

1 Watt femtosecond mid-IR Cr:ZnS laser

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ABSTRACT

A room-temperature Kerr-Lens modelocked (KLM) Cr:ZnS laser generates <70 fs pulses duration (about eight optical cycles) with 5.6 nJ pulse energy and over 100 nm FWHM spectral width at 105-157 MHz repetition rates. The laser produces 1 W average output power at 20% optical efficiency, limited by the available Er: fiber pump. For further pulse energy scaling we also realized the chirped-pulse regime, with 0.8-2 ps pulse durations. The demonstrated applications of such mid-IR source range from extra- and intra-cavity spectroscopy to subharmonic OPO pumping. For environmentally-protected delivery we suggest and realize duration-preserving soliton delivery in a ZBLAN fiber. Further bandwidth increase is demonstrated by 2.0-2.8 μm supercontinuum generation in a chalcogenide fiber.

Keywords: Mid-infrared femtosecond oscillator, Mid-IR laser, Cr:ZnS, Cr:ZnSe lasers, solid-state lasers

1. INTRODUCTION

Femtosecond coherent light sources emitting in the mid-infrared are of particular interest for a number of applications like environmental sensing, medicine, metrology, material processing and telecommunications¹. These sources are usually complex and costly, because they are built on the basis of optical parametric amplification or difference frequency generation processes. The alternative approach is a mode-locked crystalline solid-state laser setup based on Cr²⁺-doped chalcogenide crystals, namely ZnSe, ZnS, CdSe, CdMnTe, CdS, etc.²⁻⁵ These crystals are nicely suitable for high-power femtosecond pulse generation due to their broad gain, continuous tunability over the wide wavelength range and good thermo-optical and thermo-mechanical properties⁶⁻¹⁰. Until the last few years the most developed crystal for solid-state femtosecond mid-IR lasers was Cr:ZnSe. Passively mode-locked femtosecond Cr²⁺:ZnSe laser was first reported in 2006¹¹ and the first KLM laser in 2009^{12,13}. To date, output power up to 300 mW¹², pulse energy up to 2.3 nJ¹⁴, pulse duration as short as 80 fs¹⁵ were demonstrated.

Alternative crystal of Cr:ZnS received a lot of attention in the last few years. Spectroscopically both crystals are in many respects similar, and the main difference is power handling capability, which is better for Cr:ZnS because of higher thermal conductivity, higher thermal shock parameter and lower thermal lensing parameter^{2,16}. But after the first demonstrations of continuous-wave Cr:ZnS laser in 2002 [12-14] the main attention was shifted to Cr:ZnSe, mostly because of its more reproducible growth quality combined with similar spectroscopic properties. Though Cr:ZnS is formally cubic, it can co-exist in several structure types, thus exhibiting natural birefringence. Subject to the availability of good-quality single and polycrystalline materials, the interest in Cr:ZnS raised again leading to the demonstration of picosecond mode-locked¹⁷ and 10 W polycrystalline continuous-wave laser¹⁸.

Femtosecond SESAM-initiated mode-locking using Cr:ZnS crystal was obtained recently¹⁹. Pulse duration of 130 fs was demonstrated at the wavelength around 2400 nm with a pulse repetition rate of 180 MHz. The average output power of 130 mW (205 mW in the chirped-pulse mode) was obtained corresponding to the pulse energy of about 0.7 nJ. It was shown that SESAM limits the performance of the system in terms of bandwidth, output power, and third order dispersion. As a next step, the Kerr-lens mode-locked Cr:ZnS laser was just realized demonstrating 69-fs pulses with the average output power of 550 mW^{20,21}. At the pulse repetition rate of 145 MHz this corresponds to the output pulse energy of 3.8 nJ.

In this paper we present the further development of Kerr-lens mode-locked Cr:ZnS laser. The optimization of the cavity parameters allowed to obtain 1W of average output power with a maximum pulse energy of 5.6 nJ. Chirped pulse Cr:ZnS oscillator was demonstrated in the KLM mode for the first time showing the potential of further energy scaling.

