

High Repetition-Rate 2.3–2.7 μm Acousto-Optically Tuned Narrow-Line Laser System Comprising Two Master Oscillators and Power Amplifiers Based on Polycrystalline $\text{Cr}^{2+}:\text{ZnSe}$ with the 2.1 μm $\text{Ho}^{3+}:\text{YAG}$ Pulsed Pumping

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Article

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Abstract: High average power narrow-linewidth tunable solid-state lasers in the wavelength region between 2 to 3 μm are attractive light sources for many applications. This paper reports a narrow-linewidth widely-tunable laser system based on the polycrystalline $\text{Cr}^{2+}:\text{ZnSe}$ elements pumped by the repetitively-pulsed 2.1 μm $\text{Ho}^{3+}:\text{YAG}$ laser operating at the tens of kilohertz pulse rate. An advanced procedure of ZnSe -element doping and surface improvement was applied to increase the laser-induced damage threshold, which resulted in an increase of the output power of the $\text{Cr}^{2+}:\text{ZnSe}$ laser system. The high-average-power laser system comprised double master-oscillators and power amplifiers: $\text{Ho}^{3+}:\text{YAG}$ and $\text{Cr}^{2+}:\text{ZnSe}$ laser oscillators, and $\text{Ho}^{3+}:\text{YAG}$ and $\text{Cr}^{2+}:\text{ZnSe}$ power amplifiers. The output wavelength was widely tuned within 2.3-2.7 μm by an acousto-optical tunable filter inside the $\text{Cr}^{2+}:\text{ZnSe}$ master oscillator cavity. The narrow-linewidth operation at the pulse repetition rate of 20-40 kHz in a high-quality beam with the average output power up to 9.7 W was demonstrated.

Keywords: Cr^{2+} -doped chalcogenides; $\text{Cr}^{2+}:\text{ZnSe}$ polycrystals; gain switched lasers; $\text{Ho}^{3+}:\text{YAG}$ laser pumping; wavelength tuning; acousto-optical tunable filter; narrow-linewidth and wide wavelength tuneability; high-repetition-rate high-average-power mid-IR laser operation

1. Introduction

High average power tunable solid-state lasers in the wavelength range from 2 to 3 μm are attractive light sources for many applications, such as medical surgery and lasertripsy, laser processing, environmental remote sensing, wireless energy transmission technology, pumping mid-infrared optical parametric oscillators and solid-state lasers based on Fe^{2+} -doped crystals [1–7]. Cr^{2+} -doped zinc selenide ($\text{Cr}^{2+}:\text{ZnSe}$) crystals are among the most attractive active media for high-average power lasers in the 2-3 μm wavelength range due to their wide tunability, broad absorption bands, and large stimulated-emission cross section [8–13]. Gain-switched operation in nanosecond pulse mode of the $\text{Cr}^{2+}:\text{ZnSe}$ lasers and a wide wavelength tuning of their outputs were previously reported [10–17]. Various wavelength tuning techniques have been used in $\text{Cr}^{2+}:\text{ZnSe}$ lasers: intracavity wavelength-selective elements such as diffraction gratings

[11,12,19–21], birefringent Liot filters [16,22], prisms [23,24], acousto-optic tunable filters (AOTF) [10,14,17,18,25,26], liquid crystal etalon [27], or injection seeding [28]. The AOTF enabled the Cr²⁺:ZnSe laser to electronically tune every pulse in the broad-wavelength tuning range [29]. In the narrow-line widely-tunable Cr²⁺:ZnSe laser systems pumped by the Tm³⁺:YAP lasers two pulsed regimes were reported: the pulse energy of up to 50 mJ in slow pulse regime at the pulse repetition rate (PRR) of 10 Hz [18], or the average power of up to 5.1 W in the faster operating regime at 7 kHz PRR [15]. However, the scaling of the average and peak power and the pulse energy of the laser systems is hindered by the laser induced damage (LID) of both polycrystalline and single-crystalline Cr²⁺:ZnSe [12,14,15,18,30–32].

In this paper we present a high-average-power high-PRR AOTF-tunable solid-state laser system based on the polycrystalline Cr²⁺:ZnSe pumped at 2091 nm by the repetitively-pulsed fiber-laser-pumped Ho³⁺:YAG lasers. The laser system architecture comprised a tunable Cr²⁺:ZnSe master oscillator and Cr²⁺:ZnSe power amplifier pumped with 10-40 kHz PRR by a Ho³⁺:YAG mater oscillator and a Ho³⁺:YAG power amplifier, respectively. An intracavity AOTF provided 2.3-2.7 μm tunability of narrow-band generation of the Cr²⁺:ZnSe laser pulses. An advanced procedure of Cr²⁺:ZnSe-element doping and surface improvement was applied to increase the LID threshold, which resulted in an increase of the output power of the Cr²⁺:ZnSe laser system.

2. Cr²⁺:ZnSe Active Elements

The Cr²⁺:ZnSe active elements were made from laser-quality zinc selenide (ZnSe) obtained by chemical vapor deposition (CVD) and cut into blanks with a cross-section of up to 4.5×4.5 mm². The length of the blanks, and thus of the laser elements, varied from 15 mm to 33 mm. A chromium film was vacuum deposited on the side surface of the blanks. Subsequently, the samples were placed in a quartz ampoule, washed with high purity argon, and vacuumed 5 times. Metallic zinc (Zn) was also added to the ampoule; the amount of zinc was adjusted to equalize the Zn vapor pressure at the annealing temperature to atmospheric pressure. The ampoule was vacuumed, sealed, and held in a muffle furnace at 1050°C for 7 days. The ampoule was then unsealed and the elements were processed in a hot isostatic press for 20 h at 1100°C in argon to restore the ZnSe stoichiometry (to remove excess Zn). In preliminary testing the higher LID-threshold was found in Cr²⁺:ZnSe samples annealed in argon (Ar) atmosphere compared to samples annealed in Zn atmosphere or with hot isostatic pressing [32]. However, annealing in a sealed ampoule does not provide confidence that the stoichiometry of the material has been restored. In this work hot isostatic pressing was used, but compared to the procedure described in [32], the samples were placed in a graphite container to reduce contamination. As a result of this doping procedure, chromium ions (Cr²⁺) were uniformly distributed throughout the sample volume and the elements had an increased LID threshold. The side surfaces of the samples were finely ground to prevent stray reflections and to reduce the probability of parasitic generation; all right angles were chamfered at ~0.2 mm.

The Cr²⁺-ion doping concentration (N_d) and element length (L) were chosen so that the single-pass small-signal absorption coefficient ($\alpha_{\text{abs}} \times L = \sigma_{\text{abs}} \times N_d \times L$, where σ_{abs} is the absorption cross section) of the Cr²⁺:ZnSe element at 2091 nm was in the range of 1-1.5. The Cr²⁺-ion doping concentration was kept low to avoid strong thermal aberrations in the active elements. The length of Cr²⁺:ZnSe elements was optimized based on the preliminary absorption measurements; the final length of the active elements with different Cr²⁺-ion doping concentration was in the range of 14 to 32 mm. Assuming the absorption cross section of Cr²⁺:ZnSe at 2091 nm of $\sigma_{\text{abs}} = 1.4 \times 10^{-19} \text{ cm}^2$, the final concentration of the Cr²⁺ ions in the laser elements was estimated to be $N_d = 3\text{--}6 \times 10^{18} \text{ cm}^{-3}$.

The cross section of the active element was 3×3 mm² or 3.5×3.5 mm² in the 14-20 mm long samples 4×4 and 4.3×4.3 mm² in the longer active elements. A broadband (2.0-2.7 μm) antireflection coating of yttrium fluoride and zinc sulfide layers was applied on the working surfaces of the active elements. A sample active element is shown in Figure 1.

