

Efficient broadly tunable continuous-wave $\text{Cr}^{2+}:\text{ZnSe}$ laser

Irina T. Sorokina* and Evgeni Sorokin

Photonics Institute, Technische Universität Wien, Gusshausstrasse 27/387, A-1040 Wien, Austria

Alberto Di Lieto and Mauro Tonelli

Istituto Nazionale di Fisica della Materia and Dipartimento di Fisica dell'Università di Pisa, Via Buonarroti, 2-56127 Pisa, Italy

Ralph H. Page and Kathleen I. Schaffers

University of California, Lawrence Livermore National Laboratory L-482, Livermore, California 94550

Received July 19, 2000; revised manuscript received January 10, 2001

An efficient continuous-wave Cr^{2+} -doped ZnSe laser pumped by a Co:MgF_2 laser is experimentally demonstrated. In a single-pass pump scheme we observed up to 520 mW at ~ 2500 nm in 0.4-nm narrow-band operation, with 52% incident-power slope efficiency, and a tuning range between 2180 and 2800 nm. In the multipass pump scheme we also observed and analyzed the effect of dual Q -switching laser action at 1.75 and 2.5 μm in the Co:MgF_2 - Cr:ZnSe coupled-cavity oscillator. Finally, we report the measurement of the passive losses and of the ground-state absorption at the lasing wavelength. © 2001 Optical Society of America

OCIS codes: 140.3600, 140.3070.

1. INTRODUCTION

Cr^{2+} -doped broadband II-VI chalcogenide lasers, which were first suggested¹⁻³ and later studied,⁴⁻¹² represent nowadays the only room-temperature, diode-pumpable, and simple alternative to the existing midinfrared laser sources, based either on cryogenic crystalline lasers or on cascaded quantum-well materials, or use of nonlinear frequency down conversion approaches. Both pulsed⁴ as well as continuous-wave (cw) operation¹³ of Cr:ZnSe , pumped by an InGaAsP/InP laser diode, has been demonstrated so far. However, thorough investigation of the new laser system under more-optimal high-brilliance pumping (i.e., by a TEM_{00} laser-beam profile) is indispensable. Most novel concepts, physical phenomena, and limitations have been first studied under diffraction-limited laser pumping. Knowledge acquired in these studies can then be applied to the diode-pumped systems.

The first cw broadly tunable lasing of Cr:ZnSe pumped by a Tm:YALO laser was demonstrated by Wagner *et al.*^{10,11} An output power of 90 mW in the single-pass¹⁰ and 380 mW in the double-pass pumping scheme¹¹ were achieved, but with a noticeably broadened (50-nm) free-running linewidth. The authors pointed out that it was impossible to force the laser output into a single emission line even with an etalon.¹¹ Recently, 1 W of output power was obtained with a single-pass pumping by a Tm:YAG laser at 2013 nm.¹⁴ Broadband free-running oscillation has also been measured.¹⁴

In this study we investigate the cw Cr:ZnSe laser, pumped by a Co:MgF_2 laser in a single-pass pumping scheme, yielding 520-mW cw output power at 52%

incident-power slope efficiency (63% absorbed-power efficiency), tunable between 2180 and 2800 nm with a free-running linewidth of less than 0.5 nm. We discuss the unexpected observation of the dual Q -switching and lasing at both pump and laser wavelengths owing to cavity coupling in the case of multipass pumping or occasional reflections, as well as the line broadening associated with this phenomenon. We also report the measurement of the ground-state absorption (GSA) at the lasing wavelength. This knowledge is essential because GSA is intrinsic to the material and produces fundamental limitation on the laser threshold. By taking into account GSA at the lasing wavelength, we explain discrepancies in internal-loss measurements by Findlay-Clay and inverse slope-efficiency methods reported in the literature.^{3,10,11} Finally, we study how the presence of water vapor within the resonator affects the tunability range.

