Kerr-lens mode-locked Cr:ZnS laser

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We report the soft-aperture Kerr-lens mode-locked Cr:ZnS laser, generating 550 mW of 69-fs nearly transform-limited pulses at 2.39 μm wavelength. The pulse energy reached 3.8 nJ at 145 MHz repetition rate, limited by the onset of double-pulsing. This corresponds to the shortest-pulse and highest-energy direct femtosecond laser source in the mid-IR. Dispersion compensation was achieved by a single chirped mirror and a thin sapphire plate, making the laser design simple, compact and very stable, operating at ambient air and room temperature. The superb thermal and mechanical properties of Cr:ZnS, exceeding those of Cr:ZnSe and many established femtosecond laser crystals should allow further scaling of output power.

Femtosecond coherent light sources emitting in the "molecular fingerprint" mid-infrared (2 to 3 μm) spectral range are of great interest for a number of applications, first of all in environmental sensing, but also in medicine, telecommunications, material processing and metrology [1]. Such sources are mostly built on the basis of nonlinear optical conversion techniques, either optical parametric (OPO) or difference frequency generation, resulting in limited efficiency as well as high complexity and price of the system. The compact and cost-effective alternatives to the OPOs are the mode-locked crystalline solid-state lasers based on Cr^{2+}-doped chalcogenides [2-4]. Due to their broad gain and continuous tunability over a wide wavelength range (~1400 nm [5]), exceeding all known laser types, as well as high power (over 13 W in CW regime [4)], they are perfectly suitable for high power femtosecond pulse generation [6-10]. Today's most developed crystal for solid-state femtosecond mid-IR lasers is Cr^{2+}:ZnSe. Passively mode-locked femtosecond Cr^{2+}:ZnSe laser was first reported in 2006 [6] and the first KLM laser in 2009 [7,8]. To date, output power up to 300 mW [7], pulse energy up to 2.3 nJ [10], pulse duration as short as 80 fs [1,11], and parametric frequency conversion to the 4.5–5.5 μm wavelength range [12] have been demonstrated in the femtosecond regime.

The only disadvantage of the Cr^{2+}:ZnSe crystal is its comparatively high thermal lensing parameter ($70{\cdot}10^{-6}$ 1/K [2]), which potentially limits the power scalability. From this point of view a single crystalline Cr^{2+}:ZnS is a promising alternative. Together with the lower dn/dT ($46{\cdot}10^{-6}$ 1/K) it exhibits higher thermal conductivity (27 W/m·K) and thermal shock parameter (7.1 $W/m^{1/2}$) [13] resulting in potentially better power handling capability. From the spectroscopic point of view both crystals are in many respects similar with the main difference of 100-nm blue-shifted emission peak of Cr:ZnS. The first continuous-wave (CW) Cr:ZnS laser has been reported in 2002 in Ref. [13] and the diode-pumped version of it in Ref. [14]. The main reason of underinvestigation of the single crystal Cr:ZnS is the lack of its commercial availability. The formally cubic Cr:ZnS modification is a polytypical compound and can co-exist in several structure types, thus exhibiting natural birefringence [13]. Nevertheless, subject to a proper orientation, the CW output power of 700 mW at 2.35 μm with 700 nm wavelength tuning were demonstrated using Cr:ZnS single crystal as a laser active element [14]. Subsequently, the laser action in microchip configuration [15], picosecond passive mode-locking [16] and finally, femtosecond SESAM-initiated mode-locking was obtained in 2011 with 1.2 nJ 110 fs pulses [17]. The CW output power of 10 W was obtained recently using polycrystalline Cr:ZnS as an active element [18].

In this paper we demonstrate the first Kerr-Lens mode-locked Cr:ZnS laser, generating very good spectral quality and highly stable 69 fs pulses at 550 mW output power. Currently, those are the shortest mid-IR pulses at the highest reported power, generated directly from the oscillator in the mid-IR spectral region. These promising results open the way to further power scaling and reducing pulse duration down to single optical cycle.

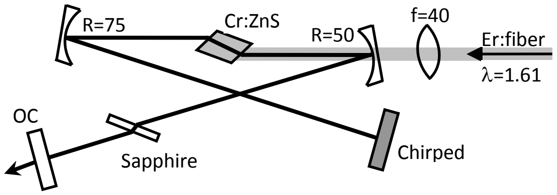

Fig. 1. Schematic of the femtosecond Cr:ZnS KLM laser.

