Continuous-Wave Fiber-Pumped $\text{Cr}^{2+}:\text{ZnSe}$ Laser

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ABSTRACT

Power performance of a compact, broadly tunable, continuous-wave (cw) $\text{Cr}^{2+}:\text{ZnSe}$ laser pumped by a thulium fiber laser at 1800 nm was investigated. In the lasing experiments, a $\text{Cr}^{2+}:\text{ZnSe}$ sample with a small-signal differential absorption coefficient of 11 cm^{-1} and a fluorescence lifetime of $4.6 \mu\text{s}$ was used. An astigmatically compensated x-cavity with 15 % output coupler produced as high as 640 mW of output power at 2480 nm with 2.5 W of incident pump power. Resonator losses were investigated using three different methods, and an in-depth analysis of the results was performed. The stimulated emission cross section values determined from laser threshold data and fluorescence measurements were in good agreement with each other. Finally, broad, continuous tuning of the laser was demonstrated between 2240 and 2900 nm by using an intracavity Brewster-cut MgF_2 prism and a single set of optics.

Keywords: Solid-state lasers, mid-infrared lasers, fiber-pumped lasers, laser tuning, transition metal ions, II-VI compound semiconductors, solid state spectroscopy, ZnSe , $\text{Cr}^{2+}:\text{ZnSe}$ laser, fluorescence measurements.

1. INTRODUCTION

When doped with transition metal (TM) ions, II-VI compound semiconductors exhibit strong absorption and emission bands in the mid-infrared region of the electromagnetic spectrum¹⁻⁹. Recently, DeLoach et al. investigated the emission characteristics of these materials (zinc chalcogenides doped with Cr^{2+} , Co^{2+} , Ni^{2+} , and Fe^{2+}) and demonstrated lasing in $\text{Cr}^{2+}:\text{ZnSe}$, $\text{Cr}^{2+}:\text{ZnS}$ ^{10, 11}. After this pioneering work, considerable attention has been focused on the development of mid-infrared TM ion-doped chalcogenide lasers, and lasing action was also demonstrated in other hosts including $\text{Cr}^{2+}:\text{ZnS}_x\text{Se}_{1-x}$ ¹², $\text{Cr}^{2+}:\text{CdSe}$ ¹³, $\text{Cr}^{2+}:\text{CdTe}$ ¹⁴, $\text{Cr}^{2+}:\text{Cd}_x\text{Mn}_{1-x}\text{Te}$ ^{15, 16}, $\text{Cr}^{2+}:\text{Cd}_x\text{Zn}_{1-x}\text{Te}$ ¹⁷ and $\text{Fe}^{2+}:\text{ZnSe}$ ¹⁸. Among all these candidates, due to its near unity quantum yield at room temperature¹⁰, $\text{Cr}^{2+}:\text{ZnSe}$ has become the most extensively studied member of this class of tunable solid-state gain media. In $\text{Cr}^{2+}:\text{ZnSe}$, broadly tunable laser emission can be obtained between 2000 and 3100 nm¹⁹, and various modes of pulsed and cw operation have been demonstrated^{10, 20, 21}. To date, $\text{Cr}^{2+}:\text{ZnSe}$ lasers have been used in applications such as medicine²², atmospheric imaging²³, spectroscopy²⁴⁻²⁶, and pumping of mid-infrared lasers and optical parametric oscillators²⁷.

