UCRL-JC-124015
PREPRINT

CONF-960848--31

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This paper was prepared for submittal to the
1996 International Symposium on Optical Science, Engineering and Instrumentation
Denver, CO
August 4-9, 1996

June 1996

Lawrence
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New tunable lasers for potential use in LIDAR systems

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ABSTRACT

We discuss the optical and laser properties of two new tunable laser crystals, Ce:LiSAF and Cr:ZnSe. These crystals are unique in that they provide a practical alternative to optical parametric oscillators as a means of generating tunable radiation in the near ultraviolet and mid-infrared regions (their tuning ranges are at least 285 - 315 nm and 2.2 - 2.8 microns, respectively). While these crystals are relatively untested in field deployment, they are promising and likely to be useful in the near future.
Keywords: Ce:LiSAF, Cr:ZnSe, tunable solid state laser

1. INTRODUCTION

The ready availability of tunable laser light sources invites the possibility of using lasers to selectively detect the presence, concentration, and distribution of certain species within the atmosphere. The tunability can be advantageous to avoid atmospheric absorption, or to tune on and off the molecular resonances. The chief means of generating tunable radiation today entails the use of optical parametric oscillators, or OPOs. While OPO devices offer tremendous flexibility in wavelength coverage, they require special attention to alignment, damage threshold, and other issues to operate effectively. Directly tunable solid state lasers, where available, could serve as a more convenient and robust approach, especially if they can be pumped directly by laser diodes.

Titanium-doped sapphire (Ti^{3+}:Al_2O_3), alexandrite (Cr^{3+}:$BeAl_2O_4$), and Cr:LiSAF (Cr^{3+}:$LiSrAlF_6$) and other well-known laser materials provide tunable laser radiation in the near-infrared region.[1] Many applications, including remote sensing and lidar, are predicated on the availability of these materials. Less available are tunable laser materials that operate in the mid-infrared and in the near ultraviolet. In fact, in the past there has been a belief that robust materials with these particular characteristics might be difficult if not impossible to identify. This is the situation because most tunable ultraviolet laser materials suffer from solarization (i.e. coloring) and from excited state absorption, while potential mid-infrared gain media are generally afflicted by nonradiative decay (thermal quenching), rendering them non-emission at room temperature. Within the last few years, however, we have been able to identify and develop these types of laser crystals, and prove that they can operate robustly in the UV and mid-IR. In particular, we have determined that cerium-doped LiSAF (Ce^{3+}:LiSAF) can serve as a practical UV laser,[2,3] tunable between 285-315 nm, and that Cr:ZnSe can operate in the range of 2.2-2.9 microns.[4,5] We expect these crystals to figure more prominently in the design of laser systems for remote sensing as they become more well-known to researchers.

2. CE:LISAF TUNABLE ULTRAVIOLET LASERS

The viability of Ce^{3+}-lasers has long depended on the need to identify a host medium that did not excessively color under conditions where ultraviolet light was employed to pump the laser crystal. After years of disappointing results from various crystalline hosts, Dubinskii and coworkers found that cerium-doped $LiCaAlF_6$ did not solarize substantially, presenting the opportunity to devise the first practical tunable UV gain medium.[6] Since then, the chemical analog $LiSrAlF_6$ (LiSAF) has been considered as well.[2,3,7] Ce:LiSAF is most conveniently pumped at 266 nm by the fourth harmonic of a Nd:YAG laser, as deduced by the absorption and emission spectra presented below in Fig. 1. Furthermore, it may be expected to lase in the region covered by the emission band. Figure 2 displays the actual gain measured by employing a pump-probe type of experimental set-up, revealing gain in the 280 - 320 nm region. It is also clear that Ce:LiSAF must be pumped with π polarized light in order to yield the maximum possible efficiency. The π-polarized operation is needed to avoid the negative influence of excited state absorption at both the pump and the laser wavelengths. Our publication contains a tabulation of the crucial parameters needed to model the performance of this system, (i.e. cross sections, lifetime, etc.).

