Tunable, Diode-Pumped Solid-State Laser for Scalpel with Adjustable Penetration Depth

R. H. Page

February 10, 1998

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96-LW-005 final report

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**Tunable, Diode-Pumped Solid-State Laser
for Scalpel with Adjustable Penetration Depth**

**Ralph H. Page
Lawrence Livermore National Laboratory
Livermore, California 94550**

I. Introduction

Three years ago, as part of a remote-sensing project, we discovered a new class of tunable mid-infrared (IR) transition-metal lasers: Cr²⁺-doped zinc chalcogenides. This is the first time that an efficient, widely-tunable, mid-IR laser operating at room temperature was produced. The broad emission linewidths of the Cr²⁺ dopant ions, and their strong, highly-luminescent optical transitions are reminiscent of the well-known Ti:sapphire system. Table I compares the spectroscopic properties of the Ti:sapphire and ZnSe:Cr systems. There is a fairly close analogy, probably due largely to the fact that both dopant ions are transition metals, and to the similarity of electronic energy level structures (evident in the E → T₂ optical transition.) A key difference—aside from 3×-longer transition wavelengths around 2400 nm—is the much larger magnitude of the absorption and emission cross sections. This leads to a much smaller value of the "pump saturation intensity," making it easier to pump the laser to threshold. In principle, a ZnSe:Cr laser could be pumped efficiently by laser diode arrays whose peak intensity is on the order of a few kW/cm². Thus we recognized the potential of creating a widely-tunable, diode-pumped mid-IR laser. By virtue of the diode pumping, its efficiency would presumably be high, and use of a powerful pump array might lead to a high laser output power (i.e. several watts.)

Table I. Spectroscopic characteristics of Ti:sapphire and ZnSe:Cr²⁺

		Ti ³⁺ :Al ₂ O ₃	ZnSe:Cr ²⁺
Transition		² E → ² T ₂	⁵ E → ⁵ T ₂
Upper-level lifetime	τ _{em} (μsec)	3	9
Peak fluorescence wavelength	λ _{max} (nm)	800	2300
Fluorescence linewidth (RT)	Δν (cm ⁻¹)	4300	1700
	Δλ (nm)	300	1000
Relative bandwidth	Δλ/λ _{max}	0.38	0.43
Peak pump cross- section	σ _{abs} (10 ⁻²⁰ cm ²)	6.5	87
Pump saturation intensity	I _{sat} (kW/cm ²)	2000	14

While there are many applications for tunable IR light—remote sensing, optical countermeasures, and pollution monitoring, to name a few—we felt that medicine (surgery in particular) would benefit from the availability of an engineered ZnSe:Cr²⁺ laser system. The reason is that wavelength tunability could add a new dimension to laser surgery. Laser energy deposition during surgery on biological tissue is largely governed by the absorption coefficient of water, which is strongly wavelength-dependent, especially in the IR region around 2000 - 3000 nm. With a wavelength-tunable laser, a surgeon would be able to control both the laser power and the penetration depth. This would allow optimization of the balance between cutting and coagulation. Both delicate cutting, such as that required for corneal sculpting, and more aggressive surgical procedures would then be possible with the same system. Thus, a ZnSe:Cr laser could potentially replace a suite of other surgical lasers, including the Er:YAG, Er:YSGG, CO₂, and Nd:YAG lasers. Its light could be delivered via a silica optical fiber (or perhaps a sapphire fiber in the longest-wavelength regions,) in contrast to a CO₂ laser, for which good fibers do not yet exist.

Yet another important variable of laser energy deposition during surgery is the timescale in which the energy is delivered. Generally, surgical efficacy improves when energy is deposited much faster than it is conducted away. This is most noticeable when small regions are illuminated, since the heat has less distance to travel to be dissipated. A short-pulse (nanosecond) system could therefore work better than a microsecond-to-millisecond-pulse system. The literature on laser surgery is rife with papers comparing the relative efficacy of lasers at different wavelengths and different pulse durations.

Our LDRD project had several goals pertaining to laser development:

- Demonstrate a widely-tunable ZnSe:Cr²⁺ laser;
- Demonstrate diode-pumped operation of a ZnSe:Cr²⁺ laser;
- Scale the laser's output power up to several watts;
- Obtain short (tens of nsec) pulse operation.

