Kerr-lens mode-locked polycrystalline Cr:ZnS femtosecond laser pumped by a monolithic Er:YAG laser*

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(Received 19 September 2016; revised manuscript received 13 October 2016; published online 8 December 2016)

We demonstrated a Kerr-lens mode-locked polycrystalline Cr:ZnS laser pumped by a narrow-linewidth linear-polarised monolithic Er:YAG nonplanar ring oscillator operated at 1645 nm. With a 5-mm-thick sapphire plate for intracavity dispersion compensation, a compact and stable Kerr-lens mode-locking operation was realised. The oscillator delivered 125-fs pulses at 2347 nm with an average power of 80 mW. Owing to the special polycrystalline structure of the Cr:ZnS crystal, the second to fourth harmonic generation was observed by random quasi-phase-matching.

Keywords: middle-infrared laser, mode-locked laser, polycrystalline Cr:ZnS

PACS: 42.65.Re, 42.55.Rz, 42.60.Fc **DOI:** 10.1088/1674-1056/26/1/014206

1. Introduction

Middle-infrared (mid-IR) laser sources, owing to the special interest in this spectral region, have received significant attention for applications in molecular spectroscopy, environmental monitoring, medical diagnostics, communications, and defense. Transition metal (TM) doped II–VI chalcogenides were considered as promising mid-IR gain media when firstly introduced by Lawrence Livermore National Laboratory (LLNL).[1,2] Owing to the low energy of the optical phonon cutoff caused by the heavy ions in II–VI crystals, the efficiency of impurity nonradiative decay is decreased. The tetrahedrally coordinated structures of II–VI semiconductor crystals enable small crystal field splitting so that the TM impurity transitions are placed in the mid-IR spectral range.[3]

Cr:ZnS is one such crystal. It has a four-level energy structure, no excited state absorption, broad absorption bands covering some efficient and reliable commercial fibre laser sources, and ultrabroad vibronic emission bands providing the ability of broadly tunable emission in the mid-IR. The structure of the ZnS crystal is not that of wurtzite but rather a modification of the cubic structure. X-ray analysis reveals the crystal to have a certain degree of hexagonality, which is one of the most abundant structures of chalcogenide compounds. Besides the two basic wurtzite (hexagonal) and zincblende (cubic) structures, a number of so-called mixed-polytype structures[4] are common in ZnS crystals. Cr:ZnS not only has excellent chemical and mechanical stability but also has a semiconducting property that gives this crystal strong nonlinear characteristics. Such nonlinear effects can induce charge transfer, photorefractive phenomenon, harmonic generation, parametric processes, and a variety of self-focusing effects. Cr:ZnS also provides a low phonon cutoff to reduce the nonradiative decay rate, resulting in a high fluorescence quantum efficiency at room temperature.[5] These properties enable Cr:ZnS to support ultrashort pulse generation as short as a few optical cycles.[6] For its favourable spectroscopic and physical characteristics, Cr:ZnS has been considered as the "Ti:sapphire" in the mid-IR.[7]

However, high-quality Cr:ZnS single-crystal materials are difficult to obtain. Crystal sublimation during the growth process results in poor uniformity of the single-crystal samples and limits the dopant concentration. Apart from its similar properties to single-crystal Cr:ZnS, polycrystalline Cr:ZnS has more advantages. An important advantage of the polycrystalline Cr:ZnS laser medium is its usefulness in the post-growth diffusion doping technology, which enables mass production of large-size laser gain elements with high dopant concentration, uniform dopant distribution, and low losses. Polycrystalline Cr:ZnS gain elements are generally fabricated by thermal diffusion doping of polycrystalline ZnS, which is grown by chemical vapour deposition (CVD).[8] Post-growth diffusion doping of CVD-ZnS retains the polycrystalline zincblende structure of the material with a grain size of 50–100 μm.[9] Polycrystalline Cr:ZnS consists of a mul-

*Project supported by the National Natural Science Foundation of China (Grant Nos. 61205130, 61465012, and 61564008).
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titude of microscopic single-crystal grains. The grain size is of the order of the coherence length of the second harmonic generation (SHG) process in the mid-IR wavelength range.[3] The random distribution of grain size and orientations results in random quasi-phase-matching.