2. EXPERIMENTAL SETUP

The experimental setup is shown in Fig. 1. The classic astigmatically-compensated four-mirror cavity was used for the laser design. Cr:ZnS crystal with Cr^{2+} concentration of about $6 \cdot 10^{18} \text{ cm}^{-3}$ was grown by physical vapour transport technique. The active element with a thickness of 2.5 mm was mounted at Brewster angle on a copper heatsink without active cooling. The near-symmetrical cavity consisted of two folding concave high-reflector (HR) mirrors, a plane high-reflector chirped mirror (CM)^{15, 22}, and a plane output coupler (OC) with a transmission of 18% at the laser wavelength. The laser was pumped by the CW Er-fiber laser from IPG Photonics providing up to 5 W of polarized collimated output at 1.61 μm . The pump beam was focused onto the crystal by an AR-coated focusing lens (FL) with a focal length of 30 or 40 mm, depending on the cavity length. The crystal absorbed about 80% of the incident pump power. The mode-locking was achieved by the soft-aperture Kerr-Lens effect with a moving chirped mirror used as a starting mechanism. The compensation of the group-delay dispersion (GDD) was achieved by the combination of material dispersion (sapphire or YAG plate installed into the OC arm of the cavity) and the chirped mirror, thus making the resonator design especially compact and stable.

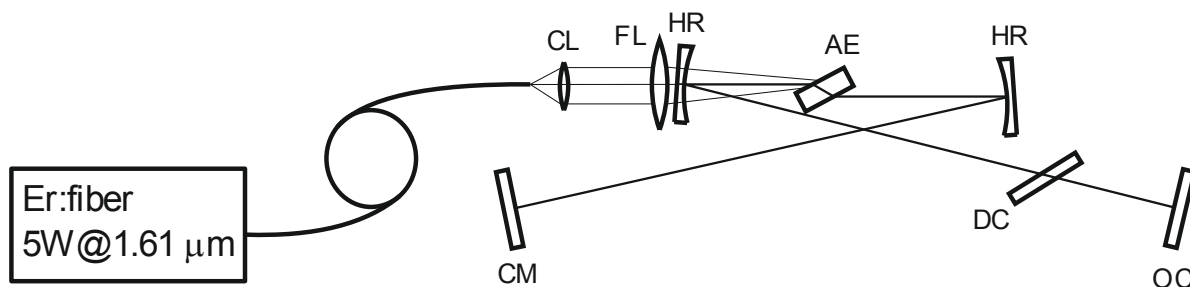


Figure 1. Schematic of the femtosecond Cr:ZnS KLM laser. CL – collimating lens, FL – focusing lens, HR – high reflector mirror, AE – Cr:ZnS active element, CM – chirped mirror, DC – dispersion compensation, OC – output coupler.

All measurements were performed in the open air with 40-50% relative humidity. The spectral distribution of the laser emission was analyzed by a Perkin-Elmer FTIR spectrometer, spectral resolution of 1 cm^{-1} was used in the most of the cases. For the measurements of the pulse duration a custom-made interferometric autocorrelator was built with a detection system based on a two-photon absorption in an amplified Ge photodetector. The pulse repetition rate was controlled by the picosecond GaAs photodetector working in two-photon absorption regime. The spatial distribution of the laser output was analyzed by the moving-slit beam profiler.

3. RESULTS AND DISCUSSION

3.1 Soliton regime

Kerr-lens mode-locked laser experiments were carried out with 1.43-m cavity having a fundamental repetition rate of 105 MHz. The maximum output power of 590 mW resulting in the pulse energy of 5.6 nJ and the minimum pulse duration of 74 fs were obtained with output coupler transmission of 18%. The interferometric autocorrelation trace of the laser pulse at maximum output power and laser beam profile are presented at Figure 2. The minimum pulse duration of 74 fs is equal to 9 optical cycles at this wavelength and corresponds to the pulse duration of 25 fs at the wavelength of Ti:sapphire laser. Laser pulses were close to transform-limited with the time-bandwidth product of 0.399. The beam profile (Fig. 2c) has a slight ellipticity.

