High power compact continuous-wave Fe:ZnSe laser at 4- μ m with >50% overall conversion efficiency

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We report on a compact, high efficiency mid-infrared continuous-wave (CW) Fe:ZnSe laser pumped by a 2.9 µm fiber laser under liquid nitrogen cooling. A maximum output power of 5.5 W and a slope efficiency of up to 66.3% with respect to the launched pump power were obtained. The overall optical-to-optical (OTO) conversion efficiency, calculated from the output of 2.9 µm fiber laser to the 4-µm laser, was as high as ~54.5%. The OTO efficiency and the slope efficiency are, to the best of our knowledge, the highest in ever reported Fe:ZnSe lasers. A rate-equation-based numerical model of CW operation was established, and the simulation agreed well with the experiment, identifying the routes used in the experiment for such high efficiency.

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Keywords: Fe:ZnSe, mid-infrared laser, high efficiency, solid-state laser.

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1.Introduction 1

There are increasing demands for high power solid-state42 2 mid-infrared lasers at 3~5 µm, for their applications in 43 3 laser radar, spectroscopy, remote sensing, infrared44 4 countermeasures, laser communication, etc^[1:3]. Currently, 45 5 the principal category of solid-state lasers at this band is 46 6 based on the nonlinear effect, including optical parametric 47 7 oscillators^[4-5](OPOs, typically using PPLN and ZGP48 8 crystals as nonlinear media), difference frequency49 9 generation (DFG)^[6], and frequency doubling^[7]. Yet there 50 10 are only a few direct solid-state mid-infrared 3~5 µm laser51 11 sources, merely including quantum cascade lasers^[8-9], 52 12 fiber lasers^[10] and transition metal (TM) ions doped 53 13 crystalline lasers (typically Fe:ZnSe or Fe:ZnS lasers)^[1,11].54 14 Compared with these lasers, FeiZnSe or FeiZnS lasers, 55 15 emitting at the mid-infrared range of 4~5 µm, enjoy the56 16 impressive advantages of high efficiency, 17 wide57 wavelength-tuning range, and compactness of optical58 18 cavity, etc^[12]. As such, lots of efforts have been made in the 59 19 development of Fe[:]ZnSe/Fe[:]ZnS lasers in the past decade.60 20 Since FeiZnSe crystal was demonstrated to be an effective61 21 gain medium for 4-um lasers for the first time in 1999^[1],62 22 lots of FeiZnSe lasers in different regimes, including63 23 CW^[2], gain-switching^[3], Q-switching^[13-14], mode-locking^[15]64 24 were demonstrated by employing various pumping65 25 sources at $\sim 3 \ \mu m$. For some practical applications, high₆₆ 26 power and highly stable CW 4-µm lasers are urgently 27 required. The upper laser level lifetime of Fe²⁺ in ZnSe 28 crystal is ~60 µs at 77 K while as short as ~370 ns at room 29 temperature^[16], as a consequence, it is necessary to cool 30 Fe:ZnSe crystal down to ~77 K by liquid nitrogen in a 31 cryostat for efficient CW operation. In 2008, Voronov 32 reported the first operation of CW Fe:ZnSe laser pumped 33 by a 2.97 µm Cr:CdSe laser^[17], of which the maximum 34 output power and an OTO efficiency (i.e., the laser output 35 power to the power of pump source output end) were 160 36 mW and 27%, respectively. Subsequently, Evans et al 37 demonstrated CW FeiZnSe lasers with output powers of 38 840 mW and 420 mW by using two Er:YAG lasers at 2.9467 39 $\mu m^{[2]}$ and an Er: Y₂O₃ laser at 2.74 $\mu m^{[18]}$, and the⁶⁸ ^b OTO efficiency with respect to power from the pump output end 40

