Efficient femtosecond mid-infrared generation based on a Cr:ZnS oscillator and step-index fluoride fibers

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Abstract

Femtosecond light sources in the 3–5 μm region are highly sought after for numerous applications. While they can be generated by using nonlinear effects in optical fibers, the efficiencies and effectiveness of frequency conversion can be significantly enhanced by using ultrashort driving pulses. Here, we report on a few-cycle Cr:ZnS oscillator driving low-order soliton dynamics in soft-glass fibers. By selecting appropriate parameters, sub-two-cycle pulses or broad supercontinua spanning over 1.7 octaves from 1.6 to 5.1 μm can be generated at average power levels exceeding 300 mW. In the same setting, Raman-induced soliton self-frequency shifting has been exploited to generate sub-100-fs pulses continuously tunable from 2.3 to 3.85 μm with a conversion efficiency of ∼50%. These results demonstrate the vast potential of using Cr:ZnS or Cr:ZnSe lasers for powerful mid-infrared generation.

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Lasers emitting in the mid-infrared (MIR) spectral range between 3–5 μm have been attracting increasing attention in recent years, driven by the vast number of spectroscopic applications that can benefit from their coherent properties [1,2]. Among others, time- and frequency-domain techniques are used to gain specific information about molecular compositions and dynamics [3]. However, most of these approaches require laser sources capable of delivering broadband, femtosecond light pulses. As there is very limited choice in the MIR region, complex nonlinear frequency conversion schemes based on optical parametric oscillators and amplifiers (OPOs and OPAs) are usually used.

A promising alternative is given by fibers based on soft glasses such as fluoride, chalcogenide, and telluride. They can support different nonlinear processes in the MIR range, including supercontinuum (SC) generation, soliton self-compression, and Raman-induced soliton self-frequency shifting (SSFS). SC generation can convert femtosecond pump light into broadband infrared radiation [4–8]. Self-compression provides near Fourier-transform-limited pulses without requiring further dispersion compensation. On the other hand, Raman-induced shifting results in broadly tunable femtosecond pulses and has recently been demonstrated using two fiber stages to reach 4.3 μm [9]. Higher average powers of approximately 2 W have also been achieved with further amplification [10]. However, the effectiveness of the various nonlinear effects can be dramatically enhanced by using shorter initial pulses. For example, the extent of frequency shifts for SSFS is inversely proportional to the 4th power of the input pulse duration. Furthermore, a shorter pulse duration also leads to a reduction in soliton order for a given fiber dispersion, nonlinearity, and input peak power. This provides compressed pulses with less pedestal, as well as supercontinua with higher spectral coherence [11]. Using the few-cycle pulses directly available from Cr:ZnS/ZnSe oscillators [12–14] will thus provide significant advantages over existing femtosecond pump sources such as Er/Tm fiber lasers [15,16] and optical parametric amplifiers [17].

The use of Cr:ZnS/ZnSe lasers to drive frequency conversion in soft-glass fibers was first demonstrated under normal fiber dispersion [18], leading to a spectral broadening factor of approximately two and a spectral bandwidth spanning 800 nm at the −20 dB level. Apart from fibers, a Cr:ZnS oscillator has also been used to drive SC generation in a silica-based double-nanospike waveguide, producing an ultra-broadband output spanning over one octave at ∼50 pJ pulse energies [19].

In this Letter, we further explore the potential of Cr:ZnS lasers as pump sources for nonlinear frequency conversion in ZBLAN fibers, which can be conveniently pumped by 2 μm lasers and are widely available. With the higher output power and shorter pulse durations generated by our oscillator, we demonstrate a spectral broadening factor of up to six, and achieve a spectral bandwidth of 3500 nm centered at 3.35 μm. Soliton self-compression down to 15 fs is also demonstrated, along with the efficient generation of wavelength-tunable femtosecond pulses via Raman-induced SSFS.
The experiment consisted of a home-built femtosecond Cr:ZnS oscillator [Fig. 1(a)] with its output coupled into a ZBLAN fiber [Fig. 1(b)]. The oscillator contained a 9 mm long rectangular anti-reflection (AR)-coated Cr:ZnS polycrystalline gain element (IPG Photonics) mounted on top of a water-cooled copper heat sink. It was placed inside an asymmetric X-fold cavity at a normal-incidence angle, similar to Ref. [13], to reduce asymmetric thermal lensing effects [20]. The oscillator was pumped at 1908 nm with a single-mode Tm-doped fiber laser (TDFL) (IPG Photonics, TLR-120), with up to 7.1 W focused into the crystal. Mode locking was achieved via the Kerr-lens effect in the gain element and by using a hard aperture (HA). With dispersive mirrors produced in-house [21], both the group delay dispersion (GDD) and third-order dispersion (TOD) were carefully adjusted, allowing for soliton mode locking in the anomalous dispersion regime. The oscillator provided up to 1 W of average power in the fundamental mode with a maximum pulse energy of 15 nJ at a repetition rate of 68.7 MHz. With the dispersion of the 6.35 mm thick fused-silica output coupler (OC) substrate compensated, the oscillator delivered 46 fs pulses. Optimizing the oscillator’s dispersion further, even shorter pulse durations can be expected, considering the recent demonstration of spectra supporting 15–20 fs from a Cr:ZnS oscillator [22].