As described in more detail below, we were partially successful in meeting these goals. Briefly, we were able to obtain laser operation over quite a large tuning range covering ~2150 - 2800 nm, and did achieve diode-pumped operation. In fact, the diode array developed for pumping the laser worked quite well. Unfortunately, obtaining a high output power and short-pulse operation are more challenging tasks on which we made only partial progress. However, since other research groups have taken interest in our work and taken up the study of Cr²⁺ lasers, progress will continue to be made, even if not at LLNL.

II. Diode-pumped laser and semiconductor saturable absorber designs

We considered a few different designs for diode-pumped ZnSe:Cr²⁺ lasers. A persistent problem in laser design is the coupling of pump light from a rectangular region (the diode array) to the resonated volume in the laser. In many diode-pumped lasers, end pumping is preferred, and sophisticated beam-shaping optics (including "lens ducts") are used to provide spatial formatting of the pump light. Effective formatting to achieve high on-axis intensities is critically dependent on the divergence of the diode light. Since we were dealing with a hitherto-uncharacterized set of long-wavelength laser diodes (see below,) we felt that end-pumping was a risky option. The diode divergence was not yet known, and lens ducts for long-wavelength light are not routinely available, and could likely be very lossy owing to OH-overtone absorption in wet fused silica. Furthermore, in end-pumped lasers a dichroic mirror is needed to transmit pump light and reflect the resonated beam, and we have not yet had experience working with such mirror coatings on ZnSe for the 1.8 - 2.5 μ m region.

A simpler design that allows facile coupling of light from diode-bar arrays is that of Figure 1, a side-pumped "brick" with a "TIR bounce" of the resonated beam. The diode light is focused with a cylindrical lens to a stripe, creating a ribbon-shaped gain volume in the brick. (Depending on how strongly the pump light is absorbed, the gain region may extend all the way through the crystal.) Antireflection (AR) coatings are applied to the ends of the brick, and since the resonated beam enters and exits at a small distance from the edges of the brick, diffraction losses (due to "scraping") are virtually eliminated. We used the design of Fig.1, popularized by Alcock[1] in all of our diode-pumped experiments.

Fig. 1. Diode-side-pumped laser design, which facilitates integration of a ZnSe:Cr slab and a multiple-bar diode array.

Fig. 2. Multiple-quantum-well (MQW) semiconductor-saturable-absorber passive Q switch.

As a passive Q-switch we had planned to employ a semiconductor saturable absorber[2] (Figure 2.) The saturable-absorber multiple quantum well (MQW) layers are sandwiched between InGaAs buffer layers, and the entire device is supported by an InP wafer. At the wavelengths of interest for the ZnSe:Cr²⁺ laser, the InP and InGaAs layers are transparent. Low Fresnel losses could be obtained by using the wafer at the Brewster angle in the cavity. Such structures have been known for roughly a decade but have not found their way into traditional "energy-storage" solid-state lasers because the intracavity fluences have been too high, causing damage to the semiconductor. Because of a high laser cross section, the saturation fluence of the Cr²⁺ ion is ~ 0.1 joule/cm², roughly 1% that of Ho:YAG and other rare-earth-ion lasers, eliminating this problem. In fact, the saturable absorber's saturation intensity must be below that of the gain medium itself to allow Q-switching,[3] and the MQW saturable absorbers have $I_{\text{sat}} \sim 1 - 10$ kW/cm², comfortably below the Cr²⁺ saturation intensity of ~ 20 kW/cm². Since our laser diode group was already capable of producing high-performance MQW structures, we felt that fabrication of a saturable absorber would play naturally to its strength.