The experimental setup is shown in Fig. 1. The laser has been assembled according to the classic X-folded astigmatically-compensated four-mirror cavity design. A PVT grown diffusion doped 2.5-mm-thick Cr:ZnS crystal with Cr^{2+} concentration of about $6{\cdot}10^{18}$ cm^{-3} [19] was mounted at Brewster angle on a copper heatsink without active cooling. The cavity consisted of two dichroic concave folding mirrors with radii of curvature 50 and 75 mm, a plane chirped high-reflector mirror [1,11] and a plane output coupler. Three different output couplers were used with transmission of about 1.5%, 4.5% and 18% at 2.4 μm. The CW Er-fiber laser from IPG Photonics providing up to 5 W of polarized output at 1.61 μm was used as a pump source. The pump beam was focused onto the crystal by a 40-mm AR-coated lens. The crystal absorbed about 80% of the incident pump power. The mode-locking was achieved by the soft-aperture Kerr-Lens effect using a moving mirror as a starting mechanism. The compensation of the group-delay dispersion (GDD) was achieved by the 1-mm sapphire plate inserted into the OC arm of the cavity and a single chirped high reflector mirror, making the resonator design especially compact and stable.

All measurements were performed in the open air with 40-50% relative humidity. The spectrum was analyzed by a commercial FTIR spectrometer at 1 cm^{-1} resolution. The pulse duration was measured using a home-made autocorrelator based on a two-photon absorption in an amplified Ge photodetector.

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In CW laser experiments a 1.5% output coupler was used. A CaF_2 prism was inserted into the HR arm of the cavity for spectral selection. The laser wavelength was tunable in the range between 2.17–2.88 μm. At the fixed wavelength of 2.367 μm about 380 mW output power was achieved for 2.75 W of incident pump power that corresponded to the slope efficiency of 15 %.

Kerr-lens mode-locked laser action was obtained after adjusting the position of the 75-mm-radii HR mirror near the end of the first stability region. The mode-locking was initiated by slightly tilting the chirped HR mirror. The available pump power was sufficient to achieve mode-locking with all the three output coupler mirrors. The laser routinely produced femtosecond soliton-like pulses at the repetition rate of 144.7 MHz. It was very stable in the certain output power range and once started could operate for several hours without readjustment.

The parameters of the mode-locked laser pulses for three different output couplers are listed in the Table 1. The interferometric autocorrelation trace of Cr:ZnS laser with the 18% output coupler as well as the beam profile are shown in Fig. 2, the spectrum is plotted in Fig. 3.

Table 1. Laser characteristics of Kerr-lens mode-locked Cr:ZnS laser with different OC mirrors

OC transmission, %	1.5	4.5	18
Pulse repetition rate, MHz		144.7	
Average output power, mW	200	380	550
Pulse energy, nJ	1.4	2.6	3.8
Pulse duration, fs	68	70	69
Peak power, kW	20	37	55
Central wavelength, nm	2430	2385	2390
Spectral bandwidth FWHM, nm	97	91	91
Time-bandwidth product ΔνΔτ	0.336	0.336	0.335

Optical-to-optical efficiency of the laser reached 13% at maximum output power. The minimum pulse duration of 69 fs is equal to 8-9 optical cycles at this wavelength. Laser pulses were close to transform-limited with the time-bandwidth product of 0.335. The beam profile has a slight ellipticity, but is much better in quality than in the SESAM-based setup [19], allowing convenient launching into a single-mode fiber [20].

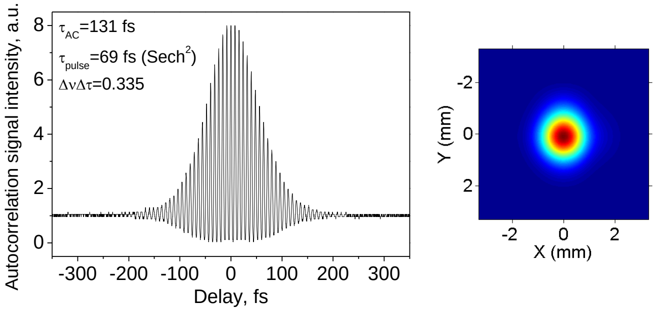

Fig. 2. (Color online) Autocorrelation trace of KLM Cr:ZnS laser pulses at highest output power (a) and beam profile at ~50 cm after the output coupler (b).

A chirp-free pulse, calculated from the measured output spectrum, would have had a duration of 65 fs. We assume that the extra 4 fs in the measured autocorrelation trace arise from the dispersion accumulated in the OC substrate (3 mm of YAG) and beamsplitters further down the beamline (3 mm CaF_2 and 1 mm ZnSe). The good beam quality of the output pulse (Fig. 2b) allowed launching into the single-mode fiber for transport and is a prerequisite for efficient nonlinear wavelength conversion like e.g. in a sync-pumped OPO [12].

Precise dispersion management is critical on the way toward few-optical-cycle pulse generation. Combination of anomalous dispersion of ZnS, normal dispersion of sapphire and a chirped mirror [1,11,19] allowed to obtain relatively flat GDD curve with total net GDD per cavity roundtrip about -450 fs^2 at central wavelength (Fig. 3).