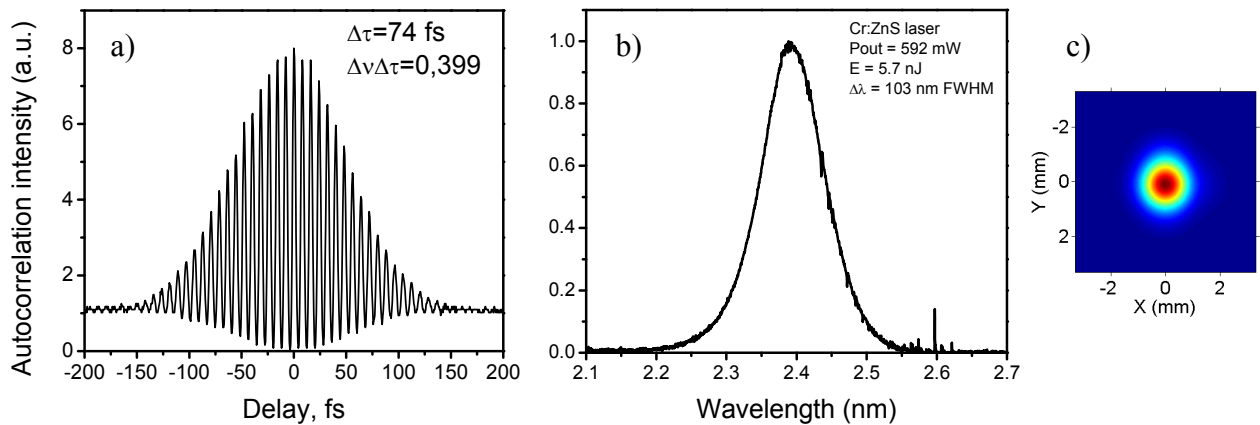


Figure 2. Interferometric autocorrelation trace of KLM Cr:ZnS laser (a), laser emission spectrum(b) and beam profile at the distance about 50 cm behind the output coupler (c).

The anomalous second-order dispersion of II-VI materials at wavelengths between 2 and 3 μm allows a simple method of material dispersion compensation^{11,14,15} to be used in chromium-doped chalcogenide mode-locked lasers. But for few-optical-cycle pulse generation management of the third-order dispersion is also critical. In the current setup both the material dispersion compensation and specially designed chirped mirror were used. Combination of methods allowed to obtain relatively flat GDD curve with total net GDD per cavity roundtrip about -450 fs² at central wavelength (Fig. 3).

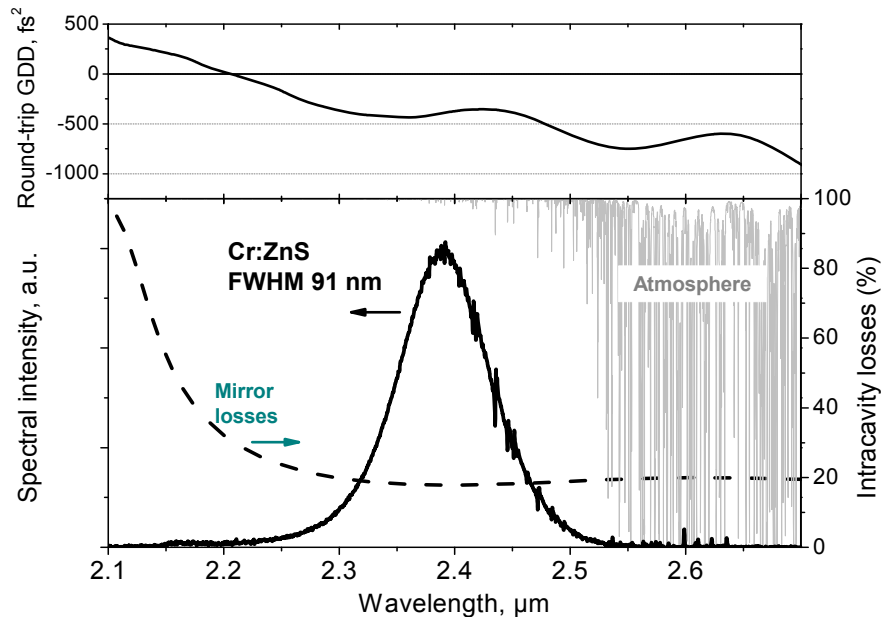


Figure 3. Output spectrum of a femtosecond Cr:ZnS laser (bold black), calculated round-trip GDD (solid black), intracavity losses due to the mirrors (dashed black). The atmospheric absorption (solid gray) is scaled to the resonator round-trip length and causes characteristic features on the spectrum²³. Data from Ref.²¹.

