$ beat signal has been measured in an additional experiment and will be reported separately. As expected, f_{CEO} can be controlled by modulation of the oscillator's pump power. We measured a response of 17 kHz/mW (3%/mW in relative terms) during slow variation of the pump power. We also observed that the reduction of

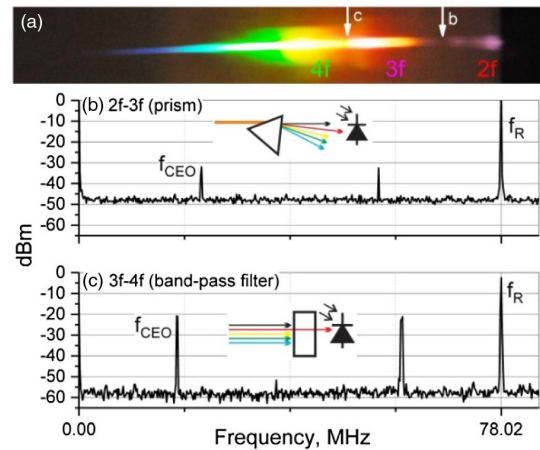


Fig. 3. (a) Photo of the visible-to-near IR part of the spectrum dispersed on a screen by a BK7 prism. Color letters show locations of the optical harmonics of the fundamental MIR. Vertical arrows show regions of spectral overlap (RSO) between the adjacent harmonics that were used to detect the interference beats at f_{CEO} . (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.

relative humidity inside the laser case from 45% to 0% resulted in ~ 30 MHz variation of the f_{CEO} .

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 $f_R = 78$ MHz. Generation of optical harmonics within the laser medium provided the means for direct and robust measurement of the laser's f_{CEO} . 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 f_{CEO} measurement for a Cr^{2+} -based laser.

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