Mid-IR laser oscillation in Cr^{2+}:ZnSe planar waveguide

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Abstract: We demonstrate 2.6 μm mid-infrared lasing at room temperature in a planar waveguide structure. Planar waveguides were fabricated using pulsed laser deposition (PLD) by depositing chromium doped zinc selenide thin films on sapphire substrate (Cr^{2+}:ZnSe/sapphire). Highly doped Cr^{2+}:ZnSe/Sapphire thin film sample was also used to demonstrate passive Q-switching of Er:YAG laser operating at 1.645 μm.

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1. Introduction

Transition metal doped II-VI semiconductor laser crystals have stimulated extensive interest in the scientific community as active materials for tunable middle infrared (mid-IR) lasers operating over the 2–5 μm range [1–5]. Of particular interest are Cr^{2+} ions incorporated in ZnSe crystals (Cr^{2+}:ZnSe) featuring intra-center 2–3 μm transitions between the crystal-field-split ground $^5T_2$ and first excited ^5E states [6]. The availability of affordable low loss polycrystalline ZnSe, and the ability to fabricate high quality uniformly doped Cr^{2+}:ZnSe crystals via post-growth thermal diffusion make them the gain materials of choice for development of high power mid-IR lasers, broadly tunable over the 2–3 μm spectral range [3,4]. Cr^{2+}:ZnSe lasers documented in the literature operate at room temperature in continuous wave (CW) regime with a slope efficiency and output power greater than 60% and 10 W, respectively [3,7–9]. Further advancements in power scaling require thorough thermal management of the active element. Among different thermal management and beam quality control approaches, waveguide laser geometry design holds a lot of promise. The majority of reported Cr^{2+}:ZnSe lasers relied on bulk gain material fabrication methods [3] and did not offer waveguiding of the optical field.

Thin film deposition techniques are promising for fabrication of Cr^{2+}:ZnSe waveguide laser and electroluminescent structures. Previously, mid-IR electroluminescence has been reported for bulk [10–12] and thin film [13,14] Cr^{2+}:ZnSe samples. Thin film fabrication technique offers a versatile fabrication method minimizing layer interface roughness and enabling the development of the entire structure in situ in vacuum. Several Cr^{2+}:ZnSe thin film structures featuring room temperature photoluminescence (PL) from the optically active Cr^{2+}ions have been fabricated by means of molecular beam epitaxy [15], radio frequency magnetron sputtering [16] and pulsed laser deposition (PLD) [17]. Building on the success of PLD growth of Cr^{2+}:ZnSe film, we report here the first PLD grown Cr^{2+}:ZnSe planar waveguide structure exhibiting mid-IR lasing. In addition, highly doped PLD grown Cr^{2+}:ZnSe thin film was used for the first time as a saturable absorber for passive Q-switching of Er:YAG laser operating at 1645 nm.

2. Thin film deposition

Chromium doped ZnSe films were deposited on epi-ready double-side-polished *c*-plane (0001) oriented sapphire substrates (Cr^{2+}:ZnSe/sapphire) by means of laser ablation of a solid chromium doped ZnSe target. Since the index of refraction of ZnSe is n_{ZnSe} ~2.44 and that of sapphire is $n_{sapphire}$ ~1.73 in the 2–3 μm region, the structure air/Cr^{2+}:ZnSe/sapphire should provide waveguiding-confinement of the optical field within the Cr^{2+}:ZnSe active region. Laser ablation targets were fabricated in-house by manually mixing powders of ZnSe (99.99%

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purity) and CrSe (99.9% purity) in the appropriate weight percentages to obtain the desired chromium doping concentrations within the targets. The mixed powders were compressed into a solid pellet approximately 1 inch in diameter which was then vacuum sealed at 10^{-4} Torr in a quartz ampoule and annealed at 1000 °C for 10 days. The target was then ablated by a KrF excimer laser at 2 J/cm^2. The details of the deposition procedure are described in Ref [17].

3. Experimental results

3.1 Characterization of the crystalline quality of deposited films

The surface roughness of the deposited films was analyzed with atomic force microscopy (AFM) in contact mode. The AFM images of a 10 μm thick Cr^{2+}:ZnSe film deposited on sapphire is shown in Fig. 1. An image of a 50 × 50 μm scan area is depicted in Fig. 1(a) and Fig. 1(b) shows a closer 5 × 5 μm scan area of the film's surface. Both images showed a surface area root mean square (RMS) roughness of less than 7 nm. On average, the area RMS roughness of all the Cr^{2+}:ZnSe film surfaces deposited by PLD was less than 10 nm which indicates a very good smooth surface morphology.