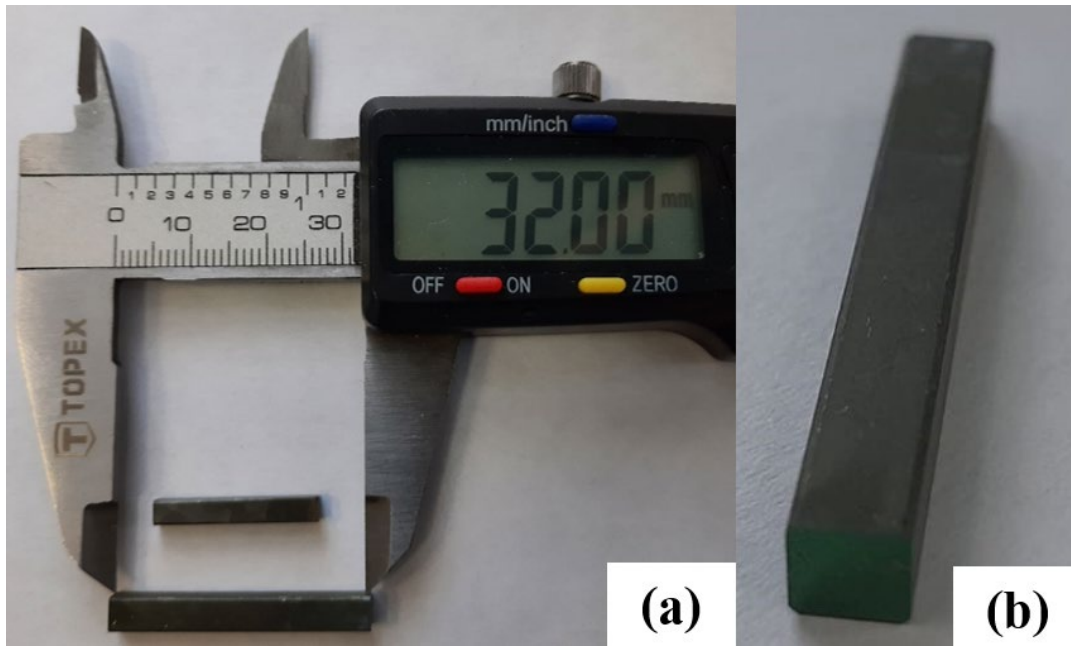


Figure 1. Photographs of the active elements made from polycrystalline $\text{Cr}^{2+}:\text{ZnSe}$: side view with measured length indication (a), the front side with antireflection coating (b).

The absorption spectrum of $\text{Cr}^{2+}:\text{ZnSe}$ crystals enables the low-quantum defect pumping of the 2.3-2.7- μm laser using a $\text{Ho}^{3+}:\text{YAG}$ laser at 2091 nm (Figure 2). At 2091 nm the absorption cross section σ_{abs} of the $\text{Cr}^{2+}:\text{ZnSe}$ crystal is less than the emission cross section σ_{em} [8,12]. Since the Cr^{2+} ion in a ZnSe crystal field is a 4-level system with fast relaxation in the vibronic sublevels [8,19], the sum of the populations of the 5E upper state N_{up} at the 2091-nm pumping and the 5T₂ ground state, N_{g} , effectively equals to the doping concentration of Cr^{2+} ions in the active sites N_{da} (neglecting an excited state absorption and an up-conversion). In this case the effective gain cross section $G(\lambda)$ of the crystal at a wavelength λ can be calculated for a given inversion fraction β using the following expression:

$$G(\lambda) = \sigma_{\text{em}}(\lambda) \times \beta - \sigma_{\text{abs}}(\lambda) \times (1 - \beta) - \gamma(\lambda)/N_{\text{da}}, \quad (1)$$

where $\beta = N_{\text{up}}/N_{\text{da}}$, $\gamma(\lambda)$ is the passive losses at a wavelength λ .

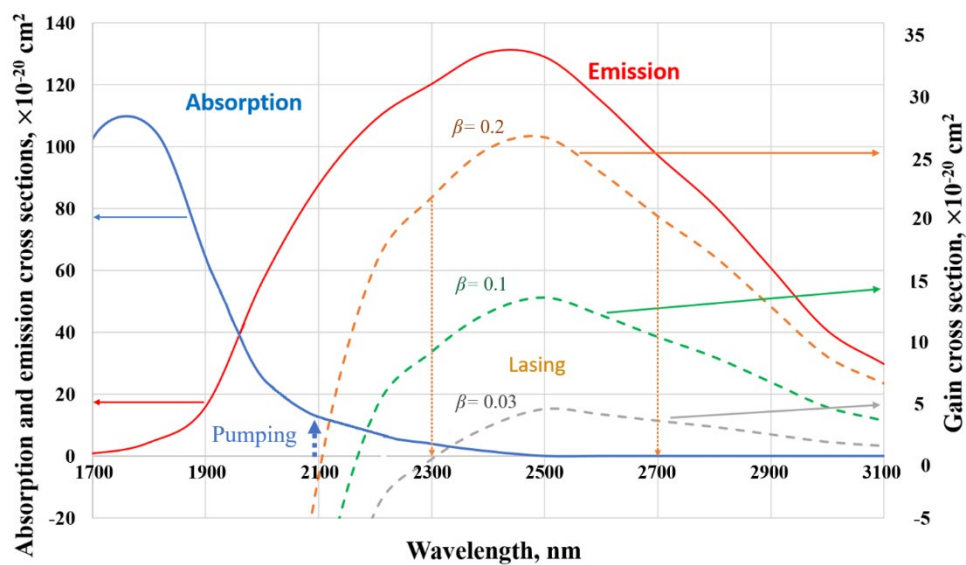


Figure 2. Absorption (blue) and emission (red) cross sections of the $\text{Cr}^{2+}:\text{ZnSe}$ crystal. The dashed lines of different colors indicate the calculated gain cross section, G , at zero passive losses ($\gamma = 0$) and for the different value of the inversion fraction β : 0.03 (gray), 0.1 (green) and 0.2 (brown). The violet and

red dotted pointers indicate the wavelengths of the pumping $\text{Ho}^{3+}:\text{YAG}$ laser and the operating $\text{Cr}^{2+}:\text{ZnSe}$ laser, respectively.

3. Laser System Architecture

The laser system comprised the double master oscillator – power amplifier: a $\text{Ho}^{3+}:\text{YAG}$ master oscillator pumping a tunable $\text{Cr}^{2+}:\text{ZnSe}$ master oscillator, and a $\text{Ho}^{3+}:\text{YAG}$ power amplifier pumping a $\text{Cr}^{2+}:\text{ZnSe}$ power amplifier (Figure 3).

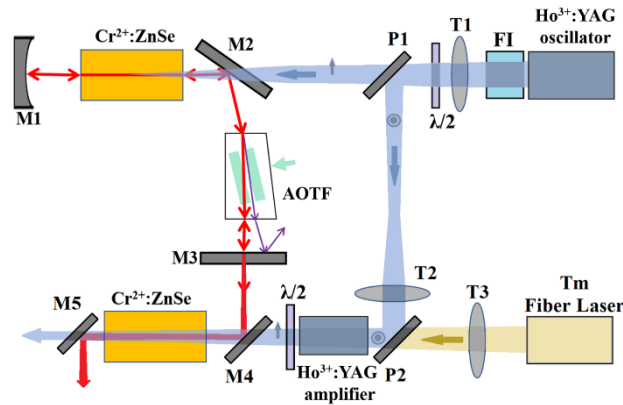


Figure 3. Schematics of the $\text{Cr}^{2+}:\text{ZnSe}$ laser system: M1-M5 are the mirrors, P1-P2 are the polarizers, T1-T3 are the Galilean telescopes, FI is the Faraday isolator, AOTF is the acousto-optical tunable filter; $\text{Cr}^{2+}:\text{ZnSe}$ and $\text{Ho}^{3+}:\text{YAG}$ are the laser and amplifier active elements. The blue pointer and the blue dot on the beams indicate the polarization direction. The red, blue, yellow and violet lines indicate the $\text{Cr}^{2+}:\text{ZnSe}$ laser beam, the $\text{Ho}^{3+}:\text{YAG}$ laser beams, the Tm fiber laser beam, and the AOTF undeflected wave, respectively. The white-green grating indicates acousto-optical wave inside the AOTF.