2. EXPERIMENTAL SETUP

The experimental setup is shown in Fig. 1. The $\text{Cr}^{2+}:\text{ZnSe}$ sample, in the form of a polished plate, was mounted at the Brewster angle on a room-temperature copper block in an astigmatically compensated X-fold four-mirror cavity, consisting of 75-mm and 100-mm radius-of-curvature dichroic mirrors, a flat high reflector (HR), and an output coupler (OC) with transmission of 1.7%, 2.7%, 6.8%, 7.8%, or 12%. These were the only curved mirrors available at the time of the experiment. The resonator-beam spot size was calculated to be approximately $70\ \mu\text{m} \times 170\ \mu\text{m}$ (astigmatism from the Brewster-angle orientation of the plate), whereas the

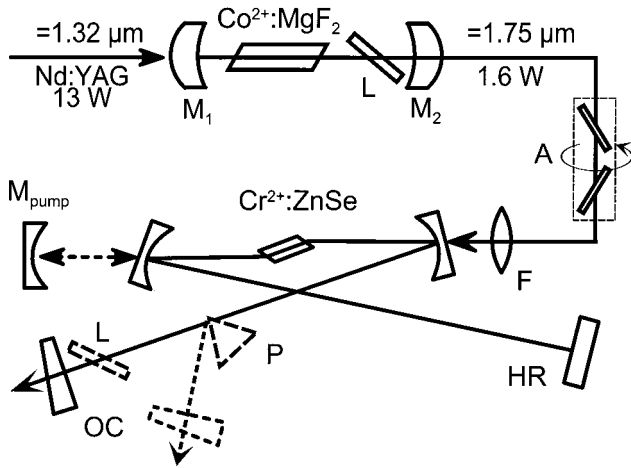


Fig. 1. Schematic of the experimental setup. The pump at 1.75 μm and the output radiation are polarized in the plane of the figure. L, birefringent filter; A, two-plate variable attenuator; F, uncoated 50-mm focusing lens.

pump beam was focused into an estimated $50\mu\text{m} \times 120\mu\text{m}$ spot inside the crystal by a 50-mm uncoated lens. This slightly nonoptimal spot-size relation was found to provide the highest output power, albeit a somewhat increased threshold. Three samples, diffusion doped with Cr, were prepared: a single crystal with thickness $d = 2.8\text{ mm}$, upper-state lifetime $\tau = 8.3\mu\text{s}$, and absorption coefficient $k = 3.2\text{--}8.0\text{ cm}^{-1}$ (owing to strong doping inhomogeneity), and two other samples with $d = 3\text{ mm}$ and 4.5 mm , $\tau = 6.6\mu\text{s}$ and $7.3\mu\text{s}$, and $k = 3.6\text{ cm}^{-1}$ and 4.5 cm^{-1} , respectively, the last being a polycrystalline material. The actual absorption during laser operation varied between 30% and 90%. The first sample performed significantly better than the latter two and is referred to below.

For Cr:ZnSe a crucial point is the choice of the pump source, since the absorption band of Cr^{2+} is centered at $1.75\mu\text{m}$, and only a few cw lasers are able to deliver sufficient power in this region. Our pump source was a cw liquid-nitrogen-cooled $\text{Co}^{2+}:\text{MgF}_2$ laser,¹⁵ tunable between 1.62 and $2.13\mu\text{m}$; it was optimized to obtain the highest possible (1.6-W) power in TEM_{00} mode at $1.75\mu\text{m}$. The broad tuning range allowed us to investigate thoroughly our Cr:ZnSe laser while pumping at different wavelengths. The effective power coupled into the Cr:ZnSe cavity was adjusted by the variable attenuator, consisting of two Brewster-angled ZnSe plates, rotating around the beam axis.