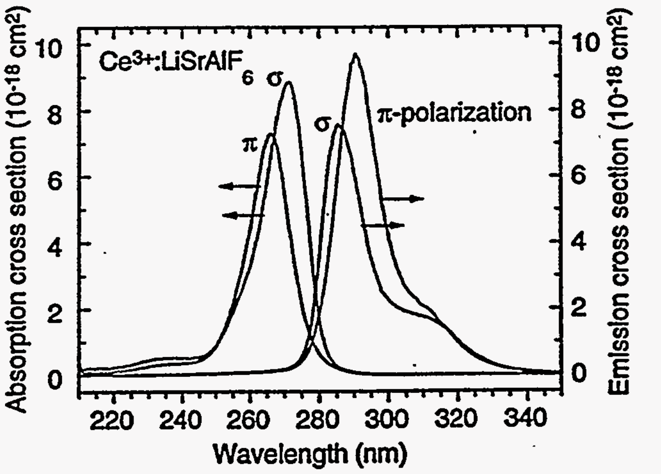

Figure 1: Absorption and emission spectra of Ce:LiSAF.

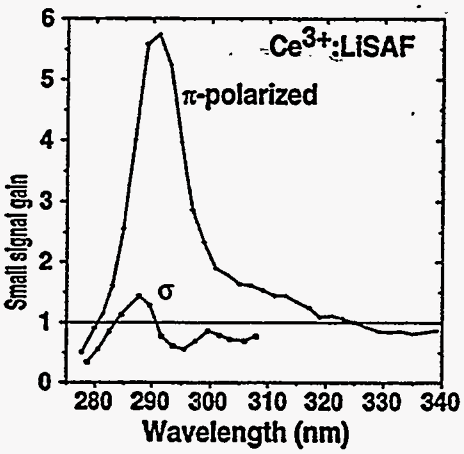

Figure 2: Pump-induced gain spectrum of Ce:LiSAF, showing that the π-polarized gain is far greater than that of the σ-polarization.

The laser slope efficiency was measured with a simple longitudinally-pumped confocal resonator, using a crystal obtained from Lightning Optical Corporation. The best laser slope efficiency obtained was 33% relative to the absorbed pump energy, as pictured in Fig. 3. This particular data was acquired using 50% output coupling. While the laser is found to be relatively efficient, there nevertheless appears to be some energy loss that has not been accounted for. Further experimentation indicates that solarization losses, while comparatively small, still have some influence on the efficiency of the laser. In particular, all the crystals experienced a pump-induced absorption feature extending from 250 to 550 nm, arising from a color center. The amount of color center absorption has been found to be directly correlated with the maximum slope efficiency that is attainable. In the plot pictured in Fig. 4 we see that the pump-induced losses associated with the solarization can be mitigated further by introducing the second harmonic (532 nm) of the Nd:YAG laser along with the fourth harmonic, since this secondary pump beam is capable of annihilating the color centers. As an example of this procedure, consider the data pictured in Fig. 4, where the 532 nm fluence is raised as the 266 nm pump is held constant. The crucial result is that the 290 nm output of the Ce:LiSAF laser oscillator is seen to rise with the 532 nm energy increase. In a separate series of experiments we were able to show that the enhanced performance is directly due to the destruction of the residual color centers that are present. With respect to the absorbed power at 266 nm, the simultaneous introduction of the 266 nm pump beam with the 532 nm "anti-solarant" beam, leads to slope efficiencies as high as 47%. In contrast to the early results obtained for Ce-lasers based on other hosts of <1%, we can now confidently note that Ce:LiSAF can offer the reliability and performance levels needed to develop reliable systems for remote sensing and other applications.