3.1. $\text{Ho}^{3+}:\text{YAG}$ Laser Oscillator

An in-house made repetitively-pulsed $\text{Ho}^{3+}:\text{YAG}$ laser at 2091 nm was used as the pump source of the $\text{Cr}^{2+}:\text{ZnSe}$ laser [33]. The $\text{Ho}^{3+}:\text{YAG}$ laser was pumped, in turn, by a CW Tm-doped fiber laser at 1908 nm (“NTO IRE-Polus”, Fryazino, Russia). The repetitively-pulsed regime was achieved by active Q-switching the cavity with a quartz acousto-optical modulator (“NII Polyus”, Moscow, Russia). The operation wavelength of ~2091 nm was selected and stabilized by a rotated silica etalon of 90- μm thickness (Figure 4a). The Q-switched laser operated with PRR varied from 10 to 50 kHz. The FWHM pulse width (τ) of 15–45 ns depended on the PRR and the pump power.

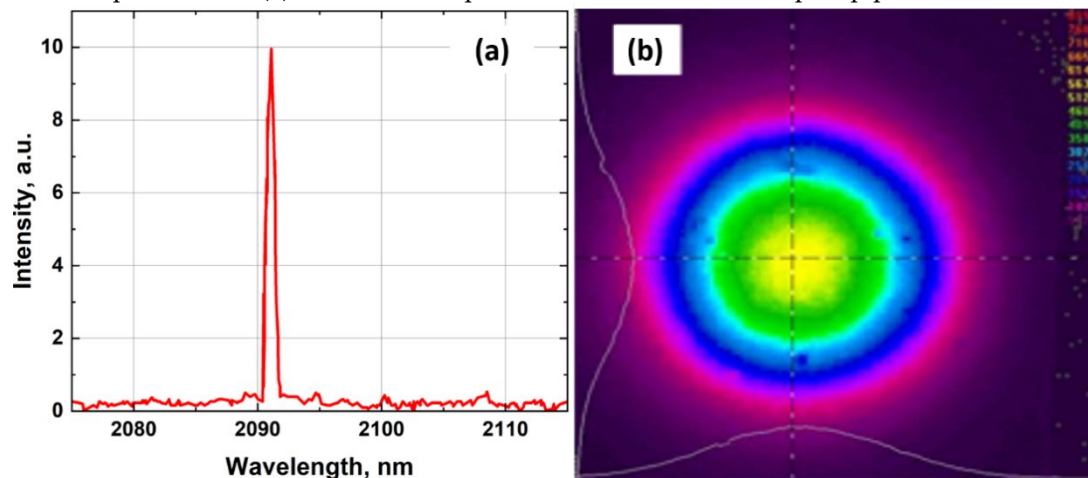


Figure 4. Operating wavelength measured by OSA203 spectrum analyzer (Thorlabs, USA) (a) and beam profile (b) of the $\text{Ho}^{3+}:\text{YAG}$ laser.

The average power of the Ho^{3+} :YAG laser measured by a power meter (Coherent PM 10 with FieldMax detector, Coherent, USA) was up to 34 W at 30 kHz PRR (at 55-W fiber laser pump power) in a linearly polarized high-quality Gaussian beam. The laser beam was monitored using a Pyrocam IV camera (Ophir-Spiricon, North Logan, USA) (Figure 4b). The M2 parameter (determined by ISO 11146-1:2021 standard with the knife-edge method [34]) was less than 1.2-1.3.

To avoid the influence of a back reflection on the Ho^{3+} :YAG laser, the Faraday isolator (EOT, Traverse City, USA) was used (Figure 3). A Galilean telescope (T1), a half-wavelength plate ($\lambda/2$) and a polarizer (P1) were placed in the Ho^{3+} :YAG laser beam path; the last two optical elements controlled the power of the linearly-polarized 2091 nm output and provided two polarization separation. Finally, the horizontally-polarized beam from the Ho^{3+} :YAG laser was focused into a Cr^{2+} :ZnSe active element, but the vertically polarized beam at 2091 nm propagated to the Ho^{3+} :YAG laser amplifier.

3.2. Ho^{3+} :YAG Power Amplifier

The power amplifier of the beam at 2091 nm was based on Ho^{3+} :YAG rods ("NII Polyus, Moscow, Russia) with the length of 50 mm or 65 mm and diameter of 5 mm or 6 mm, respectively. The Ho^{3+} -ion concentration was 0.5 at. %. The end faces of the Ho^{3+} :YAG rods were AR coated at 2050-2150 nm. The Ho^{3+} :YAG rod was pumped by a CW Tm-doped fiber laser at 1908 nm ("NTO IRE-Polus", Fryazino, Russia) with the power up to 53 W. The diameter of the pumping beam at 1908 nm in the Ho^{3+} :YAG crystal was tuned by the Galilean telescope T2 to be equal to ~ 0.8 -0.9 mm, when measured at e-2 peak intensity by the knife-edge method [34]. The diameter of the repetitively-pulsed amplified beam at 2091 nm was tuned by the Galilean telescope T3 to be equal also to ~ 0.8 -0.9 mm.

At the repetitively-pulsed input power of 3.1 W, the maximum output power of the Ho^{3+} :YAG amplifier based on a 50 mm long crystal was ~ 5.5 W. When the input power was increased to 21.6 W the output power increased to 36 W (Figure 5a). The maximum output of the Ho^{3+} :YAG amplifier based on a 65 mm long crystal was 7.5 W at 3.1 W input power, and with the input power of 21.6 W output power increased to 43 W (Figure 5b).

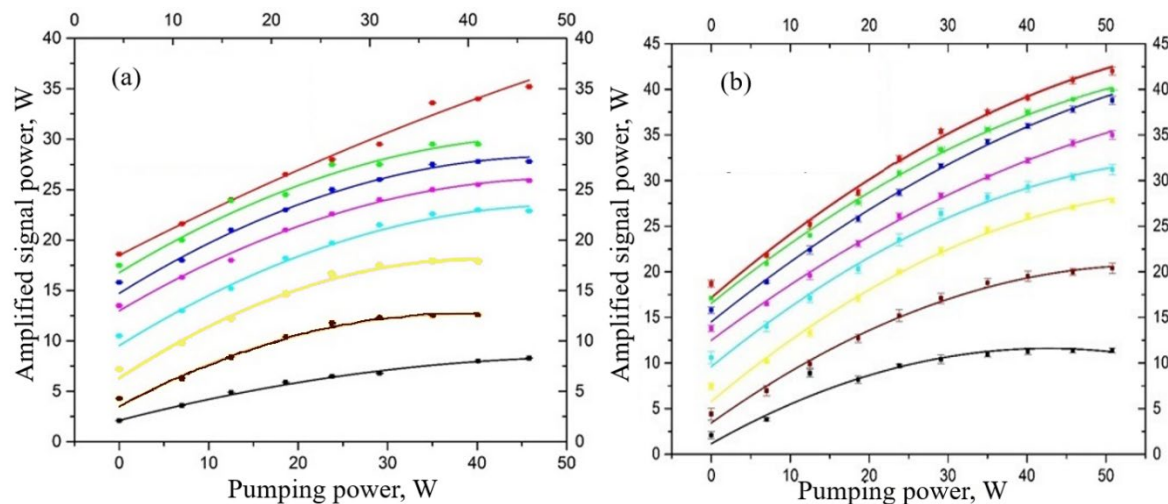


Figure 5. The amplifier power of the repetitively-pulsed beam at 2091 nm vs CW power of the fiber laser beam at 1908 nm, at 20 kHz and input power of 3.1 W (black), 6 W (brown), 9.2 W (yellow), 12.3 W (white-blue), 13.6 W (violet), 18.2 W (dark blue), 20 W (green), 21.6 W (red) for the Ho^{3+} :YAG rod with a length of 50 mm (a) or 65 mm (b).