III. Experiments

A. ZnSe:Cr Crystals

To build an efficient laser, we strove to increase the Cr²⁺ concentration while maintaining low loss at the laser wavelength. High doping is especially important in side-pumped lasers, which tend to have poor mode fill unless the pump light is absorbed in a shallow layer near the surface of the crystal. Although several techniques have been used to produce Cr²⁺-doped ZnSe and ZnS crystals, we found diffusion-doping to be a simple and effective method. Undoped blanks are placed with CrSe powder in an evacuated quartz ampoule, which is heated to a temperature of 800 - 1000 C for a few days. The blanks can be either sintered CVD-grown material (commonly used for laser windows, lenses, etc.) or single crystals. Nowadays, single crystals are grown by the Seeded Physical Vapor Transport (SPVT) technique. Diffusion of divalent transition metal ions in zinc chalcogenides is fairly thorough; the interiors of the blanks come out rather uniformly doped. On the other hand, the exact doping mechanism is not known at this time, since CrSe is not expected to volatilize at the diffusion temperatures we employed. It is suspected that volatile impurities (e.g. water) within the ampoules are acting as transport agents.

Table II. Characteristics of Cr-doped crystals used in laser experiments

Crystal Characteristics						
S/N	Origin*	Diffusion-doping conditions/remarks (CrSe unless otherwise indicated)		α_{peak} [cm ⁻¹]	α_{loss} [cm ⁻¹]	FOM
663	ZnSe poly	850°C	5 days	18.	0.65	28
666	ZnSe SPVT	900°C	2 days (FY96 DPSSL sample)	1.86	0.16	12
667	ZnS poly	900°C	3 days	1.35	0.60	2.3
669	ZnSe poly	900°C	2 days (Zn + CrSe in ampoule)	20.	0.77	26
670	ZnSe SPVT	925°C	5 days (Brewster-cut slabs)	5.2		
681	ZnSe poly	900°C	2 days (Zn)/ 900°C 2 days(CrSe)	11.7	0.32	37
690	ZnSe SPVT	900°C	14 days (FY97 DPSSL sample)	6.3	0.13	49
694	ZnSe SPVT	950°C	5 days	12.2	0.18	66
697	ZnSe SPVT	1000°C	5 days	8.7	0.16	56
701	ZnSe SPVT	950°C	5 days (Zn + CrSe in ampoule)	4.2	0.22	19
709	ZnSe SPVT	900°C	14 days (Zn + CrSe in ampoule)	5.3	0.28	19
12435	ZnSe SPVT	As grown material		2.0	0.098	20

* poly: polycrystalline CVD-grown window material; SPVT: Seeded Physical Vapor Transport

To evaluate the doped crystals, we used a spectrophotometer to obtain an absorption spectrum, from which we obtained the 1800 nm pump absorption coefficient α_{peak} . The laser loss coefficient α_{loss} (evaluated at 2350 nm) was derived by measuring attenuation of a $\text{MgF}_2:\text{Co}^{2+}$ laser beam after it traversed a substantial thickness (~ 1 cm) of material. A "figure of merit" $\text{FOM} \equiv \alpha_{\text{peak}}/\alpha_{\text{loss}}$ can be used to describe crystal quality; it is always desired to have a large FOM, and for side-pumped lasers, normally $\alpha_{\text{peak}} > 10 \text{ cm}^{-1}$ is appropriate. Table II shows some results for various doped crystals, including the starting material, doping conditions, pump absorption and laser loss coefficients, and figure of merit. We did achieve fairly high values for α_{peak} in some cases, and the FOM ranged up to 66, which is a factor of at least 3 or 4 improvement with respect to our early work. We are pleased with our ability to create useful laser material with good reproducibility from inexpensive starting materials, using simple techniques. Note, however, that the loss coefficients never got much below 0.1 cm^{-1} , so a round trip through a 1 cm crystal would entail a loss of over 20%. Virgin ZnS and ZnSe materials are prized for their extremely - high transparency and low absorption, even out to wavelengths as long as $10.6 \mu\text{m}$, so there is something associated with Cr - doping that causes loss to appear. Loss reduction remains an ongoing topic of investigation.

B. Long-wavelength diode arrays

We built and tested two different diode arrays during the project. Both were based on long-wavelength (InGaAsP/InP) diode wafers obtained from Epitaxial Products, Inc. The wafers were sectioned into individual emitters and assembled into diode bars, which were then microlensed and combined into arrays. Since the TV camera used for microlensing the "standard" (800 - 900 nm) bars did not detect the long-wavelength light, we set up a special ground-glass viewing screen and mid-IR camera for the new diode bars. Microlensing these bars turned out to be more difficult. Our first array had 4 bars and worked at 1650 nm; the second had 10 bars and generated light at 1725 - 1750 nm.

