Femtosecond operation and self-doubling of Cr:ZnS laser

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Abstract: Prismless dispersion-controlled Cr:ZnS laser generates first femtosecond (110 fs) pulses at 180 MHz repetition rate around 2400 nm with average power 200 mW. Co-propagating second-harmonic pulse at 1200 nm is simultaneously generated in ceramic sample.

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1. Introduction

Cr^{2+}-doped lasers of the II-VI family [1] operating in the "molecular fingerprint" range between ~2 and 3.5 μm [2] have recently matured to the commercial continuous-wave lasers, with the broadest among existing lasers amplification bandwidth $\Delta\lambda/\lambda$, making them a practical ultrashort-pulsed alternative to Ti:sapphire in the mid-IR wavelength range [3].

Cr:ZnS laser, first reported in continuous-wave regime [4,5] is particularly interesting for practical ultrashort pulsed sources as it has an advantage of the emission being located in the middle of the water window around 2.3 μm and shifted to shorter wavelength by about 100 nm relatively to Cr^{2+}:ZnSe. Having similar to Cr:ZnSe spectroscopic properties, Cr:ZnS crystal has a larger bandgap (compare 3.84 eV in ZnS and 2.83 eV in ZnSe), better hardness, higher thermal conductivity in the cubic phase (27 W/mK in ZnS vs.19 W/mK in ZnSe), higher thermal shock parameter (7.1 $W/m^{1/2}$ in ZnS vs. 5.3 $W/m^{1/2}$ in ZnSe) and lower dn/dT ($+46\cdot10^{-6}$ K^{-1} in ZnS vs. $+70\cdot10^{-6}$ K^{-1} in ZnSe) [5]. The most attractive feature of Cr:ZnS is therefore the better power handling capability, which is potentially higher than that of Cr:ZnSe. At the same time the gain bandwidth is comparable, making Cr:ZnS attractive for both high-power and ultrashort pulse generation.

In Cr:ZnSe the first femtosecond operation (80 fs) using SESAM has been reported five years ago [6]. Recently, KLM mode-locking has been reported using bulk dispersion compensation [7] and also prisms [8]. Attempts to mode-lock Cr:ZnS using SESAM resulted in 1 ps pulses [9]. It is worth noting that it is generally more difficult to mode-lock Cr:ZnS than Cr:ZnSe as the nonlinearity of Cr:ZnS is a factor of two lower than in Cr:ZnSe [10].

In this work, we present the first femtosecond prismless Cr:ZnS laser operating in the water-free window around 2.3-2.4 μm. The single-crystal based laser delivers up to 200 mW at fundamental wavelength, whereas the ceramic sample generated simultaneously pulses at the fundamental (2.38 μm) and the second harmonic wavelength (1.19 μm), hence, opening up interesting application opportunities in metrology and beyond. To provide better understanding of this coupled second harmonic pulse behavior, we provide a comparative analysis of this phenomenon in Cr:ZnS and Cr:ZnSe lasers. Demonstration of mode-locked operation in the normal dispersion regime is especially important in the context of power scaling capability of Cr:ZnS.

1. Experimental setup

The experimental set up is schematically shown in Fig. 1. For the active element we used the 2.5 mm thick uncoated plate of Cr^{2+}:ZnS, with over 90 % absorption at 1.61 μm (the same sample as used in [4,5]), grown using physical vapor transport method and subsequently diffusion doped with Cr. The lifetime in this sample was measured to be 3.7 μs [5]. The SESAM sample consisted of a saturable absorber based on 50 layers of InAs/GaSb quantum wells, grown on top of a dielectric mirror made from 15 alternating layers of quarter-wave thickness GaSb and $AlAs_{0.08}Sb_{0.92}$ on a GaSb substrate (for details see [11]). The SESAM had a small-signal absorption of 12% per bounce, a calculated relaxation time of 200-300 ps, and a saturation fluence of 40 $\mu J/cm^2$.