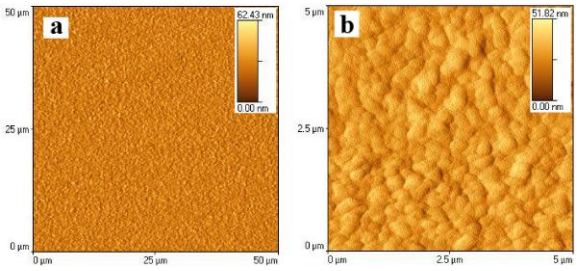

Fig. 1. AFM images of (a) 50 × 50 μm scan area and (b) 5 × 5 μm scan area of a 10 μm thick Cr^{2+}:ZnSe film deposited on sapphire. The surface areas RMS roughness was smaller than 10 nm.

The crystalline quality of the films was determined using X-ray diffraction (XRD) with CuK_α radiation of 1.5418 Å and room temperature Raman scattering. In Fig. 2(a) the θ-2θ XRD scans of Cr^{2+}:ZnSe/sapphire films deposited at various deposition temperatures are shown. All XRD patterns obtained indicated that the films deposited were polycrystalline ZnSe in a cubic (zinc blende) structure. The XRD patterns showed the (111), (220), and (311) cubic ZnSe planes perpendicular to the substrates surface along with reflections from the substrate (0006) plane. At growth temperatures lower than 475 °C, there appears to be some mixture of ZnSe hexagonal crystallites within the films as evident from the ZnSe wurtzite (100) XRD peak seen around 2θ~25.9°. As growth temperature increases, the degree of hexagonality of the films diminished as the XRD peak corresponding to the ZnSe wurtzite structure gradually disappeared. This suggests that at growth temperatures of 600 °C, the films deposited were purely cubic as oppose to a mixed-polytype structure of cubic and hexagonal ZnSe structures seen in films deposited at lower temperatures. The full width at half maximum (FWHM) of the (111) and (311) peaks decreased from 0.852° to 0.472° and 1.226° to 0.683°, respectively, with increasing deposition temperature from 350 °C to 600 °C. The narrowing of the FWHM with increasing growth temperatures indicated the formation of higher quality crystalline ZnSe lattices within the films.

The Raman spectra for Cr^{2+}:ZnSe films deposited on sapphire at various temperatures are shown in Fig. 2(b) with comparison to a bulk ZnSe polycrystal Raman spectrum. The Raman measurements are consistent with XRD analysis that the crystal quality of the films was better at deposition temperatures above 425 °C. This assertion is supported by the well resolved intense peaks at 205 cm^{-1} and 250 cm^{-1} attributed to the transversal optical (TO) and longitudinal optical (LO) phonon modes of ZnSe, respectively [18], indicating the growth of high crystalline quality ZnSe lattice in samples deposited at the higher temperatures. Also

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visible at higher growth temperatures is a broader weak transversal acoustic (2TA) ZnSe phonon mode around 140 cm^{-1}. At growth temperatures below 475 °C, only a much broader LO phonon mode of ZnSe is distinguishable from the Raman spectra indicating that the films deposited at these temperatures were partially amorphous.