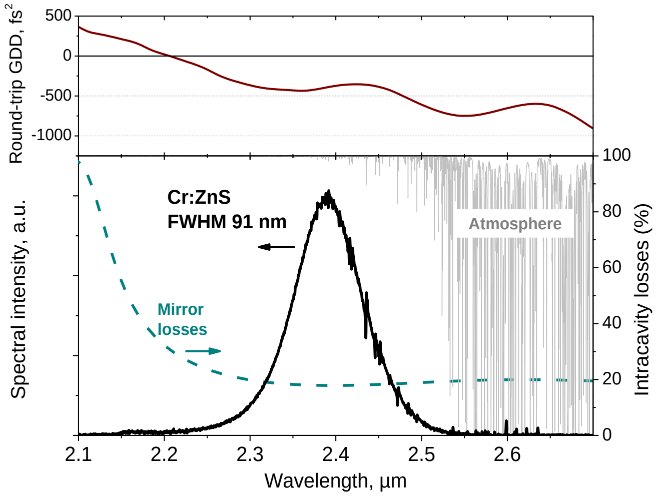

Fig. 3. (Color online) Output spectrum of a femtosecond Cr:ZnS laser (black), calculated round-trip GDD (dark red), and intracavity losses due to the mirrors (blue-green dashed) and atmospheric absorption (gray).

The important aspect of the femtosecond oscillator is its power scalability. We found the high third-order optical nonlinearity of the Cr:ZnS crystal ($90\cdot10^{-16}$ cm^2/W) to be the main power-limiting factor in our experiments. Further increasing of the pump power resulted in unstable double-pulsing and harmonic mode-locking regimes [21] for all available output couplers. With 4.5%

output coupler we were able to obtain a comparatively stable double-pulsed mode-locking regime with a reproducible pulse separation of about 2.4 ps. The laser produced 720 mW of average output power with 2.5 nJ pulse energy. The autocorrelation trace and spectrum are plotted in Fig. 4. Further increase of the pulse energy would require reducing the peak power density inside the active medium. The chirped pulse oscillator concept [22] is very promising from this point of view.

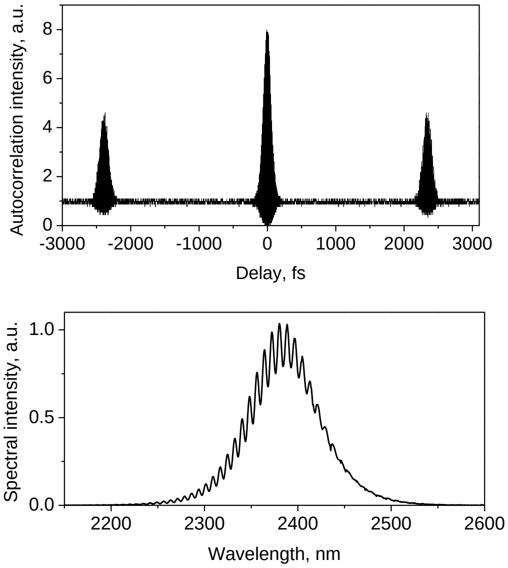

Fig. 4. Autocorrelation trace (a) and optical spectrum (b) of a double-pulsed KLM Cr:ZnS laser with pulse separation of 2.4 ps.

Since the gain bandwidth of Cr:ZnS crystal can support pulses as short as ~15 fs, it is instructive to discuss the spectrum-narowing factors preventing the pulse from further shortening. On the blue side, the spectrum is limited by the increased transmission in both input and output coupler mirrors (their combined transmission for 18% output coupler is shown by blue-green dashed line in Fig. 3) as well as by the intracavity third-order dispersion, causing the net GDD (dark-red solid line) to become positive around 2.2 μm. The red side is mainly affected by the atmospheric absorption in the 100 cm long cavity (Fig. 3, solid gray line). The effect of the water vapor absorption lines can be seen in the output spectrum as a characteristic modulation [22].

Summarizing, we report the first Kerr-Lens mode-locked laser based on a Cr^{2+}:ZnS crystal. The laser was passively mode-locked, using only one chirped mirror and a sapphire plate for dispersion compensation, and generated 69 fs pulses at 2.39 μm. The pulses are distinguished by high spectral quality, stability and the highest reported output power of 550 mW generated directly from the oscillator im the mid-IR. Those are the shortest pulses generated so far in all Cr^{2+}-based lasers. Further shortening of the pulse duration, potentially down to single optical cycle, as well as power-scaling into several Watt domain will lead to the practical and cost effective high-power ultrashort pulsed laser in the very important for applications molecular fingerprint region.

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