3. RESULTS AND DISCUSSION

A. Output Parameters and Crystal Losses

Figure 2 shows the laser output power versus incident pump power for three different output couplers: 1.8%, 6.8%, and 12%. In the optimized cavity, up to 520 mW of output power at 52% slope efficiency (63% absorbed-power slope efficiency) at $\sim 2500\text{ nm}$ was achieved without a noticeable thermal lensing effect. The output linewidth in the cw regime was measured to be less than 0.4 nm (the nominal spectral resolution of the Digikrom 480 monochromator with 50- μm slits and a 600-groove/mm

grating is 0.2 nm). It is important to notice that all the above measurements were carried out in the single-pass pumping scheme. The free-running linewidth increased to 10–20 nm in the nonstationary regime, e.g., when a chopper was used, or in a gain-switched mode (see below).

Using the standard Findlay–Clay analysis,¹⁶ we measured a value of $(10 \pm 3)\%$ (16%/cm) for round-trip passive losses [Fig. 3(a)], in agreement with the value 15%/cm.⁴ However, an alternative method of inverse slope efficiencies¹⁷ [Fig. 3(b)] yields only $(2 \pm 0.2)\%$ round-trip losses. The disagreement between the results of the measurements with different methods was repeatedly pointed out in other publications^{1,10,11} and deserves additional attention.

The consistent difference in results can be explained under the assumption of a nonzero GSA. With GSA included in the consideration, the inverse slope-efficiency method¹⁷ can still be applied to yield the correct result of 2% unsaturable losses, provided that the slope efficiency is extracted from the linear region of the output curve¹⁸

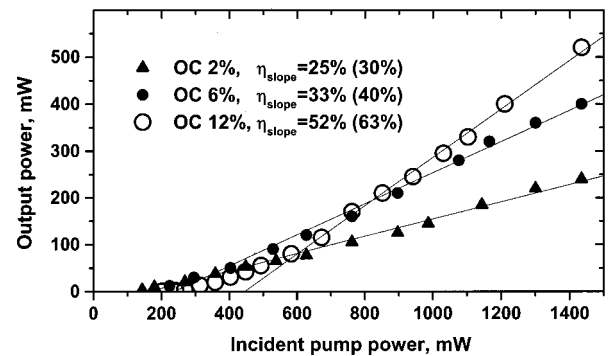


Fig. 2. $\text{Cr}^{2+}:\text{ZnSe}$ laser output power versus pump power with different output couplers (OC). In the parentheses the slope efficiency versus the absorbed power is given. Curvature in the threshold region is due to GSA bleaching.¹⁶

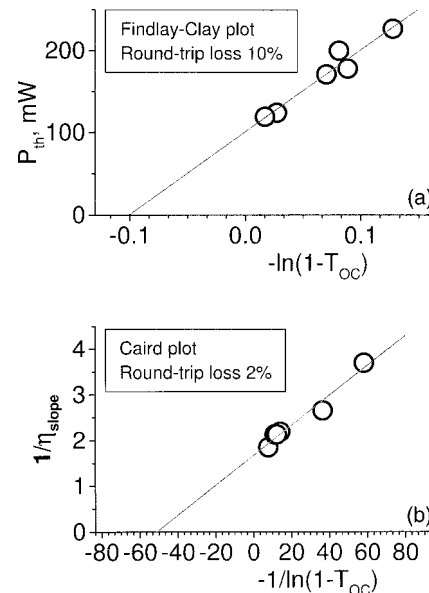


Fig. 3. $\text{Cr}^{2+}:\text{ZnSe}$ passive intracavity-loss determination by two methods: (a) Findlay–Clay plot and (b) the method of inverse slope efficiencies. In both cases the logarithmic round-trip loss value is given by the intersection of the linear fit with the abscissa. The discrepancy is due to the GSA.

far above the threshold. However, the original Findlay–Clay formula¹⁶ $P_{th} = P_{th0}[1 - \ln(1 - T_{OC})/2\delta]$ (T_{OC} is the transmission of the output coupler, 2δ is the round-trip loss, and P_{th0} is the threshold pump power with no output coupling) has to be extended to the quasi-three-level case. Using Eq. (7a),¹⁸ we can write the following expression for the threshold pump power of a quasi-three-level laser:

$$P_{th} = P_{th0} \left[1 + \frac{-\ln(1 - T_{OC})}{2\delta + 2\alpha(\lambda_0)l} \right], \quad (1)$$

where α_0 is the GSA coefficient at the oscillation wavelength λ_0 and l is the crystal length. The above expression allows use of the Findlay–Clay plot technique (Fig. 3a), but the round-trip loss value of 10% includes the GSA. The assumption of the nonzero ground-state population also explains the nonlinear initial region of the output curve (Fig. 2), a typical feature of the longitudinally pumped quasi-three-level lasers.¹⁸