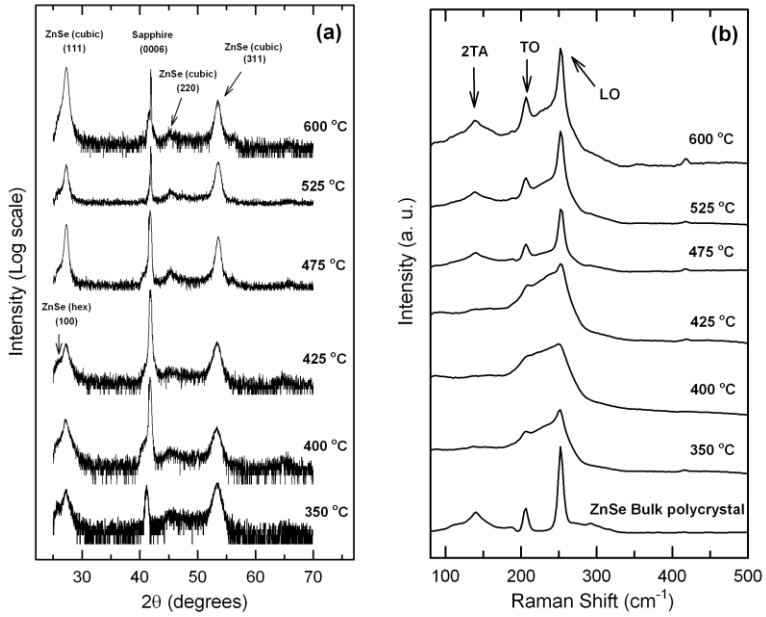

Fig. 2. (a) θ-2θ XRD patterns and (b) Raman spectra of Cr^{2+}:ZnSe films deposited on sapphire substrate at various growth temperatures.

3.2 Optical characterization of deposited films

The optical transmission spectrum over 1000–270`0 nm spectral range of a typical Cr^{2+}:ZnSe/sapphire film deposited at a substrate temperature of 400 °C is shown in Fig. 3. The interference pattern revealed a good homogeneous film. By using the transmission oscillation period with the index of refraction of n_{ZnSe} ~2.44, the thickness of each deposited film was determined. The absorption dip seen near 1700 nm resulted from the $^5T_2 \rightarrow {}^5E$ transition of Cr^{2+} ions. Using the obtained film thickness and assuming a peak absorption cross section of $\sigma \sim 1 \times 10^{-18}$ cm^2 for the Cr^{2+} ions similar to that measured through spectroscopic experiments in bulk samples [19,20], the Cr^{2+} ion concentration in each film was estimated. The Cr^{2+} concentrations were determined in this manner for several Cr^{2+}:ZnSe/sapphire films deposited at various substrate temperatures from 375 °C to 575 °C using the same laser ablation target. The overall variation in calculated dopant ion concentration was less than 15% over the analyzed films. This result indicates that the ablation targets produced in-house were fairly uniformly doped and that there was adequate transfer of stoichiometry from target to film during PLD growth independently of substrate temperature.

Figure 4(a) shows the normalized room temperature mid-IR PL spectra obtained under the same experimental conditions from Cr^{2+}:ZnSe/sapphire sample of approximately 10 μm thick films with Cr^{2+} dopant concentrations of (i) 6×10^{19} cm^{-3}, (ii) 3×10^{19} cm^{-3}, (iii) $< 2 \times 10^{18}$ cm^{-3} (estimated from CrSe powder wt. % in target since Cr^{2+} absorption was below spectrometer detection limit), and (iv) 6×10^{18} cm^{-3}. The PL measurements were obtained from direct optical excitation of the Cr^{2+} ions using a 1532 nm CW Er doped fiber laser. The PL emission signal was collected with a CaF_2 lens and detected by an Acton Research Spectra-Pro 300i (ARC-300i) spectrometer equipped with a liquid nitrogen cooled InSb

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detector/lock-in-amplifier combination. The signal intensities were collected over the 2–3 μm spectral range, while the scattered pump radiation was filtered using a 2 μm germanium filter. All PL spectra obtained were typical for Cr^{2+}:ZnSe crystals and resulted from the $^5T_2 \rightarrow {}^5E$ Cr^{2+} intra-center transition. The PL signal intensities obtained from samples with concentrations greater that 1×10^{19} cm^{-3} were significantly weaker than the PL signal obtained from samples with dopant concentration below 1×10^{19} cm^{-3}. This was attributed to concentration quenching within the highly doped samples ($N_{Cr} > 1 \times 10^{19}$ cm^{-3}) resulting in the lifetime of emission being much shorter than in lower doped films. This result is in accordance with previous studies [19,21] showing that concentration quenching in bulk Cr^{2+}:ZnSe begins above the critical dopant ion concentration of $N_{Cr} \sim 10^{19}$ cm^{-3} where the excited state Cr^{2+} lifetime drastically decreases. However, depending on the application, low-doped or highly-doped ZnSe films may be desirable. In the case of CW laser operation of Cr^{2+}:ZnSe thin films, a low dopant concentration with a longer lifetime of emissions is desirable as opposed to the case of saturable Cr^{2+}:ZnSe thin films for potential passive Q-switching and passive mode-locking where highly-doped films are more favorable.