corresponding OTO efficiencies were ~28% and 13.3%, respectively. Martyshkin et al developed a Cr:ZnSe laser at 2.94 µm with an output power of 23 W for power scaling. With the powerful pump source, they achieved a CW FeiZnSe laser with a maximum power of 9.2 W and an OTO efficiency of ~40%^[19]. Recently, Li et al presented a CW Fe[:]ZnSe laser pumped by an Er[:]YAP laser^[20], whose output power and OTO efficiency were ~1 W and 27.8%, respectively. Compared to solid-state pump lasers, fiber lasers possess the advantages of high beam quality and high stability, which are supposed to be ideal sources for pumping. Thanks to the dramatical development of fluoride fiber lasers at $\sim 3 \mu m$ in the past decades^[21-23], of which the recorded CW output power was as high as 41.6 $W^{[23]}$, therefore, it is natural to employ a fiber laser at ~3 µm to pump FeiZnSe crystals. In 2018, Pushkin et al demonstrated a compact CW FeiZnSe laser with a maximum power of 2.1 W and an OTO efficiency of 32% pumped by an Er:ZBLAN fiber laser at 2.8 µm^[24]. Table 1 presents comparison of the state-of-the-art CW FeiZnSe lasers. The lack of high power CW pump source seems to be the main obstacle for 4-µm laser power scaling. However, promotion of OTO efficiency is more important for obtaining higher output power when pump power is limited.

Tab. 1	State-of-the-art	CW Fe:ZnSe lasers
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Pump source	Fe:ZnSe	η_s^a	$\eta_o{}^b$	Refere
	laser			nce
Cr:CdSe laser(0.6 W at	160 mW	<56%	26.7%	[17]
2.97µm)				
Two Er:YAG lasers (total 3	840 mW	~40%	28%	[2]
W at 2.94 µm)				
Er:Y ₂ O ₃ (3.15 W at 2.74 μm)	420 mW	17.3%	13.3%	[18]
Cr:ZnSe (23 W at 2.94 µm)	9.2 W	41.2%	40.0%	[19]
Er:ZBLAN laser (6.5 W at	2.1 W	<59%	32.3%	[24]
2.8 μm)				
Er:YAP laser(3.6 W at 2.92	~1 W	<48.2%	27.8%	[20]
μm)				
Fluoride fiber laser (10.1 W	5.5W	66.3%	54.5%	This
at 2.91 µm)				work

^a Slope efficiency with respect to launched pump power

In this work, we report on a high power, high efficiency 48 1 and low-complexity FeiZnSe laser by adopting series of49 2 innovation, including customization of a pair of special₅₀ 3 coatings into the FeiZnSe crystal working facets for lower₅₁ 4 cavity loss, and a longer wavelength pump source for 52 5 higher pump absorption and higher quantum efficiency.⁵² The maximum output power of 5.5 W was obtained under 6 7 liquid nitrogen cooling, which is the highest in fiber-laser-54 8 pumped Fe[:]ZnSe lasers. The overall OTO efficiency with⁵⁵ 9 respect to the pump source was as high as $\sim 54.3\%$. The⁵⁶ 10 central wavelength was tuned from 3.96 to 4.15 µm by⁵⁷ 11 improving pump power. The laser was considerably⁵⁸ 12 compact and nearly alignment-free, which led to fairly 59 13 high long-term stability. 14 60 15 2.Experiments 61

The output of our CW Fe:ZnSe laser was compared under₆₂
two different pump schemes, i.e., forward pumping (FP)₆₃
and backward pumping (BP). The experimental setup of₆₄





Fig. 1. Schematic layout of the 2.9 µm fiber-laser-pumped CW⁷⁷
Fe:ZnSe laser of BP scheme. Inset: partial energy levels of Fe²⁺⁷⁸
ions involving pump absorption and laser emission.