The decrease in gain of the 65-mm long amplifier with increasing input pulsed power can be explained by the saturation effect (Figure 6). With PRR increasing, the gain increased at low input pulsed power, but the same gain decreased at high input pulsed power (Figure 6b). The gain increase in a large signal at higher PRR can be explained by the influence of the thermal lens and the electronic self-focusing effect, which reduced the diameters of the pumping and amplified beams in the laser amplifier.

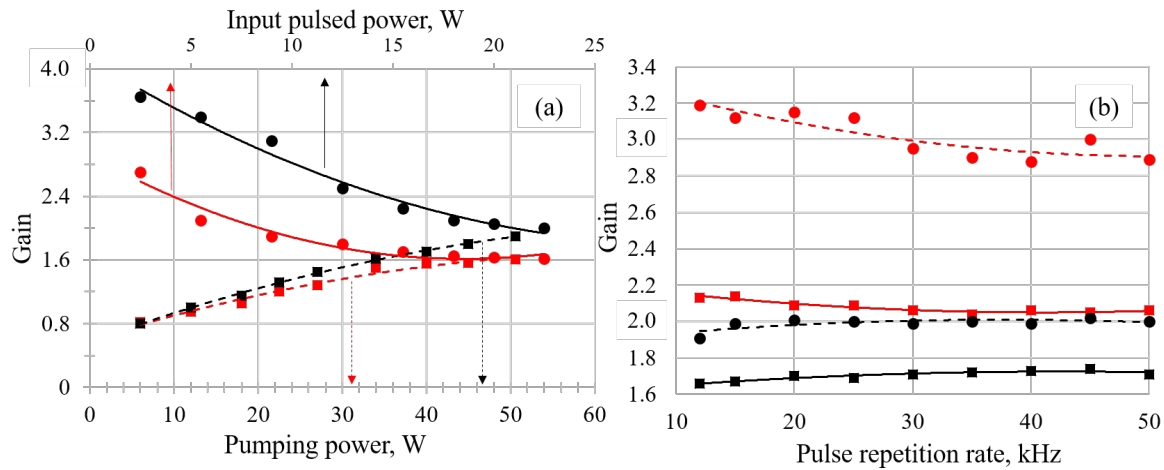


Figure 6. The Ho³⁺:YAG gain vs the CW pumping power (dashed lines) at the 21.6 W output power or the input power of the amplified beam (the solid lines) at the 50.6 W pumping power 20 kHz PRR of the 50 mm long rod (red) or the 65 mm long rod (a). The gain of the 50 mm (solid lines) or 65 mm (dashed lines) long amplifier rod vs the PRR at the input power of 9.6W (red) or 21.6 W (black) and the 50.6 W pumping power (b).

The beam quality of the amplified beam in the 65 mm rod remained high enough up to the highest power: the M2 parameter (measured using the knife-edge method [34]) was ≤ 1.8 at 30 W and ≤ 2.5 at 43 W output power (Figure 7).

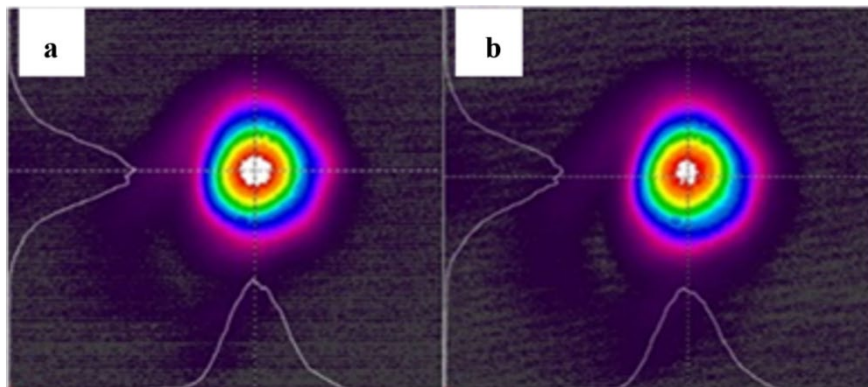


Figure 7. The beam profile at the output of the Ho³⁺:YAG amplifier with the 65 mm rod at the output power of 30 W (a) or 43W (b).

The amplified beam at 2091 nm was propagated through a half-wave-plate polarization rotator ($\lambda/2$ - plate) and a dichroic mirror M4 with a high p-polarization transmission at 2091 nm and a high s-polarization reflection at 2.3-2.7 μm before being directed into the second Cr²⁺:ZnSe crystal (Figure 3). The 2091-nm beam was the pumping beam of the Cr²⁺:ZnSe amplifier of the waves at 2.3-2.7 μm .

3.3. Cr²⁺:ZnSe Tunable Laser Oscillator

Multiple Cr²⁺:ZnSe crystals with varying lengths between 14 mm and 26 mm were tested as active elements of the laser oscillator. A part of the Ho³⁺:YAG laser beam was sent as the pump into the Cr²⁺:ZnSe laser oscillator. To avoid the LID on the Cr²⁺:ZnSe crystal surface the pump energy fluence was kept under 1.5 J/cm². For this reason, the pump beam diameter in the active element was 0.7–0.9 mm (at e⁻² peak intensity) depending on the PRR that was varied from 10 to 40 kHz.

The input dichroic mirror M2 of the Cr²⁺:ZnSe laser cavity (Figure 3) was chosen to enable high transmission of the p-polarized pump beam at 2091 nm and high reflectivity of the lasing

at 2.3–2.7 μm , which had the s-polarization. The rear mirror M1 had over 99% reflectivity both at the lasing wavelength of 2.3–2.7 μm and the pumping wavelength of 2091 nm. The M1 mirrors with the radius of curvature of 300 mm or 500 mm were tested in the experiments. The transmittance of the output coupler M3 was varied from 30% to 70% at the lasing wavelengths. The physical cavity length was also changed from 110 to 190 mm.

An acousto-optical tunable filter (AOTF) (“NII Polyus”, Moscow, Russia) inserted into the output leg of the laser cavity provided fine wavelength tuning on the deflecting acoustic wave. The driving voltage frequency was varied from 36.5 to 43 MHz. The acousto-optical element based on TeO_2 crystal had dimensions of $4 \times 4 \times 25 \text{ mm}^3$. The entrance and exit surfaces of the acousto-optical element were antireflection coated at 2.3–2.7 μm .

3.3.1. Numerical Simulation of the Thermal Lens and Optimization of the $\text{Cr}^{2+}:\text{ZnSe}$ Laser Cavity

The $\text{Cr}^{2+}:\text{ZnSe}$ -laser cavity was numerically modelled to ensure a good overlap of a fundamental mode with the pump beam. To calculate the radius of the fundamental mode in the “hot” laser cavity the thermal lens induced in the active element by the absorbed pump beam must be estimated.