The first (4-bar, 1650 nm) diode array, when operated at a low duty cycle with a 50 μsec pulsewidth, gave the slope data shown in the lower curve of Figure 3. A maximum diode power of 75 W was obtained at a drive current of 100 amp, and an array-integrated slope of 0.795 W/A, corresponding to a slope for each diode bar of ~ 0.2 W/A. The performance of the second (10-bar, 1725 nm) array was even more impressive—as evident in the upper curve of Figure 3, its output exceeded 200 Watts peak. The array's slope of 2.36 Watt/Amp corresponds to an average slope per bar of ~ 0.24 Watt/Amp. Although this is quite far below the 800 - 900 nm diode bar performance of ~ 0.8 Watt/Amp, it is still very good for this wavelength region.

Fig. 3. Slope plots for two diode arrays used to pump ZnSe:Cr²⁺ lasers.

Fig.4. Intensity distribution of the 10-bar, 1725-nm array at the focus of a 12.7 mm lens.

A razor scan of the the focused spot of the 10-bar array is shown in Figure 4, with the array driven at 70 Amp (100 Watt peak output.) A gaussian width parameter of 280 μm is derived from a fit to this data. Because of mis-aiming of some microlensed bars, not all of the stripes overlapped in the focal region, putting some light in the wings of the distribution. In spite of that, the peak intensity of 1.6 kW/cm² is appreciable with respect to the ZnSe:Cr²⁺ pump saturation intensity of 14 kW/cm². Pulsing the diodes at higher current significantly increases the focal intensity. Improvements in microlensing can probably double the intensity, with no increase in the heat delivered to the ZnSe:Cr brick, leading to diode-pumped laser performance improvements.

The long-wavelength InGaAsP/InP diodes are likely to have applications beyond pumping ZnSe:Cr²⁺ lasers. CW operation may be of interest, and to investigate the CW capability, a test was run on a single bar. It was feared that reliability would be low, but as Figure 5 shows, the output was very steady at 5 Watts for the first 100 hours (at 54 Amp drive current,) then at 8 Watts for the next 700 hours (at 75 Amp drive.) This was a surprise, especially considering that the bar was designed to have a high "fill factor" (small space between emitters,) allowing enough cooling for pulsed, but not CW operation. The implications are that a 10-bar array would deliver 80 Watts CW, and that given good thermal management of a ZnSe:Cr²⁺ laser, it too could run CW. All in all, the diode array development program was very successful.

Fig. 5. Results of CW operation of a single 1725-nm diode bar, at 54 Amp drive for 100 hours, then at 75 Amp for 700 hours.

C. Saturable absorber

Our work on the semiconductor-saturable-absorber passive Q switch positioned us for the growth and testing of MQW structures, and exposed us to some pitfalls. One stumbling block was the lack of transparent InP substrates. Doped substrates (as one would use for producing diode lasers) exhibit quasi-free-carrier absorption that causes serious IR attenuation. Even in thin samples, this renders them virtually opaque to the ZnSe:Cr²⁺ laser, and not suitable for intracavity use. An IR transmission spectrum of an undoped InP wafer (suitable for our purposes) is shown in Figure 6(a). The high refractive index (~ 3.2) of this material causes significant normal-incidence Fresnel losses, as expected. Insertion of this wafer at a steep angle into the cavity of an operating ZnSe:Cr²⁺ laser reduced its output power somewhat, but did not quench lasing. A trial growth of a single quantum well (SQW) ~ 70 Å thick sandwiched between several-micron thick InGaAs barrier layers resulted in a structure whose transmission spectrum is shown in Figure 6(b). The new features in the spectrum can be attributed to the InGaAs layers; the SQW is so thin as not to affect the spectrum noticeably. The ~ 2200 nm target bandgap of the layers is evident as a steep rise in transmittance at that very wavelength. Our ability to tailor the bandgap validates the calibration of the MOCVD growth apparatus and methods. Transmission ripples in the 2200+ nm region are due to etalon effects in the InGaAs layers; the free-spectral range is evidently ~ 100 cm⁻¹. Opacity in the region below 2000 nm is consistent with an above-bandgap absorption coefficient on the order of 10^4 cm⁻¹. Laser performance testing of a similar structure could occur in a straightforward manner if the single quantum well were replaced with a series of many (say, up to ~ 50) quantum wells.