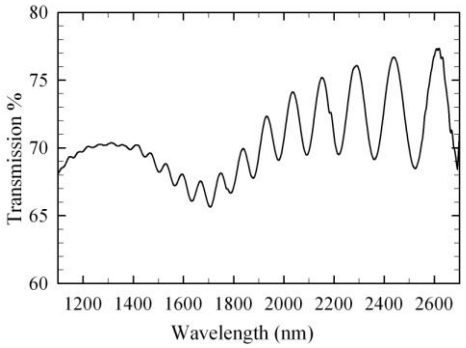

Fig. 3. The transmission spectrum of a 7.5 μm thick Cr^{2+}:ZnSe/sapphire sample with Cr^{2+} concentration of ~6×10^{19} cm^{-3} deposited at 400 °C.

Mid-IR planar waveguide lasing in gain-switched mode was demonstrated using a Cr^{2+}:ZnSe film of thickness 7.5 μm deposited on sapphire at a substrate temperature of 400 °C. From the sample's transmission spectrum shown in Fig. 3, the Cr^{2+} concentration was estimated to be 6×10^{19} cm^{-3}. The Cr^{2+}:ZnSe/sapphire sample was optically pumped at room temperature by 1560 nm D_2-Raman shifted Nd:YAG laser radiation with a pulse duration of 5 ns. The pump beam of diameter ~3.1 mm was directed perpendicularly to the front facet of the film. The resulting mid-IR emission spectrum along the waveguided direction of the Cr^{2+}:ZnSe/sapphire sample edge facet was collected with a CaF_2 lens and detected with an ARC-300i spectrometer and a liquid nitrogen cooled InSb detector. The PL kinetics were measured using a digital oscilloscope and the PL intensity was processed through a boxcar averager. At low pumped energies (Fig. 4(b)(i)), the typical PL measurement similar to the spectral characteristics seen in Fig. 4(a) was obtained from the sample. The kinetics of the luminescence from the film was measured to be shorter than the InSb detector response time (~500 ns) and significantly shorter that Cr^{2+} PL lifetime in low concentration doped ZnSe bulk samples (~5.4 μs). This is understandable since the film dopant concentration of 6×10^{19} cm^{-3} is much greater than the critical dopant concentration $N_{cr} \sim 1 \times 10^{19}$ cm^{-3} that marks the beginning of concentration quenching. However, for Cr^{2+} concentration of 6×10^{19} cm^{-3}, the estimated lifetime of luminescence should be longer than the pump pulse duration of 5 ns used for gain-switched excitation and a high concentration of the active ions should provide high amplification in the planar waveguide. Figure 4(b)(ii) shows that pumping above threshold resulted in the appearance of an intense, much narrower stimulated emission with a central peak around 2600 nm, much different than the typical PL seen at lower pumped energies. This

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stimulated emission is attributed to the planar waveguiding of the optical field set up by the lower index of refraction of the sapphire substrate with respect to the Cr^{2+}:ZnSe film. The lasing peak at 2600 nm is shifted to the longer wavelengths with respect to the PL peak around 2100 nm. This shift resulted from the trade-off between maximum emission cross-section at ~2400 nm [22] (see Fig. 4(b)(iii)) and optical losses in the waveguide due to non-saturated chromium absorption (see Fig. 4(b)(iv)). Figure 4(c) depicts the dependence of the intensity of Cr^{2+}:ZnSe planar waveguide emission at 2600 nm versus pump energy. The output-input characteristics clearly show the threshold-like behavior of the output signal with a threshold pumped energy density for planar lasing around 0.11 J/cm^2.