The pulses were then directed towards the fiber setup, where ZBLAN (ZrF4, BaF2, LaF3, AlF3, NaF) fibers from two different companies were tested. Among other soft glasses, fluoride fibers are the most mature and readily available. They also provide a matching transmission window, nonlinearity and, most importantly, favorable dispersion characteristics for the targeted wavelength [23]. When the core diameters are around 6 μm, these fibers exhibit very slight anomalous dispersion on the order of ~100 fs²/cm near the pump wavelength of 2.3 μm. Given the peak power available for the experiment, this magnitude of dispersion is essential for the formation of low-order solitons (soliton order N < 10) required for the clean soliton compression and efficient Raman SSFS aimed for in this Letter.

To increase the coupling efficiency into the fiber, the output beam diameter of the home-built oscillator was reduced to approximately 3.5 mm by a telescope (L2, L3). A coupling efficiency of 50–60%, limited mainly by the imperfect mode matching, was achieved using an AR-coated aspheric lens (L4).

To account for the fiber’s weak birefringence arising from the fiber core’s ellipticity and mounting stress, a broadband half-wave plate (HWP) was placed in the beam path to rotate the input’s initially vertical polarization and match one of the fiber’s birefringence axes. However, the resulting changes in output spectra were insignificant. Finally, by means of a self-built second-harmonic frequency-resolved optical gating device (SHG-FROG), we confirmed that the input pulse durations after the lens L4 were still relatively short (τp = 50 fs) at a maximum available input power of 730 mW.

The power into the fibers was adjusted using an iris. In addition, the fiber facets were angle cleaved to avoid back reflections into the oscillator, with the fiber stripped of its protective coating around the input and output facets to avoid absorption of stray light and subsequent damage. No major degradation was observed over multiple weeks. For our experiments, three ZBLAN fibers with different core diameters were tested for demonstrating different pulse propagation dynamics in the anomalous dispersion regime. Two fibers from FiberLabs had a core diameter of 6.3(±0.1) and 6.8(±0.1) μm. In addition, a fiber with an intermediate core diameter of 6.5 μm was provided by Le Verre Fluoré.

Solitonic effects are highly dependent on the pulse’s and fiber’s parameters. To understand and predict the effects of soliton self-compression and SSFS, numerical simulations were performed. Accurate data on the refractive index of the core and cladding of ZBLAN fibers were provided by FiberLabs, which allowed the dispersion characteristics of the fiber’s fundamental propagation mode to be calculated [24]. The results were fed into a commercially available pulse propagation simulator (fiberdesk) that solves the nonlinear Schrödinger equation by the split-step Fourier method, taking into account the higher-order dispersion, Raman effect, and self-steepening. The nonlinear refractive index of ZBLAN glass provided by FiberLabs was n2 = 4.7 × 10⁻¹⁰ m²/W, and the Raman gain was modeled using the parameters rs = 16.67 fs, rR = 20.32 fs, and f R = 0.30 [25]. In Table 1, a short summary of experimental parameters of our ZBLAN fibers is given, including the calculated second-order dispersion coefficient at 2.3 μm. Since Le Verre Fluoré did not provide the corresponding refractive index information, the dispersion of their 6.5 μm fiber was modeled using FiberLabs data. In the following, the results obtained with the three ZBLAN fibers are presented and analyzed.

We started with the 2 m long 6.8 μm fiber, which had a mode-field diameter (MFD) of 7.7 μm. As can be seen in Fig. 2, the zero-dispersion wavelength is located at 1769 nm, resulting in slight anomalous dispersion of ~109 fs²/cm at 2.3 μm. As the ZBLAN fibers exhibit very low attenuation—less than 60 dB/km for wavelengths up to 4.0 μm—the measured output powers were assumed to be equal to the launched powers. The output spectra of the fiber were recorded with a Fourier-transform infrared (FTIR) spectrometer (Bristol Instr., 771B-IR) [Fig. 3(a)] at increasing input powers, revealing SSFS.