Since firstly reported in 1996, the Cr:ZnS laser at > 2 μm has made significant progress. Initial progress in single-crystal Cr:ZnS lasers included continuous-wave (CW) tunable lasers presented in 2002,[10,11] a semiconductor saturable absorber mirror (SESAM) mode-locked picosecond laser operated at ~ 2.45 μm and generating 1.1-ps pulses,[12] a SESAM femtosecond laser with 130-fs pulse duration at 2400 nm,[13] and a Kerr-lens mode-locked (KLM) femtosecond laser operating at a wavelength of 2.39 μm with an average power of 550 mW for 69-fs pulses.[14] The first polycrystalline Cr:ZnS laser was revealed in 2009.[15] Kerr-lens mode-locked operation was reported with 125-fs pulse duration and an average power of 30 mW.[16] Most recently, pulses of only three optical cycles (< 29 fs) were realized with an average power of 0.44 W at 2.4 μm.[17] Considering the absorption peak at ~ 1.7 μm for Cr:ZnS crystal and our Er:fibre pumping source at 1.53 μm, we tried to pump a laser with a 1645-nm narrow-linewidth linear-polarised monolithic Er:YAG nonplanar ring oscillator that was pumped by the Er:fibre source. We successfully realized stable KLM operation with this pumping scheme. The Cr:ZnS laser generated 125-fs pulses at a central wavelength of 2347 nm. The average output power was 80 mW at a repetition rate of 115 MHz. Owing to the isotropic polycrystalline structure of Cr:ZnS, the second to fourth harmonic generation was also observed by random quasi-phase-matching.

2. Experimental setup

The experimental setup similar to that used in our previous work[18] is shown in Fig. 1. A standard X-folded astigmatically compensated four-mirror cavity structure was used. The Cr:ZnS crystal (from IPG Photonics) was 3.61 mm long with a Cr^{2+} concentration of about 6.4×10^{18} cm^{-3} and cut at the Brewster angle. To reduce the thermal lens effect in the crystal, we wrapped the crystal with indium foil and placed it in a water-cooled copper heat sink whose temperature was maintained at 14.5 °C. The laser cavity includes two curved high-reflection mirrors (M_1 and M_2, each with a radius of curvature of 75 mm), a flat end mirror (HR), and a flat output coupler (OC). To match the absorption peak of the Cr:ZnS crystal, a monolithic narrow-linewidth Er:YAG nonplanar ring oscillator was employed as the pump source.[19] The pump source produced a maximum 6.1-W single-frequency laser at 1645 nm with relative power stability of 0.33% within 30 min. The pump laser was collimated with a 300-mm-focal-length plano-convex lens and focused into the crystal by a 50-mm-focal-length plano-convex lens. A half-wave plate (HWP) was introduced to adjust the polarization to match that of the Cr:ZnS crystal. The absorption rate of the pump laser was measured to be $\sim 50\%$ for the crystal used in the experiment with non-lasing. The group-velocity dispersion of the Cr:ZnS crystal at 2.3 μm was calculated to be $+123$ fs^2/mm and it was -226 fs^2/mm for a sapphire plate. Therefore, for compactness and flexibility, a 5-mm-thick sapphire plate was inserted into the cavity to compensate for the cavity dispersion. The net group delay dispersion was -684.75 fs^2 at 2347 nm regardless of other optics. The total cavity length was 1.3 m, corresponding to a repetition rate of 115 MHz.

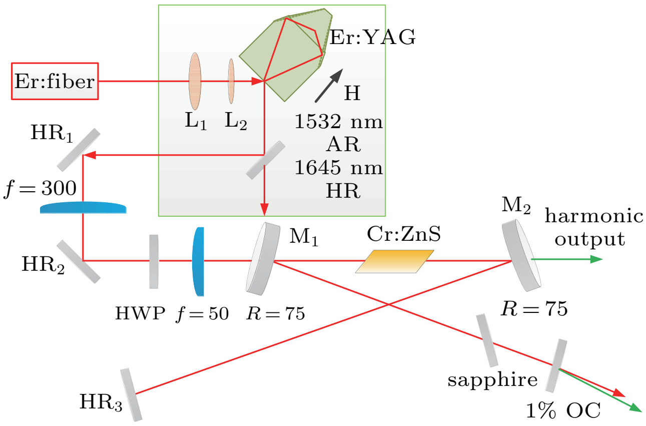

Fig. 1. (color online) Experimental setup of the Kerr-lens mode-locked Cr:ZnS laser. HR, high reflection mirror; HWP, half-wave plate; M_1, M_2, concave mirrors; OC, output coupler.

3. Results and discussion

We first characterised the CW performance of the Cr:ZnS laser. A 1% transmittance OC was used to couple out the laser. When the 5-mm-thick sapphire plate was inserted in the cavity, a maximum 212-mW CW laser at 2347 nm was obtained with 5.12-W input pump power, corresponding to a slope efficiency of $\sim 4\%$. The corresponding output power with respect to the input pump power is depicted in Fig. 2. There is no evidence of pump saturation, but we did not increase the pump power further to avoid crystal damage. The relatively low laser efficiency was probably due to the small absorption efficiency as well as to the small OC used in the experiment.