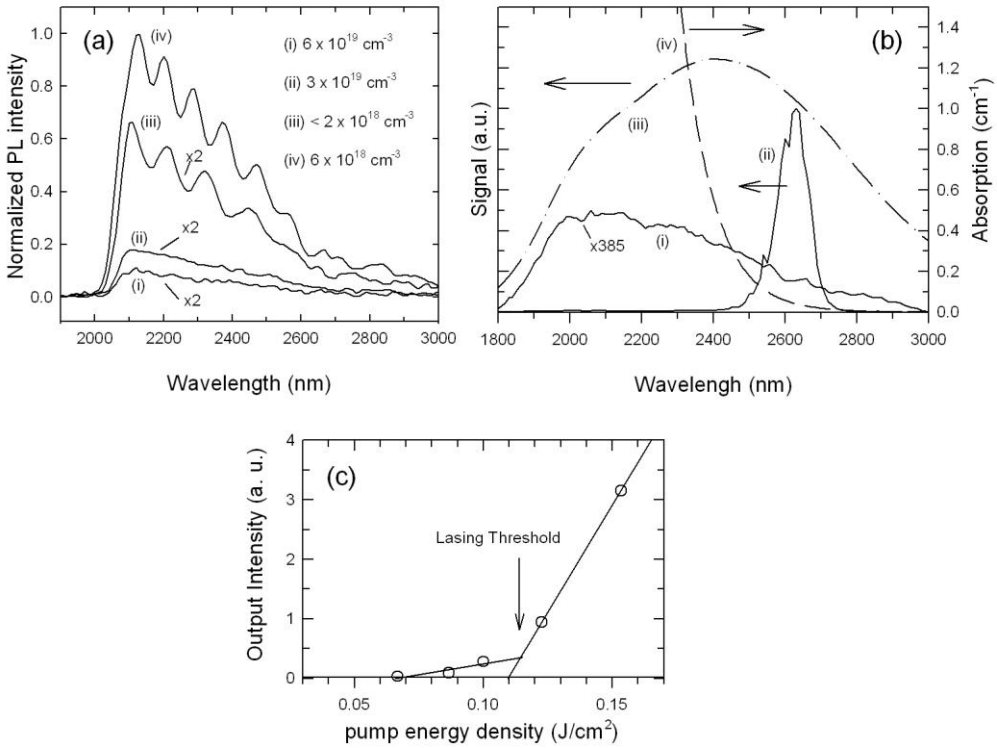

Fig. 4. (a) Normalized room temperature PL spectra measured from Cr^{2+}:ZnSe/sapphire films with various dopant concentrations deposited at a substrate temperature of 475 °C. (b) Emission spectra measured (i) below and (ii) above laser threshold from Cr^{2+}:ZnSe film of Cr^{2+} concentration of 6×10^{19} cm^{-3} deposited on sapphire substrate.(iii) Gain (in arbitrary units) and (iv) absorption (in cm^{-1}) spectra of Cr^{2+}:ZnSe with Cr^{2+} concentration of 6×10^{19} cm^{-3} from Ref [22]. (c) The output-input signal characteristics of the Cr^{2+}:ZnSe/sapphire film deposited which depicts a lasing threshold energy density of approximately 0.11 J/cm^2.

Highly-doped Cr^{2+}:ZnSe thin-film could be potentially used for passive Q-switching of the cavities of Er, Tm, and Ho lasers operating over 1.5–2.1 μm spectral range. Experiments were conducted to study CW and passively Q-switched Er:YAG laser pumped by an Er-fiber laser. A 5 mm diameter antireflection coated Er:YAG rod with Er concentration of 0.5 atomic % was used for the experiments. A 20 W Er fiber laser (ELR-20 IPG Photonics Corporation) with polarized oscillation at 1532 nm wavelength was used as the pump source. The optical scheme for the laser experiments is shown in Fig. 5(a). The features of the input-output characteristics of the passively Q-switched Er:YAG laser using a 6×10^{19} cm^{-3} doped

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Cr^{2+}:ZnSe thin film sample, and (i) 80% and (ii) 95% reflectivity of the output couplers is shown in Fig. 5(b). The output power obtained reached the level of 100 mW at 3W pumped power. For comparison, the CW oscillation of the Er:YAG laser with 80% output coupler was measured in the same cavity without the Cr^{2+}:ZnSe sample (Fig. 5(b)(iii)). As one can see from Fig. 5(b) (i) and (iii), the slope efficiencies obtained were similar. However, the use of Cr^{2+}:ZnSe thin-film Q-switcher results in laser threshold increasing from 0.6 W to 1.5 W for 80% reflectivity of the output coupler. Figure 5(c) represents a characteristic train of Q-switched Er:YAG pulses at 1.645μm obtained for input power of 1.5 W and 95% reflectivity of the output coupler. The emission spectrum shows that the passively Q-switched Er:YAG laser operates at repetition rate of 21 kHz and features an output pulse energy on the order of magnitude of several microjoules. This successful Q-switch demonstration exemplify the potential for Cr^{2+}:ZnSe thin films to be used as possible passive mode-lockers of Er:YAG laser by increasing the Cr^{2+} concentration to the level higher than that used in the current Q-switch laser experiments. By increasing the dopant concentration, the excited state lifetime of the Cr^{2+} ions continues to drastically diminish as concentration quenching becomes dominant. This short-lived excited state of the Cr^{2+} ions is desirable for passive mode-lock applications involving Er, Tm, and Ho lasers operating in the 1.5–2.1 μm.