In order to find additional evidence, we carried out absorption measurements of the two samples in a Fourier-transform infrared spectrophotometer in a wide wavelength range of up to 30 μm , using an undoped ZnSe plate as a reference. The absorption around 2.5 μm (Fig. 4) arises from the overlap between the absorption band owing to the $^5T_2 \rightarrow ^5E$ transition and the transition within the ground state 5T_2 owing to the static Jahn–Teller effect around 6.6 μm .^{19,20} It scales with Cr^{2+} concentration so that the figure of merit, defined as the ratio of attenuation coefficients at the absorption peak and at the lasing wavelength, remains approximately the same.

We can thus conclude that losses from GSA are intrinsic to Cr:ZnSe crystals and provide the upper limit for the figure of merit. The round-trip GSA loss from the spectrum in Fig. 4 is estimated to be $(13 \pm 1)\%$ for a 2.8-mm-thick sample and constitutes the major part of the losses, defined from the Findlay–Clay analysis. The actual passive round-trip loss, which is determined from the inverse slope-efficiency analysis (Caird plot¹⁷), is as small as 2%. This is an important point because passive losses can be minimized through improved growth techniques, whereas the effect of the GSA produces a fundamental limitation

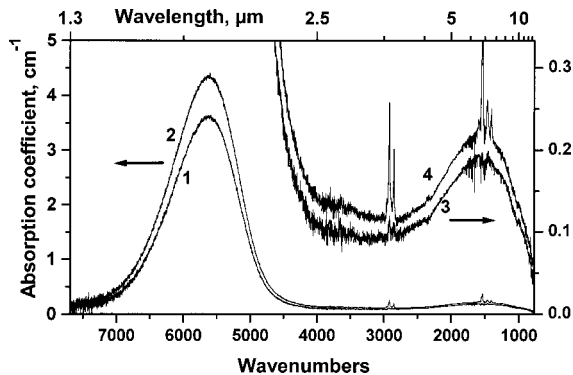


Fig. 4. Absorption spectra of the polycrystalline (curve 2) and single-crystal Cr:ZnSe samples (curve 1). Curves 3 and 4 are the $15\times$ magnifications of curves 1 and 2, respectively. The fine structure in the 3500–4000- cm^{-1} region is due to atmospheric H_2O . The sharp structure at 1600 cm^{-1} and 2900 cm^{-1} is likely due to bending and stretching vibrations of OH bonds in the crystals.

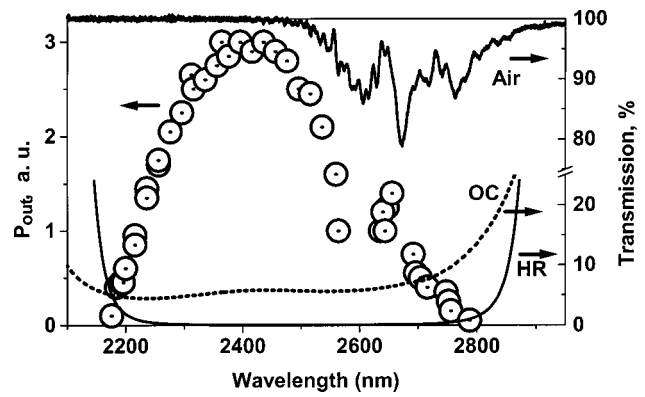


Fig. 5. Tuning range of the laser measured with a 0.8-mm Lyot filter (open circles). Also shown is transmission of the output coupler (OC), the high reflector (HR, five reflections), and 2.2 m of air at $\sim 70\%$ humidity (Air). Attenuation from intracavity air induces dips in the output power at certain wavelengths.