Compared with the pump source of a 2.8 μm fiber laser 80 24 in the previous demonstration^[24], an all-fiber laser at 2.9^{-81} 25 μm was developed to pump the FeiZnSe crystal in our 82 26 experiment mainly based on the following considerations.⁸³ 27 One is that the longer wavelength of pump results in a⁸⁴ 28 promotion of the quantum efficiency compared to⁸⁵ 29 pumping at the shorter wavelengths, and the other is that $^{\rm 86}$ 30 the absorption cross-section at 2.9 μm of Fe:ZnSe at \log^{87} 31 temperature (LT) of ~77 K is larger than 2.8 $\mu m^{[18]}.$ In 88 32 2015, a maximum output power of 30.5 W was obtained $^{\rm 89}$ 33 from an Er:ZBLAN all-fiber laser at 2.94 $\mu m^{[22]}$, which⁹⁰ 34 was an ideal source for pumping Fe ZnSe. As the⁹¹ 35 techniques of writing fiber Bragg grating (FBG) in⁹² 36 ZBLAN fiber and splicing between a ZBLAN fiber and a^{93} 37 silica fiber continue to mature, it is common to develop an⁹⁴ 38 all-fiber laser at 3-µm waveband. A single-mode all-fiber⁹⁵ 39 CW Er:ZBLAN fiber laser with a maximum output power⁹⁶ 40 of ~10.1 W at a central wavelength of 2907.3 nm was in 97 41 house built to be used as the pump source. The pump 98 42 beam was collimated with a collimator (f=12.7mm) and 99 43 then focused into the Fe ZnSe crystal through an^{100} 44 uncoated CaF₂ lens (L1, f=50 mm). The pump beam waist¹⁰¹ 45 diameter was measured to be around 400 μ m. The¹⁰² 46 Fe ZnSe crystal, grown from the vapour phase by using a^{103} 47

concurrent-doping technology^[17], was a ~9 mm in length with a labeled Fe²⁺ ions concentration of 5.0×10^{18} cm⁻³. The one-pass absorption coefficient was measured to be 5.14 cm⁻¹ at a LT of ~77 K with a similar crystal with broadband AR coated for 2.7~4.8 µm. Consequently, the Fe²⁺ ions concentration was determined experimentally to be 5.5×10^{18} cm⁻³, in which the absorption cross section at $\lambda = 2907$ nm ($\sigma_{ex}=0.94 \times 10^{-18}$ cm²) was taken from Ref[25].

For more compact cavity design, both working facets (7 mm×7 mm) of the crystal were polished carefully to reduce the residual wedge as far as possible. Two series of special coatings were deposited onto the polished facets. The coating of S1 (see Fig. 1) had a high transmittance at the pump wavelength (T \geq 95% at 2.6~3 µm) and a high reflectivity at the output wavelength (R>99% at 3.7~4.8 µm), while the coating of S2 had a high transmittance at the pump wavelength (T \geq 95% at 2.6~3 µm) and a partial transmittance at the output wavelength (R~65% at $3.7 \sim 4.8$ µm), which served as the output coupler. The crystal was wrapped by a piece of indium foil and clamped to a U-shaped copper heat sink, and the temperature of the heat sink was controlled by a liquid-nitrogen-cooled cryostat from ~77 to ~300 K to study the influence of crystal temperature to the laser conversion efficiency. Inset of Fig. 1 is the partial energy levels of Fe^{2+} ions. The lifetime of upper laser level ${}^{5}T_{2}|2>$ reduces significantly when temperature of FeiZnSe crystal higher than 120 K due to the increase of the rate of non-radiative relaxation^[26, 27]. Theoretically, longer lifetime of upper laser level makes CW emission much easier. Therefore, almost all the CW FeiZnSe lasers operated by cooling Fe[:]ZnSe crystals to low temperature, e.g., 77 K. The vacuum windows (W1 and W2) of the cryostat were 3 mm CaF_2 plates with broadband anti-reflection (AR, T~99%) coated for 2.7~4.8 µm. A dichroic mirror (DM, HR>99 % at 2.6~3 μ m, HT>95% at 3.7~4.8 μ m) was placed with an incidence angle of 45° to separate the pump beam and output laser beam. Forward pumping was carried out by just turning around the cryostat in Fig. 1.