The ^5D energy level of tetrahedrally coordinated Cr^{2+} ($3d^4$) ions is split into the $^5\text{T}_2$ and ^5E energy levels in the ZnSe crystal field. The infrared absorption and emission spectra observed in $\text{Cr}^{2+}:\text{ZnSe}$ originate from transitions between these levels. As a result of vibronic broadening the absorption band due to the $^5\text{T}_2 \rightarrow ^5\text{E}$ transition is very broad, and extends from 1500 nm to 2100 nm (absorption peak=1775 nm, width (FWHM)=365 nm)²⁸⁻³⁰. This broad absorption band makes it possible to use several alternative laser systems as pump sources. Some of the pump lasers that have been used to date include $\text{Co}^{2+}:\text{MgF}_2$ ^{10, 11, 31-33}, $\text{Tm}:\text{YALO}$ ²⁰, $\text{Tm}:\text{YLF}$ ³⁴, $\text{NaCl}:\text{OH}^-$ ³⁵, $\text{Ho}:\text{YALO}$ ³⁶, $\text{Er}:\text{YAG}$ ²⁵, laser diodes³⁷⁻³⁹, Er-doped fiber lasers^{23, 24, 40-43}, $\text{Ba}(\text{NO}_3)_2$ & BaWO_4 Raman lasers^{44, 45}, and KTiOPO_4 (KTP) optical parametric oscillators (OPO)⁴⁶. Choosing a pump laser with a wavelength close to the peak of the absorption band (1775 nm) enables the use samples with relatively low Cr^{2+} concentration and eliminates unwanted effects such as fluorescence quenching due to high ion concentrations. For example the fluorescence lifetime of the $\text{Cr}^{2+}:\text{ZnSe}$ samples decreases to half of its low-concentration value (from about $6 \mu\text{s}$ to $3 \mu\text{s}$) at a concentration level $14 \times 10^{18} \text{ ions/cm}^3$ (corresponding peak absorption coefficient of about 16 cm^{-1} at 1775 nm)⁴⁶. Also, as the doping density is decreased, passive losses at the

lasing wavelength decrease, and more efficient lasing can be obtained. Among the available pump sources, lasers based on the 2- μm transition of the trivalent thulium ion (Tm^{3+}) have been among the preferred alternatives.

In this study, we describe a broadly tunable, cw, $\text{Cr}^{2+}:\text{ZnSe}$ laser pumped by a Tm-fiber laser at 1800 nm. Continuous wave power performance of the laser was characterized by using three different output couplers. Using the 15.3 % transmitting output coupler, output laser powers as high 640 mW is obtained with 2.5 W of pump power at 2480 nm. In this study we also performed a detailed analysis of lasing performance. Resonator losses were determined using three different methods, first by using the measured threshold powers, second by using the measured power efficiency values, and lastly using the spectroscopic absorption measurements. Numbers obtained from three different methods were in good agreement with each other. From the threshold data, the stimulated emission cross section was further determined to be $4.2 \times 10^{-23} \text{ m}^2$. This was also in good agreement with the value obtained by using the emission and lifetime data. Finally, a Brewster cut MgF_2 prism was inserted into the cavity to obtain smooth, continuous tuning between 2240 to 2900 nm, with a single set of optics.

2. EXPERIMENTAL

Fig 1. Schematic of the cw $\text{Cr}^{2+}:\text{ZnSe}$ laser setup. The x-cavity containing the $\text{Cr}^{2+}:\text{ZnSe}$ crystal was end-pumped by a Tm-fiber laser at 1800 nm (See the text for description of the various components).

Continuous wave $\text{Cr}^{2+}:\text{ZnSe}$ laser experiments were carried out with a four-mirror x-cavity as shown in Fig. 1. A commercial thulium fiber laser (IPG Photonics) at 1800 nm was used as the pump source. The pump laser could deliver cw powers up to 5 W. The collimated output of the fiber pump had a $1/e^2$ beam waist of 2.25 mm with a measured M^2 of 1.03. Because power and polarization instabilities were observed at higher power levels, pump powers up to 3 W were used in the lasing experiments. The pump beam was focused to a waist of $36 \mu\text{m}$ inside the gain medium, by using an input lens with a focal length of 10 cm. The $\text{Cr}^{2+}:\text{ZnSe}$ gain medium had a thickness of 2.6 mm, and had a unsaturated single pass absorption of about 95 % at 1800 nm. The sample was wrapped in indium foil and clamped inside a copper holder maintained at 15°C . It was further placed at Brewster's angle between two curved high reflectors (M1 and M2) each with $R=10 \text{ cm}$. The Fresnel reflection loss from the sample surfaces was measured to be around 0.2 %. Long arm of the cavity was terminated with a flat end high reflector (M3), and the other contained a flat output coupler (M4). The total cavity length was 90 cm, where the long arm length was 55 cm. The cavity beam waist at the center of the stability region was estimated to be $45 \mu\text{m}$. Three different output couplers with transmission of 3, 5.8, and 15.3 % (around 2500 nm) were used in the laser power characterization measurements. The cavity high reflectors (M1-M3) had a high reflectivity between 2260 and 2900 nm (reflection $\geq 99\%$), and they had a measured leakage of about 0.4 % at 2500 nm.