An X-fold cavity configuration with a SESAM was optimized for reliable self-starting. The cavity consisted of two concave mirrors with 75 and 100 mm radii of curvature, a folding mirror with 75 mm radius of curvature, which focuses light into a ~50 μm spot diameter onto the SESAM, and a plane 2 % output coupler. For dispersion compensation, a single 3-mm thick YAG plate and one dispersive mirror were used. The diode-pumped Er-fiber laser (IPG Laser GmbH, up to 5 W polarized output) was focused onto the crystal through the $f = 30$ mm lens. The Cr:ZnS crystal was edge-mounted on a copper block without any additional cooling. All measurements were performed at open air, with relative humidity ~40%. The spectrum is analyzed by a commercial FTIR spectrometer. The pulse duration was measured using an autocorrelator based on a two-photon absorption in an amplified Ge photodetector.

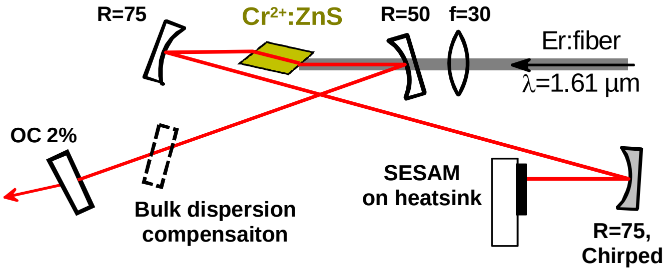

Figure 1: Experimental set-up of the Cr:ZnS mode-locked laser. Combination of the chirped mirror and a bulk dispersion compensator provided net anomalous dispersion about -1000 fs2. WWith a chirped mirror only, the laser operated in normal dispersion (dissipative soliton) regime.

The pump threshold and output power in CW regime were measured to be 70 mW and 380 mW, respectively. With SESAM in place, the threshold pump power increased to ~150 mW. With the SESAM as a starting mechanism for mode-locking and in the anomalous (soliton) dispersion regime, the laser routinely produced pulses of about 110–130 fs, corresponding to 14–16 optical cycles (Fig. 2) at the repetition rate of 180 MHz and output power up to 140 mW. An output spectrum of 50–60 nm FWHM is centered around 2350–2400 nm where calculated intracavity group-delay dispersion is about −1000 fs^2. Further decrease of the net GDD to about −500 fs^2 resulted in harmonic multiple-pulse operation regime, as predicted by theory [14]. We could also obtain mode-locked operation in the normal (dissipative soliton) dispersion regime (Fig. 2c, 2d) with about 600-fs pulses at up to 200 mW output power.