on the laser threshold. A high slope efficiency approaching the quantum-defect limit (70%) suggests negligible excited-state absorption, in agreement with spectroscopic analysis² and direct excited-state absorption measurements.¹⁴

B. Tunability

In the free-running mode the laser operated around 2490 nm. We carried out two different measurements of the tuning range by using a fused-silica prism and a 0.8-mm quartz birefringent filter: in both cases we measured a tuning curve between 2180 and 2800 nm (Fig. 5). The tunability was limited on both sides by the cavity optics. On the red side of the spectrum the emitted power is lowered owing to water-vapor absorption. In Fig. 5 we give the corresponding measured absorption spectrum of air, normalized to the length of the cavity. The dips in the tuning curve follow the corresponding water spectral peaks and could be eliminated by purging the resonator with dry nitrogen.

C. Cw and Q-Switched Operation

In order to increase the output power from the low-concentrated sample we used the power-scaling approach.^{10,11} Double-pass pumping was achieved by introducing the 150-mm curved HR mirror M_{pump} to collect back the transmitted pump radiation at 1.75 μm (Fig. 1). While allowing an increase of the output power in the low-doped sample from ~ 60 mW to ~ 170 mW, this pump scheme also created unavoidable feedback into the pump laser. The pump laser then operated in a Q-switched mode with pulses of 2.5–10- μs duration at a 3–20-kHz repetition rate, depending on the crystal absorption and the attenuator loss. Consequently, the output of the Cr:ZnSe laser was also pulsed, with a typical pulse width of 5–20 μs and with the linewidth broadened to ~ 40 nm. Such behavior can be understood, assuming that under given conditions, the Cr:ZnSe crystal acts as a coupled-cavity saturable absorber for a Co:MgF₂ laser. The low saturation intensity of Cr:ZnSe (14 kW/cm^2)³ and the strong focusing of the Co:MgF₂ radiation into the Cr:ZnSe crystal ensures easy saturation of the latter. The line-

width of ~ 40 nm is very similar to that reported,^{10,11} where the multipass regime was also used.

A more detailed treatment of the self- Q -switching regime can be performed by considering the stability conditions for cw operation in a resonator with coupled cavities.²¹ For the setup considered it can be shown that the pump laser with saturation intensity $I_{\text{sat}}^{\text{pump}}$ will switch to the pulsed regime, when

$$\frac{I_{\text{sat}}^{\text{pump}} S^{\text{pump}}}{I_{\text{sat}} S} T_{\text{OC}} T_A > 1 + \frac{-\ln(T_A) + \gamma}{\alpha_0 l} \left(\frac{P}{I_{\text{sat}} S} + 1 \right)^2, \quad (2)$$

where I_{sat} is the absorption saturation intensity of the Cr:ZnSe, $I_{\text{sat}}^{\text{pump}}$ is the gain saturation intensity of the Co:MgF₂, S and S^{pump} are the respective mode areas, P is the pump power incident upon the Cr:ZnSe crystal, and $\alpha_0 l$ is the small-signal absorption coefficient of the Cr:ZnSe. T_{OC} is the transmission of the output coupler of the Co:MgF₂ laser, and T_A is the transmission of the attenuator. Finally, γ is the unsaturable loss coefficient in the coupled cavity. With the waist diameters of $0.3 \text{ mm} \times 0.4 \text{ mm}$ in the Co:MgF₂ laser and $50 \mu\text{m} \times 120 \mu\text{m}$ in the Cr:ZnSe laser and the respective saturation intensities of 76 kW/cm^2 (Ref. 15) and 14 kW/cm^2 (Ref. 3), we find that the left-hand side of expression (2) is $\sim 5 T_A$ and the right-hand side is ≤ 2 . This explains why the pump laser switches to the pulsed regime whenever the two-pass geometry is used. Experimentally, we found that the Co:MgF₂ laser was Q switched with the attenuator transmission changing from $T_A = 1$ down to $T_A \sim 0.2$.