Elimination of the SESAM allowed shortening the pulses by about a factor of 2. Since the gain of Cr:ZnS can support few-optical-cycle pulses^{6, 15}, it is important to define the factors preventing the pulses from further shortening. The dispersion-like fractures on the spectra presented in Fig. 3 and other figures in the article are the traces of water vapor intracavity molecular absorption²³. The grey line in the Fig. 3 shows the intensity of these absorption lines in the spectral region 2.1-2.7 μm . It can be seen that water vapor absorption is one of the limiting factors for pulse shortening. Among

the other limiting factors one can consider cavity mirror losses (dashed black line in the Fig. 3) and intracavity third order dispersion causing the net GDD (solid black line in the Fig. 3) to become positive around 2.2 μm .

Another important aspect of the system is its power scalability. In the current setup the pulse energy was limited by the pulse breakup due to high third-order optical nonlinearity of the Cr:ZnS crystal ($90 \cdot 10^{-16} \text{ cm}^2/\text{W}$). Increasing the output power over 600 mW resulted in either double-pulsing with pulse separation of several picoseconds or harmonic mode-locking. At the maximum pump power of 5W the average output power reached 1W that is equal to laser optical efficiency of 20%. The pulse repetition rate was measured to be 210 MHz, twice as high as the cavity fundamental frequency, corresponding to the pulse energy of 4.7 nJ. The interferometric autocorrelation trace and pulse optical spectrum are plotted in Figure 4.

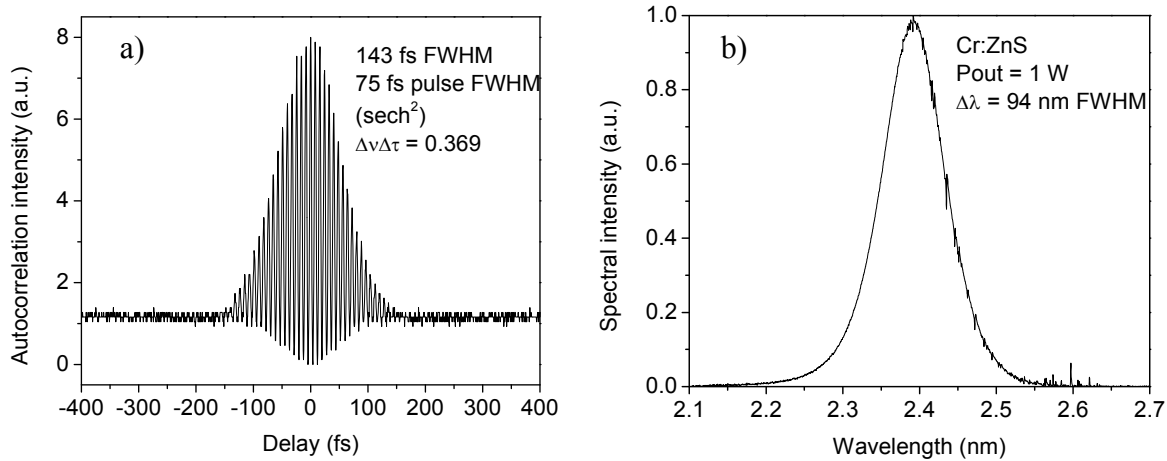


Figure 4. Interferometric autocorrelation trace (a) and optical spectrum (b) of KLM Cr:ZnS laser in harmonic mode-locking mode at the average output power of 1 W.