A transient thermal lens was induced in the $\text{Cr}^{2+}:\text{ZnSe}$ active element under each pumping pulse. The dioptric power D_T of the transient thermal lens can be estimated by the following expression [35]:

$$D_T = -2 \times \left(\frac{\partial n}{\partial T} \right)_{\text{eff}} \times \frac{\partial}{\partial r^2} \int_0^L T(r, z, t) dz \Big|_{(r=0)}, \quad (2)$$

where $T(r, z, t)$ is the temperature distribution, r and z are the transverse and longitudinal coordinates, t is the time, $(\partial n / \partial T)_{\text{eff}}$ is the effective thermo-optic coefficient of the $\text{Cr}^{2+}:\text{ZnSe}$ crystal, including the volume coefficient $(\partial n / \partial T)$, bulging of end faces and the photoelastic effect, and given for an active element by the well-known expression:

$$(\partial n / \partial T)_{\text{eff}} \approx (\partial n / \partial T) + (n - 1)\beta_T(1 + \nu), \quad (3)$$

where n is the refractive index, β_T is the thermal expansion coefficient, ν is the Poisson's ratio. The transient temperature change during each pump pulse can be estimated by

$$T(r, z, t) = \frac{\sigma_{\text{abs}}(\lambda_p) (1 - \lambda_p / \lambda_s)}{\rho c_p} \int_0^t I_p(r, z, t) \times n_{\text{up}}(r, z, t) dt = \frac{N_{\text{da}} \sigma_{\text{abs}}(\lambda_p) (1 - \lambda_p / \lambda_s)}{\rho c_p} \int_0^{w_p(r, z, t)} dw_p \left[1 - \frac{\sigma_{\text{abs}}(\lambda_p)}{(\sigma_{\text{abs}}(\lambda_p) + \sigma_{\text{em}}(\lambda_p))} \left(1 - e^{-\frac{w_p(\sigma_{\text{abs}}(\lambda_p) + \sigma_{\text{em}}(\lambda_p))}{h \nu_p}} \right) \right], \quad (4)$$

where $\sigma_{\text{abs}}(\lambda_p)$ and $\sigma_{\text{em}}(\lambda_p)$ are the absorption and emission cross sections at the pump wavelength λ_p , ν_p is the pump frequency, λ_s is the laser wavelength, h is the Plank constant, ρ and c_p are the density and the thermal capacity of the $\text{Cr}^{2+}:\text{ZnSe}$ crystal, I_p and w_p are the pump intensity and fluence. The fluence of the short pumping pulse (with the pulse width τ_p much less than the excited-state life time) $w_p(r, z, t)$ can be given by Franz-Nodvig solution [36]:

$$w_p(r, z, t) = \frac{h \nu_p}{(\sigma_{\text{abs}}(\lambda_p) + \sigma_{\text{em}}(\lambda_p))} \ln \left(e^{-\alpha_{\text{abs}}(\lambda_p) z} \left(e^{-\frac{w_p(r, 0, t) (\sigma_{\text{abs}}(\lambda_p) + \sigma_{\text{em}}(\lambda_p))}{h \nu_p}} - 1 \right) + 1 \right). \quad (5)$$

The expressions (2)–(5) determined the focal length of the transient thermal lens at the end of a single pulse.

The focal length of the steady-state thermal lens f_T was estimated by the following expression [37,38]:

$$f_T = \frac{\pi K_T a_p^2}{P_{\text{in}} \left(1 - \frac{\lambda_p}{\lambda_s} \right) \left(\left(\frac{\partial n}{\partial T} \right) + (n - 1)\beta_T(1 + \nu) \right) (1 - \exp(-\sigma_{\text{abs}}(\lambda_p)L))}, \quad (6)$$

where P_{in} is the average incident pump power with a Gaussian beam radius of a_p . The other parameters of the crystal, pump beam, and laser cavity (including the TeO₂ AOTF) are shown in Table 1.

Table 1. Key parameters of the Cr²⁺:ZnSe crystal, laser cavity and the pump beam used for the numerical calculations, and the calculated focal length of a thermal lens in the laser element.

Parameter	Value
Density of the doping Cr ²⁺ ions, cm ⁻³	5×10 ¹⁸
Density of the ZnSe crystal ρ , g/cm ³	5.27 [39]
Specific heat capacity of the ZnSe crystal, C_p , J/(g K)	0,34 [39]
Thermal conductivity of the Cr ²⁺ :ZnSe crystal, K_T , W/(cm K)	0.18 [10,12,40]
Refractive index of the Cr ²⁺ :ZnSe crystal (at 2400 nm), n	2.43 [41]
Thermo-optic coefficient of the ZnSe crystal, $(\partial n/\partial T)$, K ⁻¹	61×10 ⁻⁵ [12,42]
Thermal expansion coefficient of the ZnSe crystal, β_T , K ⁻¹	7.3×10 ⁻⁶ [43]
Absorption cross section of the Cr ²⁺ :ZnSe crystal (at 2091 nm), σ_{abs} , cm ²	14 × 10 ⁻²⁰ [11]
Emission cross section of the Cr ²⁺ :ZnSe crystal (at 2450 nm), σ_{em} , cm ²	13 × 10 ⁻¹⁹ [11]
Poisson's ratio of the ZnSe crystal, ν	0.28 [44]
Length of the Cr ²⁺ :ZnSe element L , mm	16.5
Refractive index of TeO ₂ for the ordinary and extraordinary waves, n_{eTeO} and n_{oTeO} (at 2400 nm)	2.17 and 2.3 [45]
Length of the AOTF element, mm	25
Curvature of the M1 rear mirror, mm	300
Pump beam radius a_p (at e ⁻² intensity), μ m	450
Estimated focal length of the steady-state thermal lens in the Cr ²⁺ :ZnSe crystal (at the average input pump power, P_{in}), f_T , mm	28 (at 10 W) 14 (at 20 W)
Electronic nonlinear refractive index, n_2 , cm ² /W	1.2×10 ⁻¹⁴ [11,13]

The focal length of the thermal lenses at the end of each pulse (calculated by the expressions (2)-(5)) and the steady-state thermal lens (calculated by expression (6)) are presented in the Figure 8a.

The effective inversion fraction at the axis of the Cr²⁺:ZnSe active element at the end of each pumping pulse can be estimated by the following expression

$$\beta_{ef} = \frac{1}{L} \int_0^L \frac{n_{ex}(0,z,\tau_p)}{N_{da}} dz = \frac{1}{L} \int_0^L \left[1 - \frac{\sigma_p}{(\sigma_p + \sigma_{emp})} \left(1 - e^{-\frac{w_p(0,z,\tau_p) \times (\sigma_p + \sigma_{emp})}{h\nu_p}} \right) \right] dz \quad (7)$$

Expressions (5) and (7) provide a way to estimate the inversion fraction for the average pump power at the active element input:

$$P_{in} = W_p(0) \times R_{rate}, \quad (8)$$

where $W_p(0)$ is the input pump pulse energy, R_{rate} is the PRR. In this way, the gain cross-section can be plotted for each value of the input pump power (Figure 8b).

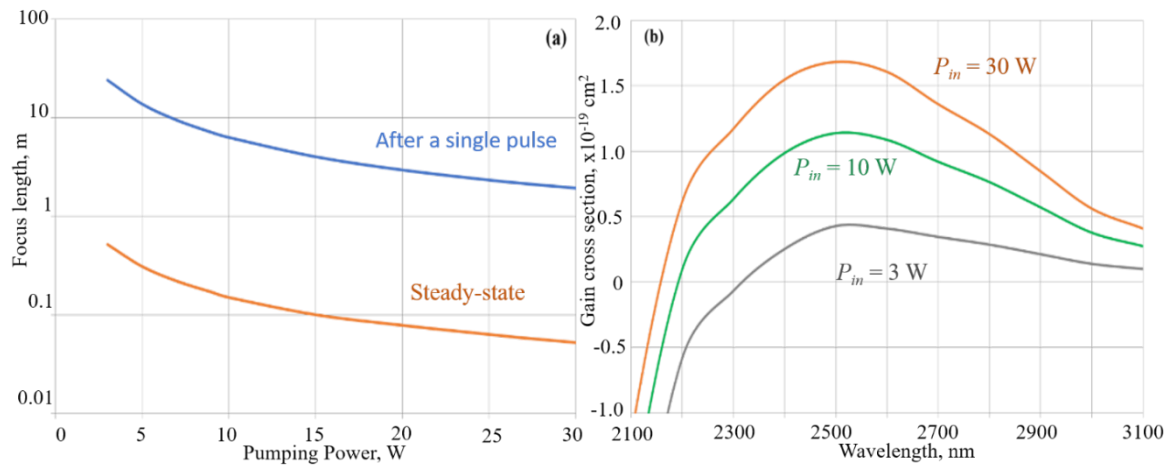


Figure 8. Numerically calculated focal length of the steady-state (brown) and transient (blue) thermal lenses in the $\text{Cr}^{2+}:\text{ZnSe}$ crystal vs the average input pump power at 2091 nm (a), and gain cross section the $\text{Cr}^{2+}:\text{ZnSe}$ crystal for the input pump power of 3 W (gray), 10 W (green), 30 W (red) (b). The calculation was made for the parameters presented in the Table 1.