Broadband coating of the high reflectors was essential in obtaining smooth and continuous tuning of the laser between 2.2 and 2.9 μm .

Fig 2. Measured absorption spectrum of pure ZnSe (thin line) and chromium-doped ZnSe (thick line) sample.

Fig 3. Measured time-dependent fluorescence decay of the $\text{Cr}^{2+}:\text{ZnSe}$ sample. Inset figure shows the time dependence of the fluorescence signal on a logarithmic scale.

Figure 2 shows the measured absorption spectrum of the $\text{Cr}^{2+}:\text{ZnSe}$ sample used in the experiments (thick line), along with the absorption spectrum of a pure ZnSe sample (thin line). After subtracting the background losses due to surface imperfections and Fresnel reflections from the $\text{Cr}^{2+}:\text{ZnSe}$ absorption spectra, the pump absorption coefficient at 1800 nm was determined to be 10.7 cm^{-1} (corresponding to a chromium concentration of about $9.5 \times 10^{18} \text{ ions/cm}^3$). Comparing the absorption spectra of doped and undoped samples, the passive single pass loss of the $\text{Cr}^{2+}:\text{ZnSe}$ sample is found to be 2.1 % at the lasing wavelength (around 2500 nm). This give a crystal figure of merit ($\text{FOM} = \alpha_{1800}/\alpha_{2500}$, α_{1800} =differential absorption coefficient at 1800 nm and α_{2500} =differential loss coefficient at 2500 nm) of 70.

A pulsed optical parametric oscillator (OPO) operating at 1570 nm was used to measure the fluorescence lifetime of the sample. The pulsewidth and the repetition rate of the OPO were 65 ns and 1 kHz, respectively. Figure 3 shows the time-dependent fluorescence decay curve of the $\text{Cr}^{2+}:\text{ZnSe}$ sample. Inset plot in Fig. 3 shows the variation of the natural logarithm of the fluorescence intensity. The fluorescence signal shows a single exponential decay (only one slope in the inset graph), and by doing a single-exponential fit to the experimental data, the fluorescence lifetime was determined to be 4.6 ± 0.2 μsec .

3. RESULTS AND ANALYSIS

Figure 4 shows the measured cw lasing performance of the $\text{Cr}^{2+}:\text{ZnSe}$ sample for the 3 different output couplers. As an example, with the 3% transmitting output coupler, 300 mW of output power was obtained with 2.3 W of pump power. The corresponding threshold pump power and the incident power slope efficiency were 207 mW and 16 %, respectively. The free running laser output wavelength shifted slightly for the different output couplers used. While the lasing peak was at 2512 nm with the 3 % OC, it shifted to 2480 nm with the 15.3 % OC. The width of the laser line (FWHM) was about 0.5 nm.

Fig 4. Power efficiency curves for the Cr:ZnSe laser taken with the output couplers having 3, 5.8, and 15.3 % transmission.