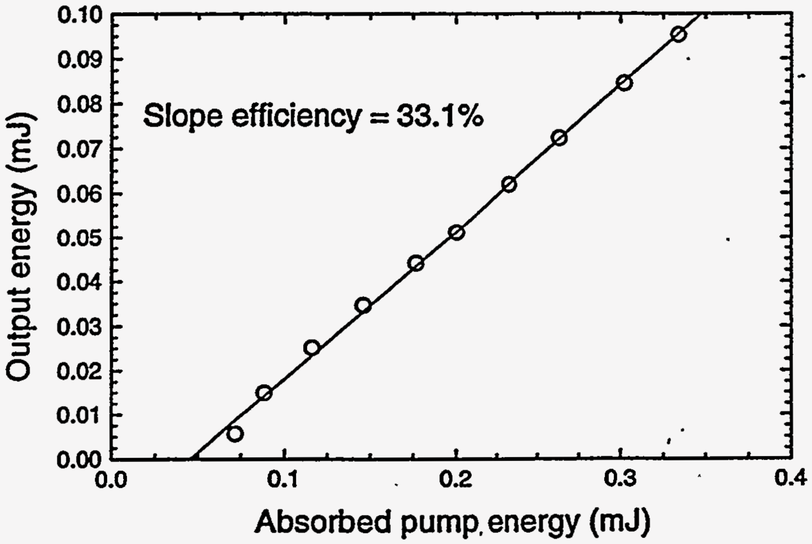

Figure 3: Measured output of a Ce:LiSAF laser plotted against the 266 nm absorbed pump energy, yielding a slope efficiency of 33.1%.

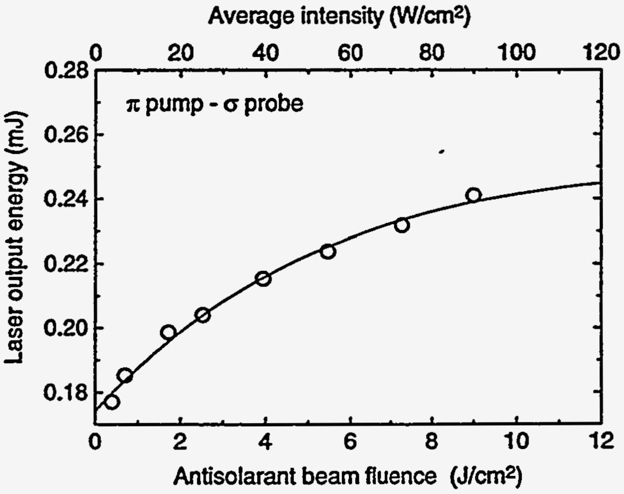

Figure 4: 290 nm output power of Ce:LiSAF laser as a function of 532 nm anti-solarant beam, with constant pump power.

3. ZNSE:CR TUNABLE MID-INFRARED LASERS

Remote sensing applications in the mid-infrared wavelength region have generally been regarded as the domain of OPOs, since direct solid state laser gain media were very limited, gas-lasers can only generate specific laser lines, and diode lasers are low power and require cryogenic cooling to operate. While OPOs have much to-offer for remote sensing, the potential availability of direct generation by a diode-pumped solid state laser has much appeal, due to the anticipated compactness, flexibility, and efficiency. With this motivation in mind, we have been pursuing a high risk approach by trying to identify a new class of solid state lasers with the following characteristics: efficient room temperature operation, tunable beyond 2 microns, and pumpable by diodes. The outcome of this search has been the discovery that divalent chromium in ZnSe offers laser action in the 2.2 - 2.8 micron range (and beyond), based on the $^5T_2$ - ^5E electronic transition of Cr^{2+} occupying tetrahedral substitutional sites.[4,5] While this novel approach to generation of laser light is at an early stage, many of the fundamental signatures of Cr:ZnSe are quite promising and warrant continued attention.