The radius of the fundamental mode in the laser cavity with the steady-state thermal lens was calculated by the "Rezonator" software package [46]. The optimal overlap of the fundamental mode at 2400 nm and the pumping beam in the active element was achieved at the pump power of 28 W in the cavity with the physical length of 140 mm (Figure 6a). The optimal cavity length for the stable mode operation decreased with the increase in the pump power (Figure 6b). The rear mirror M1 with the 300 mm radius of curvature was found to be more suitable for the used pump power and the active elements due to the good overlap of the pumping beam and the laser mode.

3.3.2. $\text{Cr}^{2+}:\text{ZnSe}$ Laser Experimental Results

In the first series of experiments the $\text{Cr}^{2+}:\text{ZnSe}$ laser was examined without the intracavity AOTF. The repetitively-pulsed operation with the highest average output power of 3.8 W at 23 W pumping power and 30 kHz PRR was obtained in the 150-mm long cavity with the 300-mm radius of curvature on the rear mirror M1 and the 35% output coupler M3 (Figure 7). The highest output power was achieved with the 16.5-mm long $\text{Cr}^{2+}:\text{ZnSe}$ element.

The Ophir-Spiricon "PYROCAM IV" beam profiler was used to analyze the $\text{Cr}^{2+}:\text{ZnSe}$ laser output. The diameter of the output $\text{Cr}^{2+}:\text{ZnSe}$ laser beam was estimated to be 800-900 μm . The divergence angle and beam quality were analyzed in the focal zone of a lens with the 70 cm focal length using the knife-edge method [34]. The beam quality was $M^2 \leq 1.5$ at the output power of 3.8 W in the single-transverse-mode regime (Figure 7).

Waveforms of the $\text{Cr}^{2+}:\text{ZnSe}$ laser pulses were registered by an in-house-made photodetector based on a PD 36-02 (or 01)-PR(TO18) photodiode manufactured by IBSG Co., Ltd., St. Petersburg, Russia (spectral response range is 2.3–3.6 μm) in comparison with the pumping pulses of the $\text{Ho}^{3+}:\text{YAG}$ laser at 2091 nm, which were measured by an in-house-made photodetector based on HAMAMATSU InGaAs PIN G8422-03 photodiode (spectral response range is 0.9–2.1 μm). The $\text{Cr}^{2+}:\text{ZnSe}$ laser operated in the gain-switch mode: the laser pulse delayed with respect to the pumping pulse, the delay time decreased with an increase the pump pulse energy (Figure 8). The laser pulse width decreased from 50-70 ns near threshold to 20-25 ns far above threshold.

The output spectrum was measured by an IR Fourier spectrometer FT-801 (Simex, Russia). The wavelength of the power maximum and the linewidth were found to depend on both the pump power and the cavity mirrors. The wavelength of the spectral maximum tuned from 2400 nm to 2450 nm with an increase of the average pump power from 8 W to 22 W. The self-tune of the spectral maximum wavelength can be explained by tuning of the laser operation from the

mirror-reflection maximum at the low pump power to the gain maximum at the higher pump power. The linewidth at half maximum was measured to be ~ 70 nm at 18 W pump power (Figure 9a), and decreased as the pump power decreased.

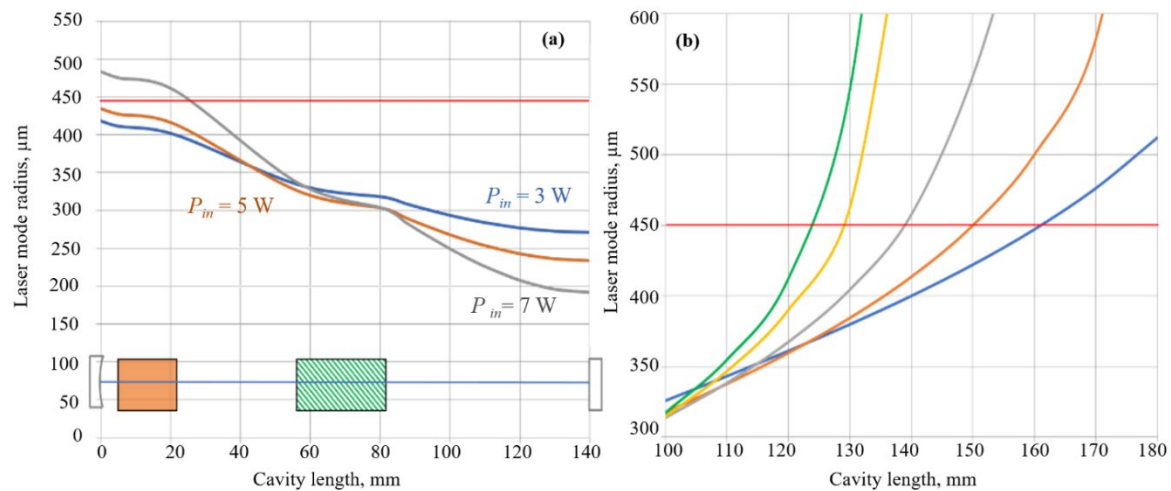


Figure 9. The calculated radius of the fundamental mode in the $\text{Cr}^{2+}:\text{ZnSe}$ -laser cavity with the active rod (brown) and the AOTF (green) for the input pumping power of 3 W (blue line), 5 W (brown line), 7 W (gray line) (a); the cavity mode radius at the right end of the active rod vs the total cavity length for the input pumping power of 10 W (green), 9 W (yellow), 7 W (gray), 5 W (orange), 3 W (blue). The pumping beam radius at the active rod is 450 μm (the red lines on in both figures).

The operating wavelength of the $\text{Cr}^{2+}:\text{ZnSe}$ laser with the intracavity AOTF was tuned by frequency tuning of the HF driver: from 2300 nm at ~ 42.6 MHz, to 2700 nm at ~ 36.6 MHz (Figure 12). The discreteness of the wavelength tuning was determined by the discreteness of the AOTF-driver frequency tuning, which was ~ 1 kHz (corresponding to the discreteness of the laser wavelength tuning of ~ 0.2 nm). The linewidth of the $\text{Cr}^{2+}:\text{ZnSe}$ laser with the intracavity AOTF was measured to be strongly ≤ 0.8 nm (the measurement accuracy was limited by the spectrometer used). The average output power of the electronically-tuned $\text{Cr}^{2+}:\text{ZnSe}$ laser at 23 W pumping power and 30 kHz PRR had a maximum of 3.1-3.3 W at 2410 nm, and decreased to 1.1-1.2 W at 2300 nm and to 0.6-0.8 W at 2700 nm (Figure 10). The beam quality of the tuned $\text{Cr}^{2+}:\text{ZnSe}$ laser remained high, $M^2 \leq 1.5$.