Fig. 6. Transmission spectra of undoped InP wafers (a) before and (b) after deposition of a single-quantum-well (SQW)/barrier-layer sandwich.

D. ZnSe:Cr laser results

One of our goals was to demonstrate wide tunability of a ZnSe:Cr²⁺ laser. Although we had done some tuning experiments with a birefringent tuner, we realized that a diffraction grating would provide simpler tuning and wider spectral coverage. The laser's gain was sufficient to overcome the additional loss introduced by a grating. Grating-tuning experiments were done by replacing the cavity high-reflector with a 420 line/mm diffraction grating on a rotation stage, and using curved output couplers. The diode array was removed and a pump beam from a MgF₂:Co²⁺ laser was focused onto

the crystal using the same cylindrical lens. Since the $\text{MgF}_2:\text{Co}^{2+}$ laser has a low-divergence beam, its brightness is much higher than that of the diode array, making it easier to reach threshold with a lossy cavity element (i.e. grating.) Output wavelengths were checked with a monochromator. According to the tuning curve in Figure 7, the long-wavelength limit of operation appears to be ~ 2670 nm, but in fact the OH - induced absorption in the BK - 7 mirror substrate blocked the laser output, which actually extended to 2799 nm. The short-wavelength cutoff was 2134 nm; even though the emission cross section remains substantial, self-absorption most likely inhibits laser operation. Higher-power pumping of this laser may well extend the tuning range at both the long- and short - wavelength ends. With the laser running at ~ 2600 nm, we measured its linewidth at less than 1 nm. Such a resolution is adequate for various condensed-matter studies, including sublimation of solid hydrogen in ICF target capsules. Considering that there was no beam expander to increase the number of illuminated grating grooves, and no additional line-narrowing elements (e.g. étalons), the laser should ultimately be capable of very-high-resolution operation, making it suitable for IR spectroscopy and remote sensing. The demonstrated wide spectral coverage (out to almost 2800 nm) represents a validation of our claims regarding the tunability.

Fig. 7. Tuning characteristics obtained with $\text{MgF}_2:\text{Co}^{2+}$ laser pumping of ZnSe:Cr, resonated with a diffraction grating. The output - coupler transmittance spectrum reveals the OH absorption in the BK - 7 glass substrate material that blocked the laser's output from ~ 2650 - 2800 nm.

We have performed some experiments to test the power output with diode pumping. Under short-pulse (50 μsec) conditions, with a low duty cycle, we obtained the data in Figure 8. Using crystal #690 ($\alpha_{\text{peak}} = 6.3 \text{ cm}^{-1}$; $\alpha_{\text{loss}} = 0.13 \text{ cm}^{-1}$) in a near-confocal cavity with 10-cm mirrors, and a series of 3 different output couplers, we found that the 85% reflector gave the highest output, at slightly over 3 Watts peak. This corresponds to an absolute optical-to-optical efficiency of 2% and an optical-to-optical slope efficiency of 3%. (Figures given are ZnSe:Cr $^{2+}$ output with respect to diode-array output.) The low efficiencies can be blamed largely on the poor mode overlap between the pump volume and the resonated wave; in fact, much of the pump light would pass all the way through a crystal 1 mm thick! Our experiments done with end-pumping using a $\text{MgF}_2:\text{Co}^{2+}$ laser have shown that slope efficiencies over 25 - 30% (with respect to absorbed pump energy) can be obtained when the mode-matching is good.

Fig. 8. Peak output power vs. diode-array drive current in a near-confocal resonator with three different output couplers, and crystal #690.

Fig. 9. Average power vs. duty cycle for crystals #690 and #701.