The best power performance was obtained with the 15.3 % transmitting output coupler. Here, as high as 640 mW of output power was obtained with 2.5 W of pump. The slope efficiency with respect to incident pump power was 34 %. Due to thermal loading saturation of the output power was observed at pump powers beyond 2.5 W³⁴. The slope efficiencies were calculated from a linear fit to the laser efficiency data at low pump powers where the output power saturation is not yet effective. Threshold pump powers were estimated by using these linear fits. Table 1 lists the threshold pump power and the slope efficiency obtained with each output coupler.

Table 1. Threshold pump power and slope efficiency values for each output coupler (OC).

OC Transmission (%)	Slope Efficiency (%)	Threshold Pump Power (mW)
3	16	208
5.8	23	277
15.3	34	413

Resonator losses at the lasing wavelength were estimated by using the Findlay-Clay analysis⁴⁷, Caird⁴⁸ analysis, and spectroscopic absorption measurements. In the Findlay-Clay analysis one can use the laser threshold data to estimate the cavity round trip passive loss. Under several simplifying assumptions (low output coupling, absence of excited-state absorption, uniform pump and laser spot sizes), the threshold pump power P_{th} required to attain lasing can be expressed as⁴⁹⁻⁵¹

$$P_{th} = \frac{\pi}{4} \frac{h\nu_p}{\sigma_{em}(\lambda_L) \tau_f} \frac{(\omega_L^2 + \omega_p^2)}{(1 - \text{Exp}(-\alpha_p \ell))} (T+L) = A(T+L), \quad (1)$$

where $h\nu_p$ is the energy of pump photons, $\sigma_{em}(\lambda_L)$ is the emission cross section at the laser wavelength, τ_f is the fluorescence lifetime of the upper laser level, ω_L and ω_p are the average values of the laser and pump spot sizes in the gain medium, $(1 - \text{Exp}(-\alpha_p \ell))$ is the total absorption at the pump wavelength, L is the passive round trip resonator loss, and T is the transmission of the output coupler. Collecting all the parameters in one constant, A , clearly shows that threshold pump power is proportional to the total round trip loss L . A plot of P_{th} versus T gives a straight line and the loss L can be determined by using the best-fit values of the slope (A) and intercept (AL).

In the Caird analysis, one can use the experimentally measured laser slope efficiency values for different output coupler transmissions to estimate the laser passive losses. According to the Caird analysis, the slope efficiency η of the laser be expressed to a good approximation as

$$\eta = \eta_0 \frac{T}{(T+L)}, \quad (2)$$

where η_0 is the maximum slope efficiency that can be obtained at high output coupling. η_0 is determined by such factors as the finite quantum defect of the laser transition, mode matching between the pump and the laser beams, and luminescence quantum efficiency. A plot of $1/\eta$ as a function of $1/T$ hence gives a straight line and L can again be determined in a straightforward way.

Fig 5. (a) Measured variation of the incident threshold pump power as a function of the output coupler transmission (Findlay-Clay plot) and (b) variation of the inverse of the slope efficiency with the inverse of the output coupler transmission (Caird plot). L was determined to be 10.6 and 6.2 % from the Findlay-Clay and Caird analyses, respectively.

Figure 5 (a) shows the variation of the laser threshold power with output coupler transmission, and Figure 5 (b) shows the dependence of the inverse of the slope efficiency ($1/\eta$) on inverse of the output coupling ($1/T$) (Caird Plot). A linear fit to the graphs gave an L value of 10.6 % for Findlay-Clay analysis (Fig. 5 (a)) and 6.2 % for Caird analysis (Fig.