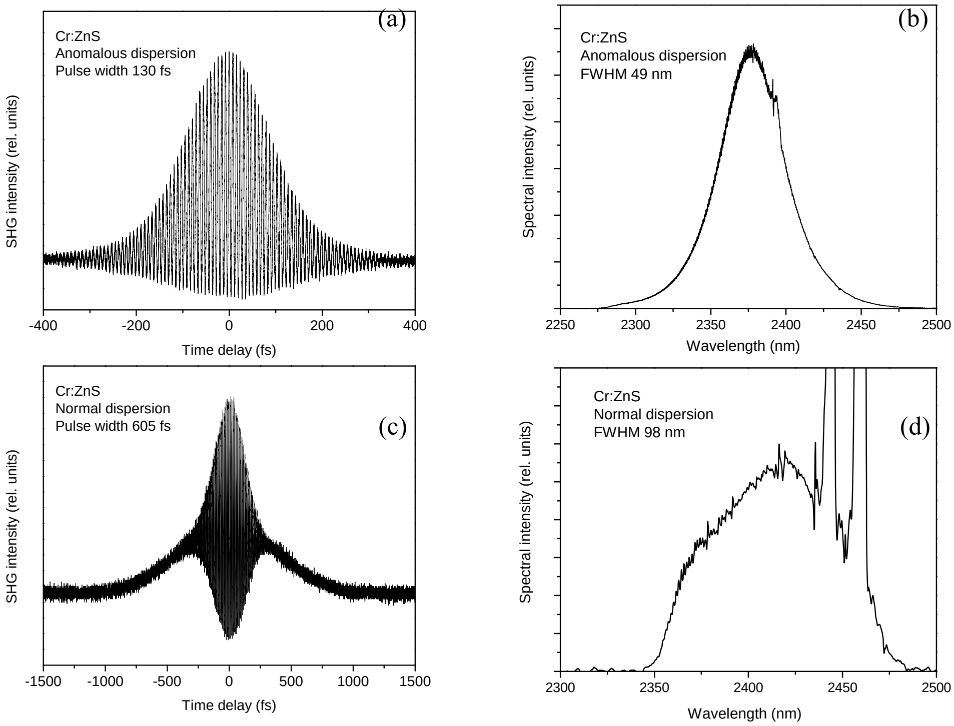

Figure 2: Autocorrelation traces and a corresponding spectra of a Cr:ZnS laser, operating in normal (soliton) dispersion regime (a,b) and in anomalous (chirped-pulse) dispersion regime (c,d). Note that the narrow features on the spectra (b) and (d) are due to the intracavity water absorption [12,13].

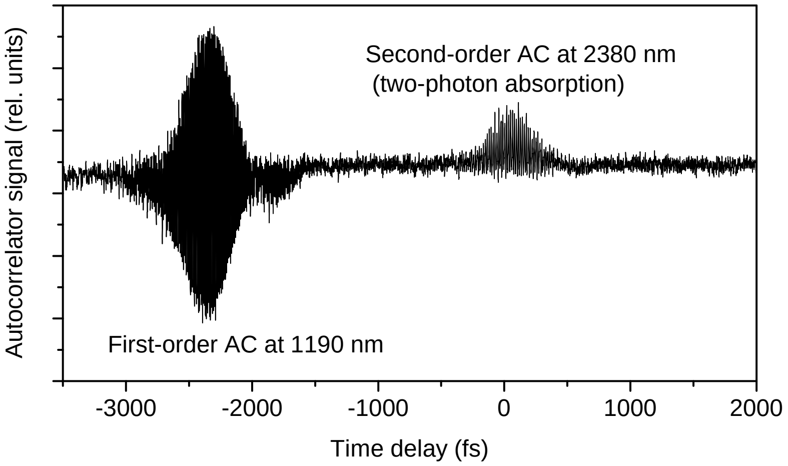

Figure 3: Autocorrelation trace of the mode-locked Cr:ZnS ceramic laser, showing the first-order interferogramm of the second-harmonic radiation at -2.2 ps time delay (in the beamsplitter) with respect to the second-order autocorrelation peak at 0 ps.

An interesting from the nonlinear-optical standpoint observation was made, when the single crystalline Cr:ZnS is replaced by a ceramic sample. Similarly to ceramic Cr:ZnSe [15] the autocorrelation trace then reveals that besides the main pulse at 2380 nm, the laser also emits radiation at second harmonic around 1190 nm (Fig. 3). The observed second harmonics is due to the fact that the ceramic sample consists of a multitude of differently oriented microcrystallites, providing for the quasi-regular switch of the nonlinear interaction phase. This is similar to the orientation patterning in OP-GaAs devices and, in a broader sense, to periodical poling, but with significantly reduced efficiency.

3. Conclusion

Summarizing, we have demonstrated the first single-crystalline KLM femtosecond Cr:ZnS laser, generating high power (up to 200 mW) stable and clean 110 fs pulses at 180 MHz repetition rate. The ceramic Cr:ZnS in the same setup emits radiation at two wavelengths: 2.4 µm and 1.2 µm. We provide the physics behind this phenomenon as well as give outlook on future improvements in operation of such a "double pulse" laser and possible applications.

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