Average-power test results are shown in Figure 9. The diode-pumped laser was run with 50 μ sec pump pulses whose repetition rate was increased to raise the duty cycle. For example, 200 Hz operation corresponds to a pump duty cycle of 1%. During the experiments, the duty cycle was increased until "rollover" was encountered. In Figure 9(a), crystal #690 was again tested, and in the low-duty-cycle portion of the plot, the peak power of ~ 3 W was maintained. At a 1% duty cycle, the power had fallen from the expected 30 mW, to a value ~ 15 mW. A similar experiment using a "worse" crystal (#701; $\alpha_{\text{peak}} = 4.2 \text{ cm}^{-1}$; $\alpha_{\text{loss}} = 0.22 \text{ cm}^{-1}$; lower doping, and higher loss) showed that at low duty cycle, the power was lower, but that the rollover did not occur until a duty cycle $\sim 2\%$. This allowed the laser to produce a higher average output power over 20 mW. Our interpretation of these results is that the heat load on the crystal (~ 2 W at 1% duty cycle) is high enough to cause thermal lensing that destabilizes the laser cavity. Even if the gain per pass at low drive is reduced, a lower-doped crystal experiences a smaller temperature gradient in the gain region, mitigating thermal lensing. ZnSe, being a semiconductor whose band gap declines with increasing temperature, exhibits a comparatively-large value of $dn/dT \sim 7 \times 10^{-5} \text{ K}^{-1}$. In spite of the high thermal conductivity of ZnSe (0.17 W/cm-K,) the thermal-lens focal length (in the vertical axis) is estimated at only a few cm

when 2W pump power is absorbed. Horizontal-axis focusing is much less severe in the side-pumped geometry employed to date. Use of a shorter cavity (than the 5 - 10 cm lengths tried to date) may well allow operation at higher duty factor and higher output power. Typical crystal lengths of ~1 cm do allow for cavity shortening. However, realization of the full average-power potential of ZnSe:Cr²⁺ lasers mandates another look at the cavity configuration. Perhaps a monolithic, end-pumped design, a waveguide, or an "active mirror" configuration will cope better with thermal lensing.

IV. Discussion

Wide tunability and diode-pumped operation of ZnSe:Cr²⁺ lasers have been well demonstrated in our laboratory. There is still important work to do in the quest for higher average power. New resonator designs that mitigate thermal lensing and provide good "mode fill" will be needed. CW lasing and ultra-short-pulse generation are potential operating regimes we have not reached yet, but they will probably be demonstrated within a year or two, likely by our colleagues in other laboratories.

Interest in Cr²⁺ lasers in other parts of the solid-state-laser community has been high, and there have been spectroscopic studies of Cr²⁺-doped CdSe and CdMnTe, and laser experiments reported on CdMnTe:Cr²⁺. [4] Transition-metal-doped zinc chalcogenides can also function as saturable absorbers (Q switches.) Collaborations between LLNL and various groups worldwide have begun, for the purposes of producing Cr²⁺-doped samples and exploring some of the desirable properties of the new laser materials. Crystal-growth companies (e.g. Cleveland Crystals) are now selling Cr²⁺-doped material on a custom basis. So, with time, we should be seeing all kinds of interesting progress in mid-IR solid-state lasers, as a result of the original work done at LLNL.

V. Bibliography

In the course of the projects, we published several refereed papers and obtained a patent; a list follows. We also gave numerous conference presentations, some invited.

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VI. Acknowledgements

Many people have contributed to the success of our ZnSe:Cr²⁺ laser projects. Bill Krupke and Steve Payne initiated our program; Laura DeLoach and Gary Wilke participated in the first spectroscopic and laser experiments; Prof. Arnold Burger taught us about diffusion-doping; Kathleen Schaffers and John Tassano have done numerous diffusion-doping growth runs; Jay Skidmore, Mark Emanuel, Barry Freitas, Victor Sperry, Everett Utterback, Larain Di Mercurio, Cathy Reinhardt, and Joe Satariano have produced the long-wavelength diode arrays and saturable absorbers; John Lang has produced a variety of machined parts; Falgun Patel, Randy Bartels, and Phil Waide have helped with laser experiments; Ron Vallene and Peter Thelin have taken care of cutting and polishing crystals; Ray Beach spearheaded the laser design and modeling; and Howard Powell has provided moral support.

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