![Fig. 1. Setup. (a) Cr:ZnS oscillator, containing GDD mirrors (CM, ~200 fs²/bounce), TOD mirrors (TM, ~3200 fs²/bounce), curved dichroic mirrors (DM, ROC = ~100 mm), and an output coupler (OC, 32%), TDFL, Tm-doped fiber laser; L1 (f = 75 mm), plano-convex lens; HA, hard aperture. The round-trip GDD is ~500 fs² at 2.3 μm. (b) Fiber spectral broadening setup. A 5 mm thick AR-coated ZnSe substrate and TM-mirrors were added to compensate for the OC’s dispersion. The beam diameter was reduced by two lenses (L2, f = 100 mm; L3, f = 40 mm). HWP, half-wave plate; L4 (f = 5.95 mm, NA = 0.56), black diamond-2 aspheric lens; L5 (f = 7 mm), parabolic mirror.](image-url)
effects shifting the spectral components towards the longer wavelengths. The central wavelengths of the Raman-shifted pulse are in general agreement with the simulated results (in brackets): 3.85 μm (3.85 μm) at 380 mW, 3.67 μm (3.61 μm) at 300 mW, 3.22 μm (3.51 μm) at 260 mW, 3.09 μm (3.43 μm) at 220 mW, and 3.01 μm (3.36 μm) at 180 mW. The dynamics can be modeled even more accurately by taking into account the variation of the effective mode area at different wavelengths [9].

To determine the power content of the Raman-shifted solitons in the MIR, a long-pass (LP) filter with a cut-on wavelength of 2.8 μm was used. The measured MIR power at various total input power levels is 165, 136, and 112 mW for 380, 300, and 260 mW, respectively. These results—taking into account the approximately 15% loss of the LP filter—indicate that 50% of the input power is converted into the soliton pulse. This represents more than two times higher efficiencies compared to other non-amplified SSFS schemes in the 3–5 μm range [9]. Longer wavelengths could potentially be reached by replacing the ZBLAN fiber with an InF₃ fiber, which is transparent up to 5.5 μm [9].

To verify the femtosecond nature of the output, SHG-FROG measurements were performed on the solitons shifted beyond 3 μm. A 140 μm thick GaSe crystal was used as the nonlinear medium for SHG due to its high nonlinearity and transmission in this wavelength range. The retrieved spectral and temporal intensities of the redshifted pulse, centered at 3.25 μm at a total input power of 260 mW, are plotted in Figs. 3(e) and 3(f), along with the corresponding FROG traces (FROG error: 0.0034). The measured pulse duration is 70 fs, consistent with the Fourier-transform limit of the spectrum (at ~30 dB). The spatial output beam profile shows the pulse being guided in the fundamental mode [Fig. 3(e)], and was recorded using a pyroelectric camera (Ophir Optronics, Pyrocam III) behind the 2.8 μm LP filter.

The next fiber tested had a core diameter of 6.5 μm and a MFD of 7.6 μm, whose stronger nonlinearity and lower dispersion give a stronger self-compression. A fiber length of 2.5 cm was used, matching the calculated soliton fission length [11] for the fiber and input pulse parameters. In Fig. 4, the spectral and temporal profiles of the shortest output pulse are plotted. We generated sub-two-cycle pulses with a spectrum extending from 1.4 to 3.5 μm (~30 dB) at an output power of 350 mW. A retrieved pulse duration of 15 fs (FROG error: 0.0058) shows over 63% of the pulse energy residing within the FWHM of the compressed pulse, in virtue of the low soliton order. The measured pulse was slightly chirped, since the pulse travelled through 1.3 meters of air from the fiber tip to the diagnostics. Additionally, the pulse spectrum overlapped with the water vapor absorption region around 2.7 μm. Based on the retrieved data, the duration can be recompressed down to the Fourier-transform limit of 10 fs by an additional ~35 fs². This pulse duration would be equivalent to a single-cycle pulse.