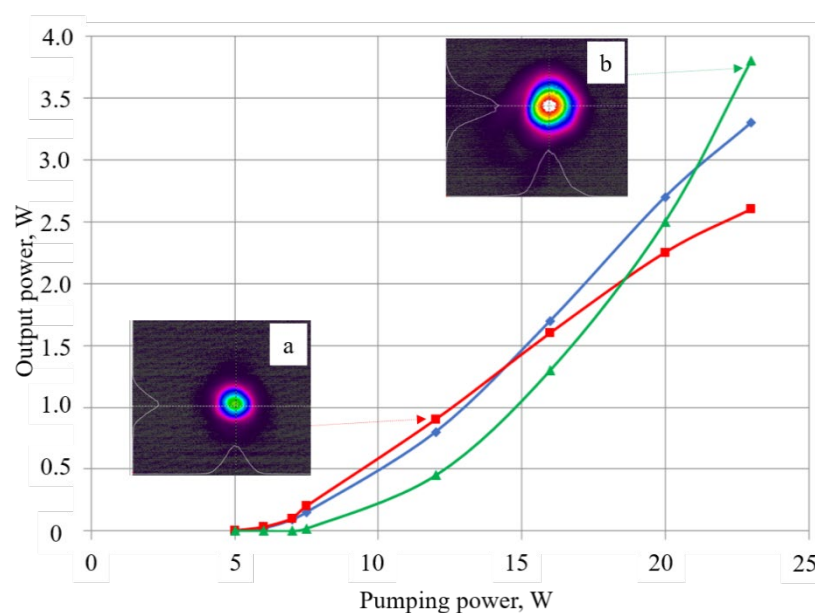


Figure 10. The average output power of the Cr²⁺:ZnSe laser vs the pumping power at PRR of 15 kHz (red), 25 kHz (blue) or 30 kHz (green) in the cavity with the output coupler M3 with the transmission of 50% (red and blue) or 65% (green). The inserts show the PYROCAM images of the Cr²⁺:ZnSe laser output beam at the average power of 0.9W (a) or 3.8W (b).

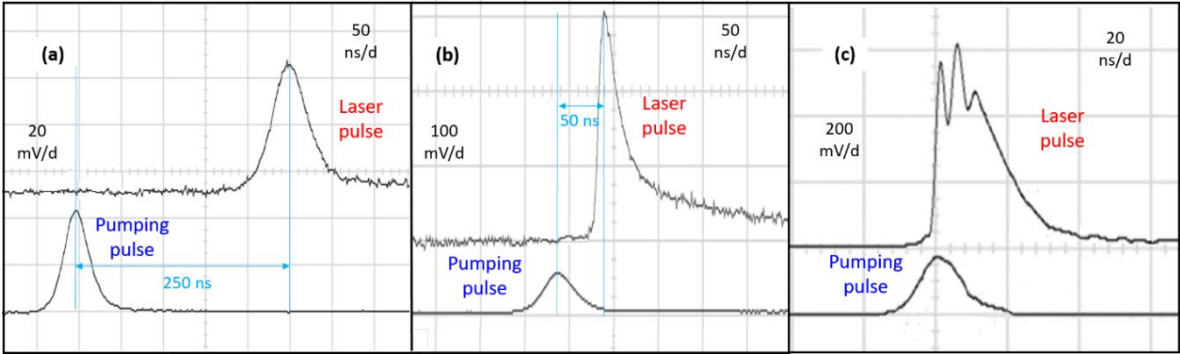


Figure 11. Waveforms of the Cr²⁺:ZnSe laser pulses (upper curves) and the input pumping pulses (lower curves) for different input pumping power, P_{in} : near the oscillation threshold at $P_{in} = 7$ W (a), above the threshold at $P_{in} = 15$ W (b), and far above the threshold at $P_{in} = 23$ W (c). PRR was 20 kHz.

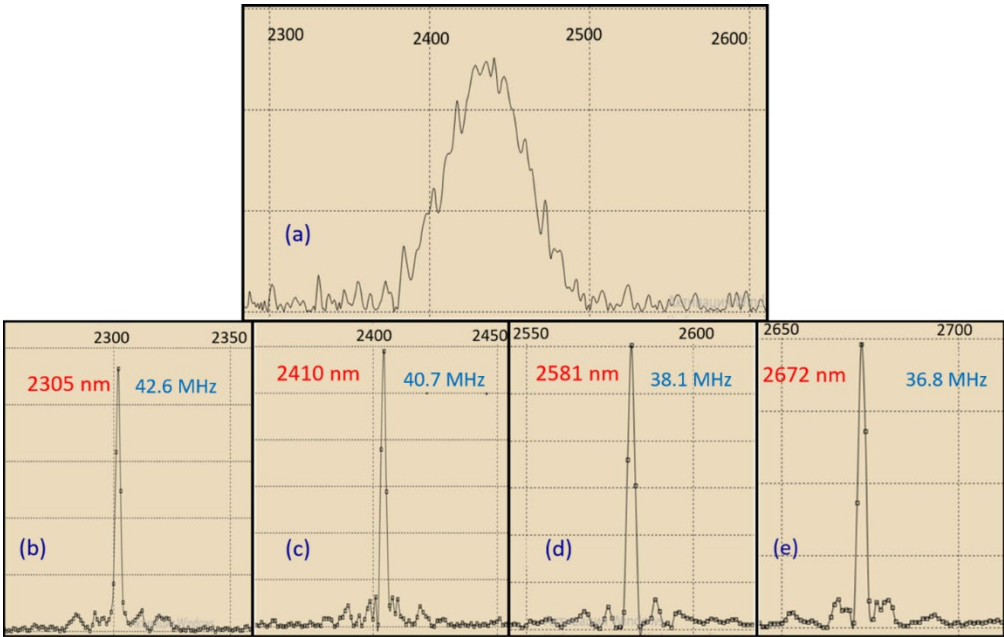


Figure 12. The output spectrum of the Cr²⁺:ZnSe laser in the cavity with 50% output coupler without AOTF at 18 W pump power (a), and with AOTF at tuning frequencies of 42.6 MHz (b), 40.7 MHz (c), 38.1 MHz (d) and 36.8 MHz (e).

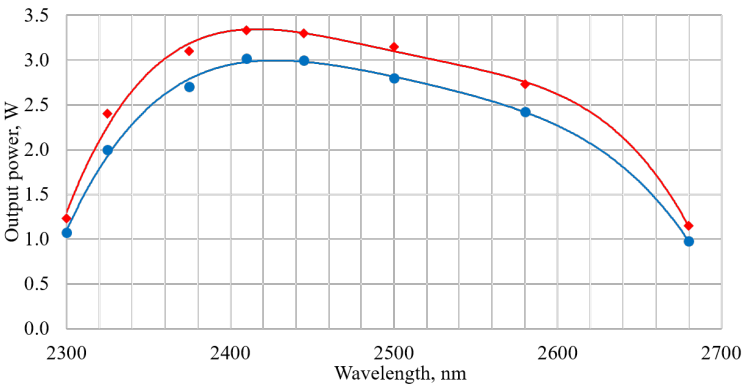


Figure 13. Output power of the wavelength-tuned $\text{Cr}^{2+}:\text{ZnSe}$ laser with the active element length of 16.5 mm (brown) or 20 mm (blue) at 23 W pump power and 30 kHz PRR vs the operating wavelength.

The $\text{Cr}^{2+}:\text{ZnSe}$ laser wave had linear polarization due to the polarization selectivity of the cavity mirror M2 and the AOTF. The polarization contrast (measured by a single-crystal silica wedge) was $\sim 70:1$ at the highest output power. The output beam of the $\text{Cr}^{2+}:\text{ZnSe}$ laser was directed to the $\text{Cr}^{2+}:\text{ZnSe}$ power amplifier by the dichroic mirror M4 (Figure 3).

3.4. $\text{Cr}^{2+}:\text{ZnSe}$ Power Amplifier

The $\text{Cr}^{2+}:\text{ZnSe}$ crystals with length varied from 16 to 32 mm were tested as active elements of the $\text{Cr}^{2+}:\text{ZnSe}$ amplifiers. The highest output power at 2.3–2.7 μm was achieved with the longer active element of 26–32 mm length.