Kerr-lens mode-locking was realized by finely adjusting the cavity to the stability edge. This was implemented by care-

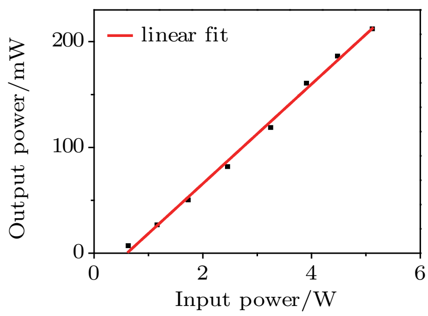

Fig. 2. (color online) CW output power as a function of the input power of the Cr:ZnS laser with 1% OC.

fully adjusting the position of the gain medium and M_2 mirror. As a starting mechanism, we slightly tilted the HR_3 mirror to initiate the mode-locking operation. With 2.88-W pump power, we achieved 80-mW mode-locked average power. We found that further increasing the pump power by 10%–15% led to unstable mode-locking with multi-pulsing and spikes in the spectrum. Therefore, to obtain higher output power, better thermal management should be taken into account. The mode-locking spectrum was measured by an optical spectral analyser (A.P.E WaveScan), which is shown in Fig. 3(a). The laser spectrum had a nearly Gaussian shape centred at 2347 nm with a 50.2-nm-wavelength bandwidth (full width at half maximum). The pulse duration was measured by an intensity autocorrelator (A.P.E PulseCheck), as shown in Fig. 3(b). Assuming a sech^2 pulse shape gives a pulse duration of 125 fs. The time–bandwidth product was calculated to be 0.342, which was close to the Fourier transform limit of the sech^2 pulse.

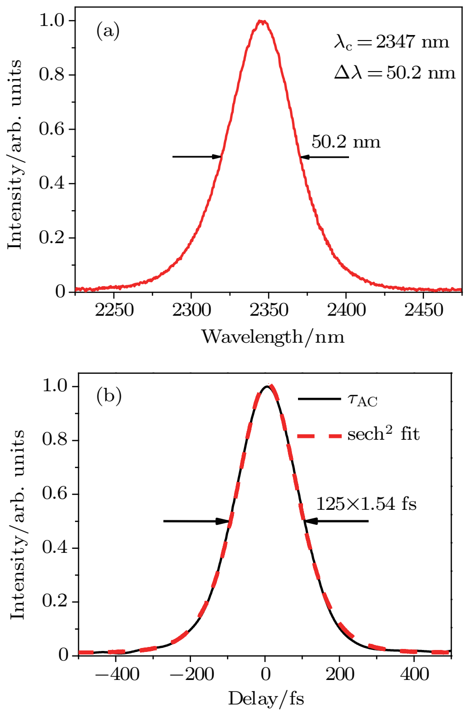

Fig. 3. (color online) (a) Laser spectrum and (b) autocorrelation trace of the KLM Cr:ZnS laser.

During the mode-locking operation, we could observe second to fourth harmonic generation in the visible to the near-infrared region. The harmonic spectra were measured behind the M_2 mirror, as shown in Fig. 4. The spectral width of the second harmonic is 18.7 nm, centred at 1164 nm. The widths are 11.8 and 5 nm for the third and the fourth harmonics, centred at 783 and 586 nm, respectively. The generation of nonlinear harmonics is due to the polycrystalline structure of the Cr:ZnS crystal. Random quasi-phase-matching may be fulfilled easily by the random distribution of grain size and orientations. Consequently, the phase-matching condition is not sensitive to the orientation of the sample.[20] However, the harmonics yield is very low for polycrystalline Cr:ZnS, which is mainly due to the large mismatch between the average size of the grains and the coherence length of the harmonics.

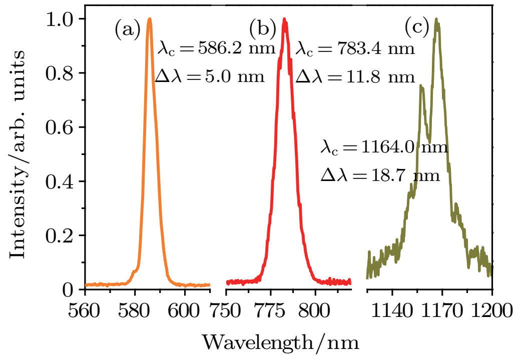

Fig. 4. (color online) Spectra of the harmonic wavelengths.

4. Conclusion and perspectives

We have demonstrated a monolithic Er:YAG-laser-pumped Kerr-lens mode-locked polycrystalline Cr:ZnS oscillator. The laser generates 125-fs pulses at 2347 nm. With a pump power of 2.88 W, an average power of 80 mW is obtained at a repetition rate of 115 MHz. By optimising the thermal management and dispersion compensation, sub-100-fs pulse duration with watt-level average power is feasible. An ultrafast laser at > 2 μm is an attractive driving source for high-cutoff, high-harmonic generation and 3- to 6-μm MIR pulse generation.

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