5(b)). This disagreement between the passive round-trip cavity loss estimations with the Findlay-Clay and Caird analyses was also observed in several previous laser studies with $\text{Cr}^{2+}:\text{ZnSe}$ ^{11, 20, 31, 37} gain medium, and attributed to the existence of residual ground state absorption at the lasing wavelength^{19, 31, 37, 52, 53}. Since $\text{Cr}^{2+}:\text{ZnSe}$ medium has near unity quantum yield at room temperature¹⁰, the existence of ground state absorption affects only the threshold power (but not the slope efficiency)^{31, 49} and Findlay-Clay analysis gave higher L values than Caird analysis^{11, 20, 31, 37}. To check this further, we carried out direct loss measurements taking into account absorption and reflection losses of the crystal as well as the leakage from high reflectors. This method gave a value of 6.6 % (2 x 2.1 % gain medium passive loss, 2 x 0.2 % reflection loss from the gain medium surface, 5 x 0.4 % leakage from the cavity high reflectors) in reasonable agreement with the result of the Caird analysis. The average cavity passive round-trip loss was hence estimated to be 6.4 %. The excess part in the Findlay-Clay analysis value (10.6 % - 6.4 % = 5.2 %) may be attributed to the round-trip ground state absorption loss (L_{GSA}) for this sample. The calculated value is in good agreement with previous studies^{31, 49}.

The stimulated emission cross section of the gain medium at the laser wavelength can be determined from the threshold data by rewriting Eq. 1 as^{49, 51}

$$\sigma_{\text{em}}(\lambda_L) = \frac{\pi}{4} \frac{h\nu_p}{P_{\text{th}} \tau_f} \frac{(\omega_L^2 + \omega_p^2)}{(1 - \text{Exp}(-\alpha_p \ell))} (T + L + L_{\text{GSA}}). \quad (3)$$

Findlay-Clay analysis result value of $L + L_{\text{GSA}}$ (10.6 %) determined above was used in the calculation of the stimulated emission cross section. Using the threshold data taken with the 3, 5.8, 15.3 % output couplers, the value of σ_{em} was calculated to be $4.2 \pm 0.2 \times 10^{-23} \text{ m}^2$ at the laser wavelength (near 2500 nm).

Fig 6. Emission spectrum of the $\text{Cr}^{2+}:\text{ZnSe}$ medium excited with a cw Cr:YAG laser operating at 1490 nm.

Alternatively one can determine the stimulated emission cross section from fluorescence measurements, which require the fluorescence spectrum of the sample as a function of the wavelength and lifetime data. According to the Fuchtbauer-Ladenburg formula^{45, 54} the stimulated emission cross section $\sigma_{\text{em}}(\lambda)$ at a wavelength of λ within the emission band is given by

$$\sigma_{\text{em}}(\lambda) = \frac{\lambda^5}{8 \pi c n^2 \tau_{\text{rad}} \int_{\text{band}} \lambda I_e(\lambda) d\lambda} I_e(\lambda). \quad (4)$$

In Eq. 4, c is the speed of light, n is the refractive index of the medium, τ_{rad} is the radiative lifetime of the upper laser level (not the fluorescence lifetime), and $I_e(\lambda)$ is the wavelength dependent fluorescence intensity distribution.