The diameter of the pump beam at 2091 nm in the $\text{Cr}^{2+}:\text{ZnSe}$ amplifier was determined by the telescope T2 and the thermal lens inside the $\text{Ho}^{3+}:\text{YAG}$ amplifier. It was chosen to be approximately equal to the diameter of the amplified laser beam in the $\text{Cr}^{2+}:\text{ZnSe}$ crystal and had a value of 0.9–1.0 mm. To avoid LID in $\text{Cr}^{2+}:\text{ZnSe}$ the PRR of the master $\text{Ho}^{3+}:\text{YAG}$ oscillator was varied only within 20–40 kHz.

The power of the seed (amplified) beam at 2.3–2.7 μm and the pumping beam at 2091 nm was varied by adjusting the polarization beam splitter “P1 - $\lambda/2$ ” and the pumping power of the $\text{Ho}^{3+}:\text{YAG}$ oscillator and amplifier.

The 2.3–2.7 μm signal pulse lagged 10–30 ns behind the 2091 nm pump at the $\text{Cr}^{2+}:\text{ZnSe}$ amplifier. The delay depended on the $\text{Cr}^{2+}:\text{ZnSe}$ master oscillator pump power. However, this did not affect the amplifier gain, because the lifetime of the $\text{Cr}^{2+}:\text{ZnSe}$ upper-laser level is much longer.

The maximum average output power and pulse energy of the narrow-line wave at 2.41 μm reached 9.7 W and 0.32 mJ, respectively, at 30 kHz PRR; the total maximum amplifier gain was over a factor of 7 (Figure 14a). The maximum of average power and pulse energy was achieved using the 24-mm long $\text{Cr}^{2+}:\text{ZnSe}$ amplifier element. The beam quality of the amplified beam was $M^2 < 2$ at the maximum output power (Figure 14b). A slight deterioration in the beam quality of the amplifier output compared to the master-oscillator output can be explained by thermal effects in the long $\text{Cr}^{2+}:\text{ZnSe}$ amplifier element. The output pulse energy increased, but the average output power decreased with PRR decrease from 30 kHz to 20 kHz (Figure 14c).

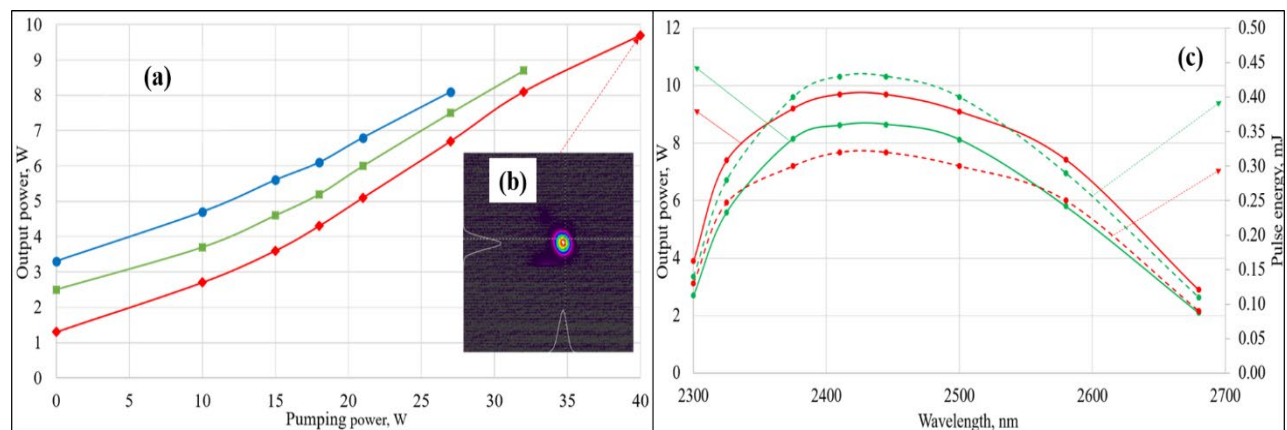


Figure 14. The average output power of the $\text{Cr}^{2+}:\text{ZnSe}$ amplifier at 2410 nm and 30 kHz PRR vs the pumping power at 2091 nm at different average input power 1.3 W (red), 2.5 W (green), 3.3 W (blue). The 2.4- μm output beam image at 9.7 W average power in the insert (b). The average power (solid lines) and pulse-energy (dashed lines) spectra of the narrow-line output for PRR of 20 kHz (green lines) or 30 kHz (red lines) (c). The $\text{Cr}^{2+}:\text{ZnSe}$ elements length was 28 mm.

4. Discussion

Power scaling of the Cr²⁺:ZnSe laser system could be achieved by adding power amplifiers [12,18]. However, several factors limit the pulse energy and the average power of the output beam in the Cr²⁺:ZnSe lasers: the LID, aberrative thermal lens or electronic self-focusing and gain saturation.

The rather high saturation fluence of the Cr²⁺:ZnSe amplifier (62 mJ/cm² at 2.45 μm) allows for further increases in the output power of the high-PRR nanosecond laser system with strong extraction efficiency. The thermal lens effect and electronic self-focusing can result in decreased beam diameter and deterioration of beam quality; these effects are more pronounced in longer Cr²⁺:ZnSe amplifiers. The thermal lens and electronic self-focusing are weaker at the lower beam fluence. For example, a *B*-integral estimation in the 30-mm amplifying crystal gives the value of approximately 0.34 for the input fluence $w_{in} = 1$ J/cm² at 2400 nm. Therefore, by increasing the diameter of the signal and pump beams in the amplifier and increasing the PRR, the influence of gain saturation, thermal lens could be reduced, and electronic self-focusing, and the Cr²⁺:ZnSe-crystal LID in the amplifier chain could be avoided. The use of a protective container during the hot isostatic treatment after Cr²⁺:ZnSe doping helps prevent contamination, resulting in an increase in the LID to 2.0-2.5 J/cm² at the high PRR.

5. Conclusions

The narrow-line broadly-tunable high-PRR laser system based on polycrystalline Cr²⁺:ZnSe elements pumped by repetitively-pulsed Ho³⁺:YAG lasers was developed. Wide-band wavelength electronic tuneability was achieved by using the TeO₂ AOTF. The average output power of up to 9.7 W at 30 kHz PRR in the single-stage Cr²⁺:ZnSe power amplifier was reached. The average output power and the pulse energy can be scaled by adding Cr²⁺:ZnSe amplifier stages.

The compact and powerful Cr²⁺:ZnSe laser system tunable within 2.3-2.7 μm was designed for a long-range LIDAR environmental monitoring in the transparency window of the Earth's atmosphere. Several pollutant gases (such as CH₄, NH₃, CO) have specific absorption bands in the wavelength range of 2.3-2.7 μm and can be detected using the developed laser source [47]. The high-repetition rate tunable Cr²⁺:ZnSe laser system can be used for other applications in medicine, material processing, and pumping of mid-infrared optical parametric oscillators and Fe²⁺-doped solid-state lasers.

6. Patents

This work resulted in a patent EA 041501B1 20221031 from the Eurasian patent office: "IR Laser source based on Cr²⁺-doped chalcogenide crystals with acousto-optical wavelength tuning", by O.L. Antipov, I.D. Eranov, Yu.A. Getmanovskiy.

Author Contributions: Conceptualization, O.A., and S.B.; methodology, O.A.; software, A.D., Yu.G., and V.Sh.; validation, I.E.; formal analysis, A.D., Yu.G.; investigation, O.A., I.E., N.Yu.; resources, S.B. and O.A.; data curation, Yu.G. and N.Yu.; writing—original draft preparation, O.A., S.B.; Yu.G.; writing—review and editing, O.A., S.B.; visualization, Yu.G.; supervision, O.A., S.B.; project administration, O.A.; funding acquisition, O.A. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, or in the decision to publish the results.

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