Another ZBLAN fiber from FiberLabs with a core diameter of 6.3 μm and a MFD of about 7.6 μm was also investigated. As can be seen in Fig. 2, the dispersion curve of this fiber approaches zero. Since the soliton order, which correlates with the extent of the spectral broadening, is inversely proportional to the square root of the dispersion magnitude [11], this fiber is ideal for generating an ultra-broadband SC, similar to that observed by Salem et al. [6]. Yet, the inverse relationship also

![Fig. 2](image-url) Calculated fiber dispersion for different core diameters. The dashed black line marks the central wavelength of the laser pulses.

![Fig. 3](image-url) Results with the 6.8 μm ZBLAN fiber. (a) Measured spectra of the Raman-shifted solitons at increasing input powers. (b) Simulated pulse propagation at 380 mW input power, with the shifted spectrum after 2 m placed on top for comparison. (c) and (d) Measured and retrieved FROG traces, respectively, at 260 mW of input power. (e) Spectral results. Inset: measured output beam profile of the soliton. (f) Temporal results.

![Fig. 4](image-url) Results with the 6.5 μm ZBLAN fiber. (a) and (b) Measured and retrieved FROG traces. (c) Results in the spectral domain. (d) Temporal profile of the pulse, showing a FWHM pulse duration of 15 fs and the corresponding Fourier-transform limit of 10 fs.
makes the output spectrum sensitive to slight variations in absolute dispersion when the magnitude of dispersion is small, as in this case. Furthermore, given the experimental uncertainty in the fiber’s core diameter ($\pm 0.1 \mu m$), there may be regions where the dispersion becomes positive in the actual fiber. Consequently, simulations for this particular fiber are only indicative, and the presence of possible zero-dispersion crossings means the generation of non-solitonic dispersive waves is also likely [11]. Nevertheless, a 1.7 octave-spanning spectrum extending from 1.6 to 5.1 $\mu m$ ($\pm 30$ dB level) at an average power of 350 mW was measured using the 6.5 cm long fiber (Fig. 5). To the best of our knowledge, this is the broadest bandwidth generated in fibers and waveguides with Cr:ZnS/ZnSe pump sources [18,19]. The high output power and ultrashort pulse duration generated directly from the oscillator, together with the central wavelength of 2.3 $\mu m$, means a stronger spectral broadening towards the longer wavelengths can be achieved compared to typical thulium-fiber-based pump systems [6]. The newly generated frequency components measured behind a 2.8 $\mu m$ LP filter reached an average power of 47 mW. These power levels are already enough to saturate typical HgCdTe detectors or to generate a beat signal in a F2f interferometer for stabilizing the carrier-envelope phase of the Cr:ZnS oscillator. In addition, the stability of the newly generated spectral components was measured using a thermal power meter (Coherent, PS19) behind a 3.6 $\mu m$ LP filter. The root-mean-square (RMS) value of the power fluctuations turned out to be less than 0.4% over 1 h.

In summary, we have demonstrated the applicability of ultrashort few-cycle pulses from a Cr:ZnS oscillator as a new source for driving different nonlinear effects in the 3–5 $\mu m$ spectral region. By coupling these pulses into ZBLAN fibers, durations of 15 fs and, potentially, down to 10 fs (single-cycle) can be generated at 2.3 $\mu m$. In addition, a spectrum spanning from 1.6 to 5.1 $\mu m$ has been measured at over 300 mW of average power. This power level can be used in many applications such as nano-spectroscopy with a scattering-type scanning near-field optical microscope (s-SNOM) [26], or for time-domain spectroscopic techniques [3]. Taking advantage of the Raman SSFS effect, continuously shifting sub-100 fs pulses from 2.3 to 3.85 $\mu m$ wavelengths was also demonstrated at over twice the efficiencies compared to state-of-the-art non-amplified SSFS systems in the 3–5 $\mu m$ region [9,27]. By replacing the ZBLAN fibers with InF$_3$ fibers, whose transmission window extend beyond 5 $\mu m$, the SSFS can in principle be further shifted to the longer wavelengths [9,28]. Further experiments are planned with customized fibers that match the available laser parameters. While the output pulse duration of the Cr:ZnS laser used in the experiments presented here was 46 fs, even shorter pulse durations down to possibly 19 fs can be used [22], promising further improvement in the nonlinear frequency conversion. Finally, all of the presented nonlinear effects, despite their very different output characteristics, can be obtained by simply switching between fibers with different lengths or core diameters—a unique flexibility not available from other MIR generation systems. Thus, the combined system of Cr:ZnS/ZnSe lasers and soft-glass fibers has the potential to become the new workhorse in the 1.7–5 $\mu m$ range.

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**REFERENCES**