Figure 6 shows the normalized emission spectrum of $\text{Cr}^{2+}:\text{ZnSe}$ gain medium excited at 1490 nm with a 50 mW cw $\text{Cr}^{4+}:\text{YAG}$ laser. In the measurements the emitted fluorescence was collected with a concave gold mirror and imaged at the entrance slit of a 0.5-m Czerny–Turner type monochromator, after passing through a high-pass optical filter that blocked the pump radiation. The fluorescence signal was detected with a PbS detector and amplified in two stages by using a preamplifier and a lock-in amplifier. The fluorescence spectrum was corrected for filter, detector and grating response. $\text{Cr}^{2+}:\text{ZnSe}$ gain medium has overlapping absorption and emission bands between 1.7–2.3 μm . To minimize the reabsorption of the of the emitted photons in this region a sample with a low chromium concentration ($3 \times 10^{18} \text{ cm}^{-3}$) was used in the emission measurement. Other parameter values appearing in Eq. 4 are $n = 2.45$, $\tau_{\text{rad}} = 6 \mu\text{s}$ ⁴⁶. Here as the radiative lifetime we take the measured value of the fluorescence lifetime at room temperature for a very low Cr^{2+} doped sample, where concentration quenching of fluorescence is negligible⁴⁶. Low temperature fluorescence lifetime measurement is not necessary for the $\text{Cr}^{2+}:\text{ZnSe}$ gain medium, since non-radiative decay rates are negligible at room temperature¹⁰. The normalized emission spectrum data were used to determine the lineshape function and the area under the emission spectrum. The peak of the observed emission spectrum was at a lower wavelength (around 2050 nm), than most of the previously reported values (around 2400 nm)^{10, 45, 55}, but came close to the peak position reported by Mirov et al. (around 2150 nm)⁴¹. This difference is possibly due to the minimization of reabsorption of emitted photons in the lower wavelength side by using a sample with low doping concentration, and use of a low wavelength (1490 nm) pump source. Using Eq. 4, peak emission cross section was determined to be $12 \times 10^{-23} \text{ m}^2$ (at 2050 nm), which is in good agreement with previously reported values in the literature^{10, 19, 44, 45, 56}. However, as noted earlier, the reported peak emission wavelengths are different (around 2400 nm). Using the shape of the emission spectrum the emission cross section at the wavelength of 2500 nm is determined to be $4.3 \times 10^{-23} \text{ m}^2$. This value is in very good agreement with the emission cross section value ($4.2 \times 10^{-23} \text{ m}^2$) determined from the laser threshold data at 2500 nm.

Fig 7. Continuous-wave tuning curve of the $\text{Cr}^{2+}:\text{ZnSe}$ laser taken with an intracavity MgF_2 prism. Using a single set of optics, broad tuning could be obtained between 2240 and 2900 nm. Solid and dashed lines show the variation of the cavity high reflector and output coupler transmissions with wavelength, respectively.

Finally, using a Brewster-cut MgF_2 prism, we investigated the wavelength dependence of the output power. The prism was inserted in the high-reflector arm of the resonator, and the output coupler with 3 % transmission was used. In these measurements, the laser cavity was not purged, and the relative humidity was around 67 %. The tuning curve was taken with at an incident pump power of 1.8 W, and the output power of the laser decreased from 255 to 250 mW after the insertion of the prism. Figure 7 shows the obtained tuning curve, where the square dots represent the measured output power as a function of the emission wavelength. Using a single set of optics, broad tuning could be obtained between 2240 and 2900 nm. While the free running laser wavelength was around 2500 nm, the peak of the laser tuning curve occurred at 2320 nm after the insertion of the tuning prism. The dashed and solid lines in Fig. 7 show the transmission of the output coupler and the cavity high reflectors, respectively. Due to the atmospheric losses inside the cavity the tuning

curve has several deeps between 2.6 and 2.9 μm as it was previously observed^{31, 57} in non-purged cavities. Using a purged cavity could eliminate the dips in the tuning curve and increase the laser efficiency¹⁹. Tuning on both sides was limited by the reflectivity of the cavity high reflectors (see the solid line in Fig. 7). Previous studies demonstrated tuning with $\text{Cr}^{2+}:\text{ZnSe}$ gain medium between 2000 and 3100 nm, using different mirror sets⁵⁵.

In conclusion, we have described in detail the operation of a cw broadly tunable $\text{Cr}^{2+}:\text{ZnSe}$ laser pumped by a thulium-fiber laser at 1800 nm. A maximum output power of 640 mW and a slope efficiency of 34% with respect to the incident pump power were obtained, using a 15% output coupler. Lasing performance data taken with 3, 6, and 15 % output couplers were analyzed to determine the passive resonator losses and the stimulated emission cross section of the gain medium. The emission cross section value was determined to be $4.2 \times 10^{-23} \text{ m}^2$ from the laser efficiency data and $4.3 \times 10^{-23} \text{ m}^2$ from the fluorescence spectrum measurements at 2500 nm. Using an intracavity Brewster-cut MgF_2 prism and a single set of optics, continuous broad tuning of the laser was demonstrated between 2240 and 2900 nm.

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