4. CONCLUSION

In conclusion, this study presents, for the first time to our knowledge, a room-temperature cw Cr:ZnSe laser, pumped by a cryogenically cooled Co:MgF₂ laser, capable of more than 0.5-W output power in the single-pass regime. Here we should note that the system as a whole is not a room-temperature device. However, the laser is distinguished by a narrow linewidth, a slope-efficiency value close to the theoretical limit, and continuous tunability in an interval $\sim 25\%$ of the central wavelength. We also present an explanation of the observed dual Q switching and lasing at both the pump and the laser wavelengths owing to the coupling with a pump laser cavity, developed in the case of the multipass pumping. We believe this phenomenon to be responsible for the broadening of the linewidth observed in the latter case. Finally, there is an intrinsic loss owing to GSA, which sets the limit to the threshold.

ACKNOWLEDGMENTS

The authors wish to express their sincere gratitude to W. F. Krupke, whose encouragement, personal support, and stimulating exchange of ideas at the early stage are the key factors for the success of this study. The research was supported by the Austrian National Science Foundation Projects P12756-TPH, P14704-TPH, and T-64, and Austrian National Bank Project 7913.

*E-mail address: sorokina@tuwien.ac.at.

REFERENCES

1. R. H. Page, L. D. DeLoach, K. I. Schaffers, F. D. Patel, R. J. Beach, S. A. Payne, W. F. Krupke, and A. Burger, "Recent developments in Cr²⁺-doped II-VI compound laser," in *Advanced Solid-State Lasers*, S. A. Payne and C. Pollock, eds., Vol. 1 of OSA Trends in Optics and Photonics Series (Optical Society of America, Washington, D.C., 1996), pp. 130–136.
2. L. D. DeLoach, R. H. Page, G. D. Wilke, S. A. Payne, and W. F. Krupke, "Transition metal-doped zinc chalcogenides: spectroscopy and laser demonstration of a new class of gain media," *IEEE J. Quantum Electron.* **32**, 885–895 (1996).
3. R. H. Page, K. I. Schaffers, L. D. DeLoach, G. D. Wilke, F. D. Patel, J. B. Tassano, S. A. Payne, W. F. Krupke, K.-T. Chen, and A. Burger, "Cr²⁺-doped zinc chalcogenides as efficient, widely tunable mid-infrared lasers," *IEEE J. Quantum Electron.* **33**, 609–619 (1997).
4. R. H. Page, J. A. Skidmore, K. I. Schaffers, R. J. Beach, S. A. Payne, and W. F. Krupke, "Demonstration of diode-pumped and grating tuned ZnSe:Cr²⁺ lasers," in *Advanced Solid State Lasers*, C. R. Pollock and W. R. Bosenberg, eds., Vol. 10 of OSA Trends in Optics and Photonics Series (Optical Society of America, Washington, D.C., 1997), pp. 208–210.
5. U. H  mmerich, X. Wu, V. R. Davis, S. B. Trivedi, K. Grasza, R. J. Chen, and S. Kutcher, "Demonstration of room-temperature laser action at $2.5 \mu\text{m}$ from Cr²⁺:Cd_{0.85}Mn_{0.15}Te," *Opt. Lett.* **22**, 1180–1182 (1997).
6. K. L. Schepler, S. K  ck, and L. Shiozawa, "Cr²⁺ emission spectroscopy in CdSe," *J. Lumin.* **72–74**, 116–117 (1997).
7. J. McKay, K. L. Schepler, and G. C. Catella, "Efficient grating tuned mid-infrared Cr²⁺:CdSe laser," *Opt. Lett.* **24**, 1575–1577 (1999).
8. J. T. Seo, U. H  mmerich, S. B. Trivedi, R. J. Chen, and S. Kutcher, "Slope efficiency and tunability of a Cr²⁺:Cd_{0.85}Mn_{0.15}Te mid-infrared laser," *Opt. Commun.* **153**, 267–270 (1998).
9. U. H  mmerich, M. Turner, A. Bluiett, J. T. Seo, H. Zong, S. B. Trivedi, S. W. Kutcher, C. C. Wang, R. J. Chen, P. R. Boyd, and W. Tardiff, "Mid-infrared luminescence properties of Cr²⁺ and Co²⁺ doped CdTe and cadmium manganese telluride," in *Digest of Conference on Lasers and Electro-Optics* (Optical Society of America, Washington, D.C., 1999), p. 421.
10. G. J. Wagner, T. J. Carrig, R. H. Page, K. I. Schaffers, J. Ndap, X. Ma, and A. Burger, "Continuous-wave broadly tunable Cr²⁺:ZnSe laser," *Opt. Lett.* **24**, 19–21 (1999).
11. G. J. Wagner, T. J. Carrig, R. H. Page, K. I. Schaffers, J. Ndap, X. Ma, and A. Burger, "High-efficiency, broadly tunable continuous-wave Cr²⁺:ZnSe laser," in *Advanced Solid-State Lasers*, M. M. Fejer, H. Injeyan, and U. Keller, eds., Vol. 26 of OSA Trends in Optics and Photonics Series (Optical Society of America, Washington, D.C., 1999), pp. 427–434.
12. T. J. Carrig, G. J. Wagner, A. Sennaroglu, J. Y. Jeong, and C. R. Pollock, "Mode-locked Cr²⁺:ZnSe laser," *Opt. Lett.* **25**, 168–170 (2000).
13. E. Sorokin, I. T. Sorokina, and R. H. Page, "Room-temperature CW diode-pumped Cr²⁺:ZnSe laser," in *Advanced Solid-State Lasers*, OSA Trends in Optics and Photonics Series (Optical Society of America, Washington, D.C., 2001), paper MB11.
14. A. V. Podlipensky, V. G. Shcherbitsky, N. V. Kuleshov, V. I. Levchenko, V. N. Yakimovich, A. Diening, M. Mond, E. Heumann, S. K  ck, and G. Huber, "1 W continuous-wave laser generation and excited state absorption measurements in Cr²⁺:ZnSe," in *Advanced Solid State Lasers*,