3. Results and discussion

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Firstly, output characteristics of the Fe[:]ZnSe laser were compared under different pumping schemes at a LT of ~77 K. The laser cavity was self-aligned and had a relatively low loss by customizing special coatings into the facets of the FeiZnSe crystal. It was easy to obtain the 4µm laser output without any tedious alignment of the laser cavity in both schemes when the launched pump power was higher than the threshold of around 0.4 W. The output powers of FP and BP as a function of launched pump power were shown in Fig. 2. The maximum powers of FP and BP were 5.3 and 5.5 W at the full pump power of 10.1 W. respectively. The slope efficiencies were 61.9% in FP and as high as 66.3% in BP, corresponding to overall OTO efficiencies of 52.4% and 54.5%, respectively. Both the slope efficiency and the overall OTO efficiency are the highest in ever reported FeiZnSe lasers. The slope

efficiency in BP was close to the limited laser efficiency,44 1 i.e., quantum efficiency (i.e., $\lambda_{\text{pump}}/\lambda_{\text{laser}}$), which is 69.9 % in 45 2 3 our case. The results suggested that the performance of 4 BP was slightly better than that of FP, which was similar 5 to fiber lasers with different pump schemes^[28]. The laser cavity length, i.e., the length of the crystal, was ~9 mm, 6 which was nearly one order of magnitude shorter than 7 previously reported Fe[:]ZnSe laser^[3]. Such short cavity 8 9 brought several advantages, including better cavity 10 stability and lower cavity loss. In laser cavity design, an important parameter denoted Fresnel number can be 11 expressed by N=d²/L λ , where d is the diameter of 12 excitation region in crystal and L is the cavity length. The 13 14 shorter the cavity, the higher the Fresnel number, which in turn led to lower diffraction loss of cavity. The 15 geometric loss was very small as well due to the high $_{47}^{46}$ 16 17 parallelism of cavity mirror. Thus, much lower cavity loss, 48 18 through integrating the cavity mirrors and the crystal⁴⁹ 19 instead of cavity mirrors, was mainly responsible for such 50 high efficiency. Further in this work, if not specially stated⁵¹ 20 21 our considerations are confined to the laser in BP regime.⁵² 22 No thermal roll-off at the maximum pump power was⁵³ observed, which indicated that further power scaling⁵⁴ 23 would be available by just increasing pump power. The⁵⁵ 24 25 output beam profile of BP at the maximum output power⁵⁶ 26 captured with a camera is shown in the inset of Fig. 3,57 27 from which we could found that the far-field beam spot58 28 had a symmetrical good Gaussian distribution. We59 29 measured the divergence of output beam in the forward60 pumping scheme with a CaF_2 lens (=100 mm). The spot61 30 diameter at the focal plane was around 1.5 mm, therefore, 62 31 the divergence was calculated to be 15 mrad (full angle). 32 63 64



Fig. 2. Output powers of FP and BP as a function of launched
pump power. Inset: the beam profiles of BP at the maximum
output power.

The output spectra at various output powers were captured by a mid-infrared optical spectrum analyzer with a resolution as high as 0.2 nm, as typically shown in Fig. 3. The gain spectra of Fe:ZnSe at different temperatures are very broad, as shown in Fig. 4. The evolution trend of spectra was that the central wavelengths were red-shifted to longer⁷⁶ positions with bandwidths broadened as increasing the pump power.



Fig. 3. Output spectra of Fe:ZnSe laser (BP) at low and high output powers.