The absorption and emission spectra of Cr:ZnSe are plotted in Fig. 5, revealing that the material can be pumped near 1.8 microns, and should lase from about 2.2 microns to beyond 3 microns. Notice that the cross sections are on the order of 10^{-18} cm^2. The emission lifetimes are shown in Fig. 6, where they appear as a function of temperature. Because the lifetime does not precipitously fall until the temperature rises above room temperature, this data suggests that nonradiative decay does not compete significantly with radiative emission at room temperature. In other words the emission is efficient at room temperature, providing the potential for laser operation at the lowest possible threshold, (or allowing greater energy storage for a given pump intensity). This observation of high luminescence efficiency in the 2 - 3 micron region, broadband, is unusual if not entirely unique.

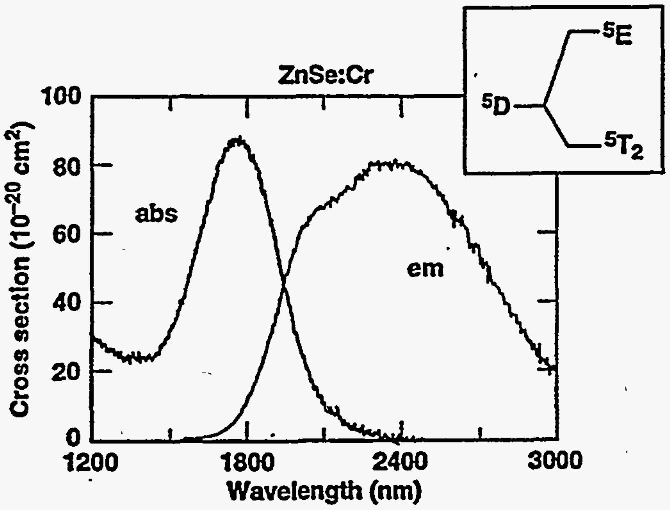

Figure 5: Absorption and emission spectra of Cr:ZnSe.

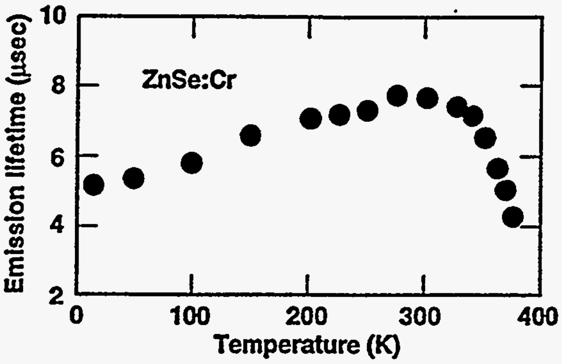

Figure 6: Emission lifetime of Cr:ZnSe as a function of temperature.

The laser pumped laser efficiency of Cr:ZnSe is shown in Fig. 7, where a MgF_2:Co laser has been used as the excitation source. Using a simple longitudinally-pumped oscillator, a slope efficiency as high as 30% was attained (pumping at 1.8 microns, lasing at 2.3 microns). This slope efficiency was compared at a number of different output couplers to determine that excited state absorption does not seriously degrade the performance of the laser. This finding is actually not surprising since only the ground and lowest excited states are spin quintets, while all higher lying electronic states are triplets and singlets. So in other words Cr^{2+} in tetrahedral sites has an intrinsic electronic structure where the laser transition is spin-allowed, and all excited state absorptions are necessarily spin-forbidden.

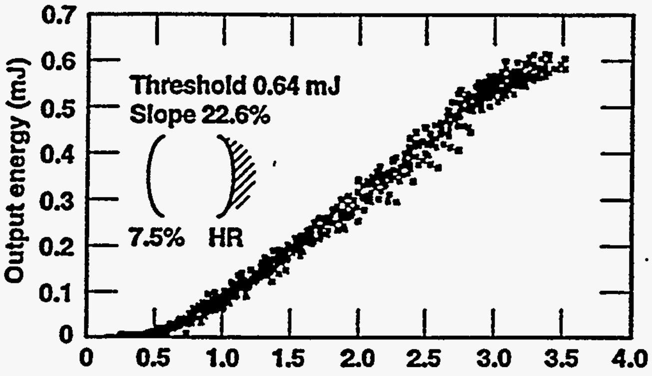

Figure 7: Laser-pumped laser output curve, for pumping near 1.8 microns.