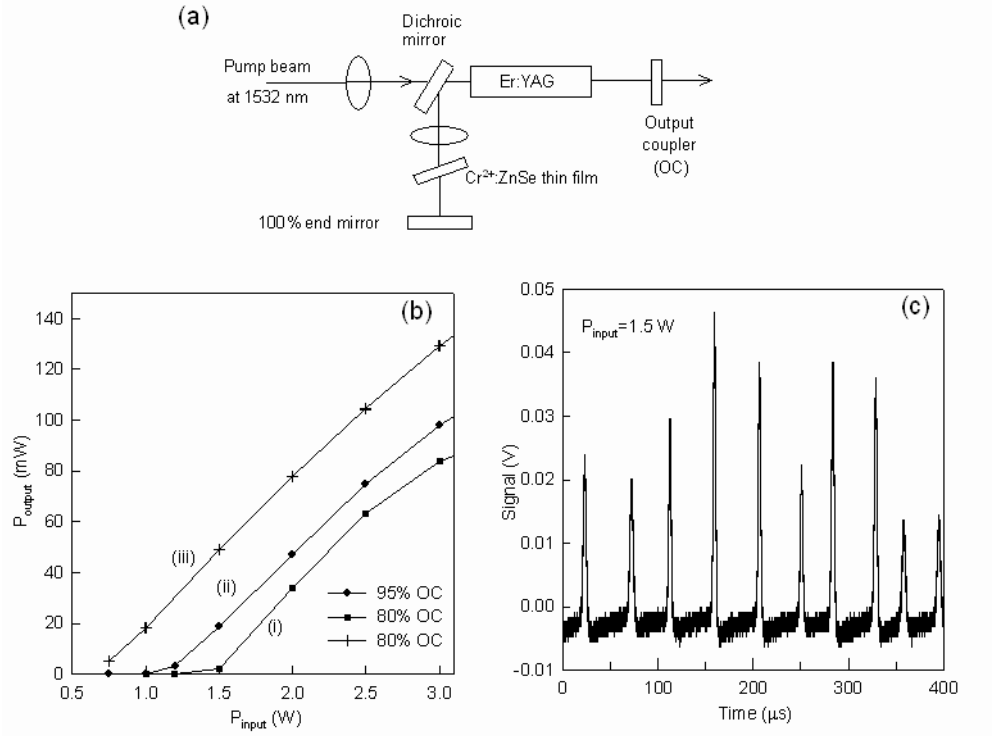

Fig. 5. (a) The optical scheme used for the laser experiments. (b) Comparative Er:YAG input-output laser characteristics in Cr^{2+}:ZnSe thin film Q-switched regime with (i) output coupler reflectivity R = 80%, (ii) R = 95%, and (iii) free running (R = 80%) regimes of operation. (c) The output pulses from passively Q-switched Er:YAG laser at pump power of 1.5 W with 95% output coupler.

4. Conclusions

In summary, we report that highly doped Cr^{2+}:ZnSe thin films can be used as saturable absorbers for passive Q-switching of Er:YAG laser cavity with potential to be used as a passive mode-locker of Er, Tm, and Ho lasers operating over 1.5–2.1 μm spectral region. The first demonstration of 2.6 μm mid-IR lasing in Cr^{2+}:ZnSe waveguide structures grown by PLD

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is also reported. The transition metal doped II-VI planar waveguide laser structures could be attractive for chip-integrated optical laser design with diode laser excitation and electronic control of tunability required for highly sensitive portable opto-chemical sensors.

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The work reported here partially involves intellectual property developed at the University of Alabama at Birmingham. This intellectual property has been licensed to the IPG Photonics Corporation. The UAB co-authors declare competing financial interests.

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