The peak wavelength was 3.98 µm at the output power of ~ 1 W, and then shifted to 4.15 µm at ~ 5 W. The red-shift of central wavelength at different pump power is very common in free-running solid-state which is interpreted as increase lasers. of temperature of pumping region by local heating of gain crystal. A model was established to estimate the temperature distribution in the pumping area, and the highest temperatures generated in the center of pump beam spot were ~82.5 K at the output power of ${\sim}1~$ W and ${\sim}96.1~$ K at ${\sim}5~$ W, respectively. The increase of temperature was supposed to be responsible for such wavelength shift. There was a dip in the spectrum at the output power of 5W, the cause of which was attributed to the absorption by atmospheric CO_2 in the path from the output chamber window to the optical spectrum analyzer when measuring the spectrum. Compared to the spectra of a spiky structure in previous reports, the spectra in our case had a relatively smooth curve. It is interesting that the curves had only one peak with a FHWM bandwidth (3 dB) of ~20 nm at a low output power, and the bandwidth was slightly narrower (~12.3 nm) at a high power. The signal-tonoise ratio (SNR) was measured to be roughly 25 dB for low powers, and it was nearly 40 dB at the output power of 5 W.

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Fig. 4 Temperature-dependent gain spectra of Fe[:]ZnSe^[29]

The laser output was fairly stable by integrating the41 1 2 cavity coatings into the crystal to avoid any possible42 3 factors causing instability. The long-term stability of the43 4 laser at an output power of over 5 W over a 1 h period was 5 measured with a power meter, as shown in Fig. 5. The be⁴⁴ 6 peak-to-peak fluctuation was calculated to 7 approximately 0.7% at nearly the maximum output power.



Fig. 5. The measured long-term stability at an output power of
~5 W in BP scheme over a 1 h period with a power meter with a
response time of 0.6 s.

The conversion efficiency of our experiment was 12 even higher than the previous simulation of CW 13 operation^[30], in which the slope efficiency at a LT of $_{56}$ 14 77 K was only 42%. To gain insight into the laser $_{57}$ 15 dynamics and get an explanation for such high₅₈ 16 efficiency, we developed a numerical model of steady-59 17 state CW operation with rate-equation theory. $Only_{60}$ 18 the ground state $({}^{5}E|g>)$ and upper laser level₆₁ 19 $({}^{5}T_{2}|2>)$ of Fe²⁺ (see the inset of Fig. 1) need to be₆₂ 20 considered in the calculation. The ground $state_{63}$ 21 population and the upper laser level population were 64 22 denoted as N_0 and N_2 , respectively. 23 65

The power evolution of pump $P_p^{\pm}(z)$ and laser signal $_{66}^{66}$ $P_s^{\pm}(z)$ (+ forwards, - backwards) along the cavity axis $_{67}^{67}$ (with longitudinal coordinate z) could be expressed as $_{68}^{68}$ follows: 69

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$$\pm \frac{dP_{p}^{\pm}(z)}{dz} = -\sigma_{gsa}N_{0}(z)P_{p}^{\pm}(z) - \sigma_{esa}N_{2}(z)P_{p}^{\pm}(z) - \alpha_{p}P_{p}^{\pm}(z) \qquad (1$$

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$$\pm \frac{dP_s^{\pm}(z)}{dz} = P_s^{\pm}(z)(g(z) - l) + P_{ASE}$$
(2)72
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30 where the gain g(z) is:

$$g(z) = \Gamma \sigma_{\rm se} N_2(z)$$

32 in Eq. (1)~(3), σ_{gsa} , σ_{esa} and σ_{se} are the ground absorption, 33 excited-state absorption and emission cross sections of the Fe:ZnSe crystal, respectively; α_p is the intrinsic absorption 34 coefficient of the crystal; Γ is the overlap coefficient of 35 pump spot and laser spot; l is the total loss of the cavity 36 37 including the intrinsic loss of crystal, geometry loss and diffraction loss. P_{ASE} is the power of amplified spontaneous 38 emission (ASE), which is given by^[31]: 39