Another area of research has been our finding that laser quality Cr:ZnSe crystals can be prepared by diffusion-doping -- with either single crystal ZnSe or with polycrystalline (pressed) window material. In this methodology CrSe powder is included with the ZnSe in a quartz ampoule, and the vessel is held at about 800 degrees Celsius for a few days. The interesting result is that low-cost ZnSe windows (e.g. as used for CO_2-lasers) can be diffusion-doped to produce laser material. This idea invites consideration of the use of very large gain elements if needed. At this time, the losses of the Cr:ZnSe optical elements are still rather high, on the order of 10%/cm, due to a combination of Se precipitates and Fe impurities. We are in process of improving the crystalline quality and see no major impediments to obtaining low-loss material (as we have already obtained on occasion).

We have begun to develop a diode-pumped solid-state laser based on the Cr:ZnSe gain medium. To do this, we fabricated a prototype laser diode array that generates light near 1.63 microns, and constructed a four-bar stack; the device is pictured in Fig. 8. We have reached threshold with this laser module and are encouraged by the performance. At this juncture the laser diodes can only be operated at very low duty factor. The peak power obtained from the Cr:ZnSe laser was 0.34 Watts, and we think that our Cr:ZnSe diode-pumped solid state laser design can ultimately offer several Watts of power, once the laser diode array technology can be operated at high duty factor.

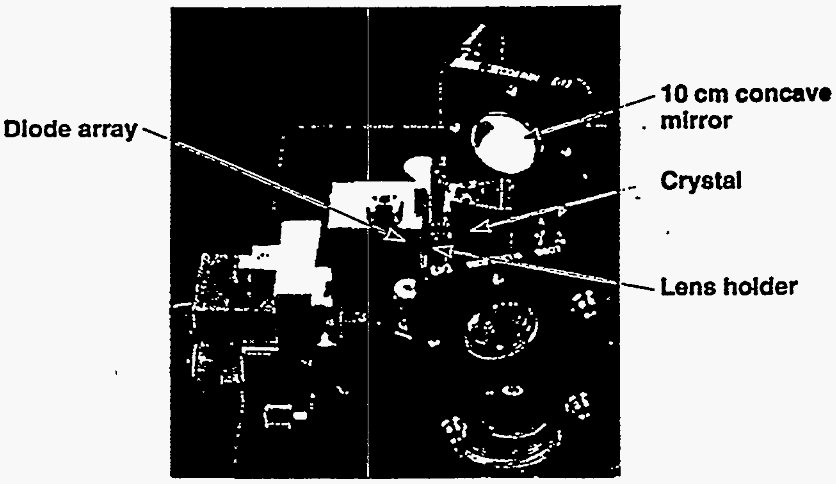

Figure 8: Photograph of first-generation prototype Cr:ZnSe diode-pumped solid state laser.

4. SUMMARY

We have reported on our efforts to extend the wavelength range of tunable solid state gain media into the ultraviolet and the mid-infrared. Although these wavelength ranges are beset by fundamental complications (excited state absorption and solarization in the ultraviolet, nonradiative decay in the mid-infrared), special optical materials have been identified where these impediments are overcome to a substantial degree, such that the identified materials appear to be practical. If one considers the "enabling" nature of new laser materials, their potential pay-off can be enormous and far-reaching. We think that Ce:LiSAF tunable ultraviolet lasers and Cr:ZnSe tunable mid-infrared lasers will present many new opportunities for remote sensing applications.

5. ACKNOWLEDGMENTS

We gratefully acknowledge the laser diode array produced by Jay Skidmore and the members of the diode group at LLNL, Professor Arnold Burger for the collaboration with the diffusion-doping process, and Greg Quarles and his team at the VLOC Division of II-VI Corporation for Ce:LiSAF crystals. This work was performed under the auspices of the US Dept. of Energy by Lawrence Livermore National Laboratory under contract number W-7405-ENG-48.

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