- H. Injeyan, U. Keller, and Ch. Marshall, eds., Vol. 34 of OSA Trends in Optics and Photonics Series (Optical Society of America, Washington, D.C., 2000), pp. 201–206.
15. P. F. Moulton, “An investigation of the Co:MgF₂ laser system,” *IEEE J. Quantum Electron.* **21**, 1582–1595 (1985).
 16. D. Findlay and R. A. Clay, “Measurement of internal losses in 4-level lasers,” *Phys. Lett.* **20**, 277–278 (1966).
 17. J. A. Caird, S. A. Payne, P. R. Staver, A. J. Ramponi, L. L. Chase, and W. F. Krupke, “Quantum electronic properties of the Na₃Ga₂Li₃F₁₂:Cr³⁺ laser,” *IEEE J. Quantum Electron.* **24**, 1077–1099 (1988).
 18. T. Y. Fan and R. L. Byer, “Modeling and CW operation of a quasi-three-level 946 nm Nd:YAG laser,” *IEEE J. Quantum Electron.* **24**, 895–912 (1988).
 19. B. Nygren, J. T. Vallin, and G. A. Slack, “Direct observation of the Jahn–Teller splitting in ZnSe:Cr²⁺,” *Solid State Commun.* **11**, 35–38 (1972).
 20. M. Kaminska, J. M. Baranowski, S. M. Uba, and J. T. Vallin, “Absorption and luminescence of Cr²⁺(d⁴) in II-VI compounds,” *J. Phys. C* **12**, 2197–2214 (1979).
 21. E. Sorokin and I. T. Sorokina, “Coupled-cavity passive Q-switching,” in *Conference on Lasers and Electro-Optics/Europe* (IEEE, New York, 2000), p. 359.