3.2 Chirped pulse (CPO) regime

For further increasing of the pulse energy of mode-locked $\text{Cr}^{2+}:\text{ZnS}$ laser one should reduce the peak power density inside the active medium. That could be done by using the technique of chirped-pulse-oscillator (CPO). This technique is well-established in the fiber²⁴ and Ti-sapphire lasers^{25,26}, and has recently been demonstrated with the Cr:YAG²⁷ and Yb-doped thin-disk lasers^{28,29}. For Cr:ZnSe the analytical theory³⁰ predicts pulse energies up to 0.5 μJ ³¹ and initial demonstration of CPO technique has already been performed for Cr:ZnSe as well as Cr:ZnS lasers¹⁹.

Practical realization of the chirped pulse regime has been performed by removing the bulk dispersion compensation elements from the cavity, bringing the net intracavity GDD to nearly zero. The experiments were carried out in 1-meter-length cavity with output coupler having 4.5 % transmission. The average laser output power could be scaled up to 730 mW that is equal to 17% slope efficiency. Despite the lack of the active cooling, the laser showed no evidence of thermal degradation demonstrating straight input-output curve in the whole available pump power range (Fig. 5a). The pulse durations are in the range of 0.8-2 ps, typical autocorrelation trace is plotted in the Fig. 5b. The spectra showed distinctive shape with steep edges reaching maximum width of 138 nm (Fig. 5c). Some asymmetry is due to the residual higher-order dispersion²⁷. The pulse energy slightly below 5 nJ was reached.

In the current configuration CPO oscillator has an important advantage over the SESAM-version¹⁹ that there is no energy leakage into the higher-order modes. Further pulse energy scaling is thus a subject of optimization of OC transmission, cavity length, available pump power, and active cooling.

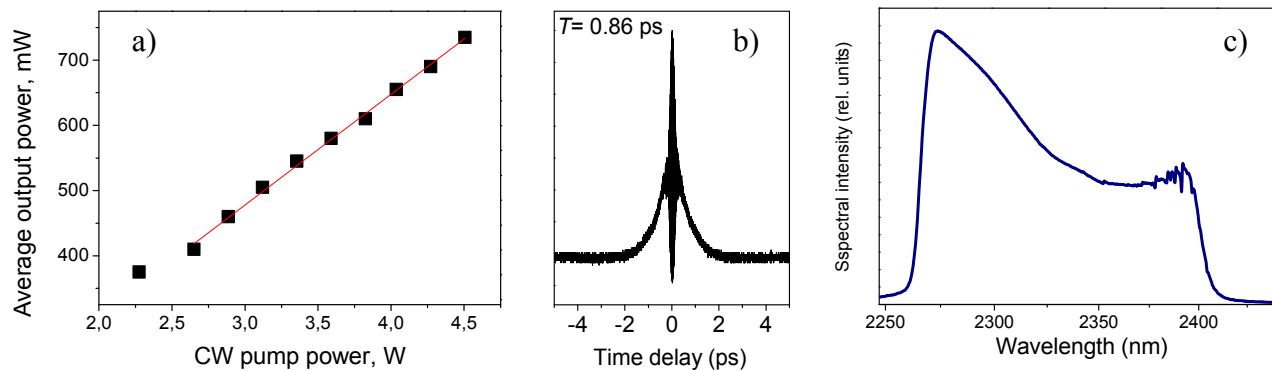


Figure 5. Input-output characteristics a), interferometric autocorrelation trace (b), and optical spectrum (c) of KLM Cr:ZnS laser in chirped pulse (CPO) regime. The noise-like features near 2390 nm originate from the atmospheric water absorption lines that are strongly enhanced at the spectrum edge³².

4. DEMONSTRATED APPLICATIONS

At this power and energy level, Cr:ZnS oscillator is now capable of number of important spectroscopic and nonlinear-optical applications. Already at 100-mW power range, high-sensitivity spectroscopy³³⁻³⁵ and wavelength conversion further to the infrared in subharmonic GaAs OPO³⁶ have been demonstrated. These applications will strongly benefit from the increased average power and mode quality. Important for applications is also the demonstrated possibility for environmentally-protected and duration-preserving delivery of the pulses via single-mode infrared fibers in solitonic regime³⁷. Further bandwidth increase can be achieved by supercontinuum generation in step-index chalcogenide fiber reaching 2.0-2.8 μm range³⁸.

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