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$$P_{ASE} = \Gamma \frac{2hc^2}{\lambda_s^3} \sigma_{se} N_2(z) \Delta_{ASE}$$
(4)

where \triangle_{ASE} is the ASE bandwidth centered at signal wavelength; *h* is the Planck constant; *c* is the speed of light in vacuum; λ_s is the laser wavelength.

$$\frac{dN_{2}(z,t)}{dt} = \frac{\lambda_{p}(P_{p}^{+}(z,t) + P_{p}^{-}(z,t))}{\pi r_{p}^{2}hc} (\sigma_{gsa}N_{0}(z,t) - \sigma_{esa}N_{2}(z,t)) -\frac{\lambda_{s}(P_{s}^{+}(z,t) + P_{s}^{-}(z,t))}{\pi r_{s}^{2}hc} \sigma_{se}N_{2}(z,t) - \frac{N_{2}(z,t)}{\tau_{f}}$$
(5)

where λ_p is the pump wavelength, and τ_r is the fluorescence lifetime of upper laser level; r_p and r_s are the beam radii of pump and laser, respectively.

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(3)

The output power of the Fe:ZnSe laser could be expressed by:

$$P_{\rm out} = (1 - R_2) P_{\rm s}^{+}(L) \tag{6}$$

where R_2 is the reflectivity of output coupler, L is the position of output end of the cavity.

We carried out the model with parameters in line with our experiment, as shown in Tab. 2.

Parameter	Value	Parameter	Value
σ_{gsa}	0.94×10 ⁻¹⁸ cm ^{-2[25]}	$ au_{f}$	57 μs ^[27]
σ_{se}	1.1×10 ⁻¹⁸ cm ^{-2[25]}	N_t	5×10 ¹⁸ cm ⁻³
$\sigma_{esa}/\sigma_{gsa}$	0.12[32]	Δ_{ASE}	1.13 µm ^[25]
l	variable	α_p	0.01 cm ⁻¹
λ_p / λ_s	2.9 μm / 4.1 μm	n	2.4
R_{I}	0.99	R_2	0.65

The spectroscopic parameters of Fe²⁺ were taken from Ref[25]. According to Ref[32], the particles in upper laser level $({}^{5}T_{2}|2)$ jump to the higher level via excited-state absorption process by absorbing the pump photons not the laser photons, which is different from the process of the simulation in Ref[30]. The ratio of σ_{esa} to σ_{gsa} was deduced to be 0.12 in terms of the experiment results in ReF[32]. The simulation of output power with these input conditions under various cavity losses was shown in Fig. 6. The slope efficiency decreased as increasing the total loss. The maximum slope efficiency was 63.1%, which was very close to our experimental result. That is, the cavity loss in our case was slightly lower than 0.05 cm^{-1} due to such compact cavity with a length of ~1 cm. The geometry loss and diffraction loss would be increased when the cavity length was lengthened by at least one order of magnitude in previous demonstrations^[18-20]. Therefore, reducing the loss as much as possible is beneficial for high conversion efficiency.



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Fig. 6. The calculated output power as a function of launched pump
 power.

4 4.Conclusion

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5 In conclusion, a high power, high efficiency fiber-laser-63 pumped all-solid-state CW Fe:ZnSe laser with a current-64 6 recorded overall efficiency as high as 54.5% is⁶⁵ 7 demonstrated. Output power of over 5 W with a central⁶⁶ 8 wavelength of around 4.1 μm was obtained from a^{67} 9 Fe:ZnSe crystal under liquid nitrogen cooling. A_{co}^{68} 10 numerical model was established to analyze factors of $^{69}_{70}$ 11 influencing high efficiency CW operation. The simulation₇₁ 12 was in line with our experiment, indicating that, output₇₂ 13 14 power scaling to tens of watts or even higher could be73 15 possible by employ this configuration just by using higher₇₄ pump power. 16 75

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