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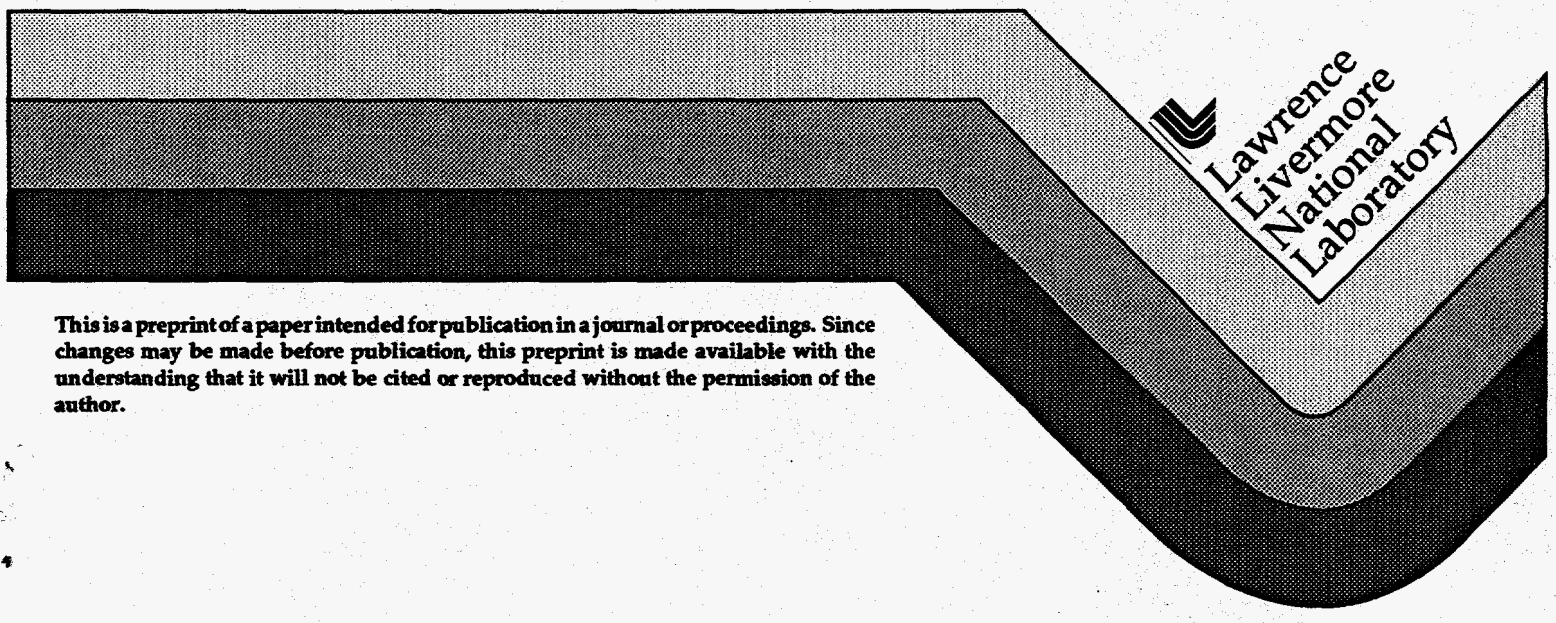

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New Solid State Lasers from the Ultraviolet to the Mid-Infrared

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Abstract

We discuss three new laser materials that offer improved access to the ultraviolet, near infrared and mid-infrared spectral regions. In order for each of these materials to have been identified, a particular hurdle needed to be overcome with respect to the fundamental laser physics impacting the material. In the case of the 280-320nm Ce:LiSAF laser, the main issue is the need to reduce the loss associated with excited state absorption, while for 1047nm Yb:S-FAP it is the ground state absorption at the laser wavelength that must be minimized. Cr:ZnSe has been down-selected from a number of potential candidates which could lase in the 2200-3000nm region, in order to mitigate the detrimental impact of nonradiative decay. In all three cases we discuss how appropriate consideration of fundamental concerns has led to the identification and understanding of the new laser system.

1. Introduction. One of the major goals facing the laser community has been to develop means of conveniently producing laser light from the mid-infrared to the ultraviolet. In the past there has been a very broad range of wavelengths demonstrated, but most of these laser sources have not proved to be practical devices. It turns out, however, that the insights derived from solid state physics, material science, and laser physics can now be collectively engaged to develop new laser media that are of practical interest to the user community. In other words, while the new laser media discussed in this paper are at a pre-commercial stage of development, we believe that these particular material systems offer exceptional prospects for future deployment in real laser applications. Now, although the overall utility of a laser material is potentially impacted by a very large number of factors other than the fundamental physics (e.g. crystal growth, thermomechanical properties, cost, etc.), this paper will address only the fundamental areas at this time (references quoted herein can be reviewed for greater detail).

Figure 1 reveals the fundamental physics that can affect the utility of a laser material. While there are many other issues of crucial importance to the gain medium, this article will focus on three aspects of the laser physics: excited state absorption, ground state absorption at the laser wavelength, and nonradiative decay of the metastable upper laser level, as illustrated in the figure.

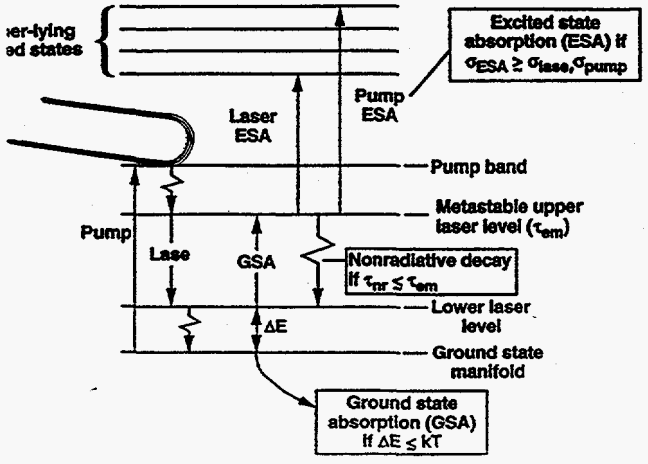

Fig.1: Energy level diagram depicting ground and excited state absorption, nonradiative decay, and other fundamental processes.

2. Ce:LiSAF lasers.[1] Excited state absorption (ESA) can detract from both the gain and the pumping efficiency, depending on whether the ESA exists at the laser or the pump wavelength. In the case of Ce:LiSAF (Ce^{3+}-doped into $LiSrAlF_6$), it turns out that both mechanisms are important to the performance and efficiency of the laser material. This situation can be addressed by first examining the absorption and emission spectra contained in Fig. 2. The absorption spectra may be interpreted to indicate that the material can be pumped in the range of 255-275nm, and that it should be relatively insensitive to the polarization of the pump source. (The $LiSrAlF_6$ host is uniaxial in character, and therefore exhibits the so-called p unique axis and the s plane perpendicular to it.) Furthermore, the emission spectra offer similar cross sections in the two polarizations, suggesting that we should expect similar gain in both cases.

As revealed in Fig. 3 however, it is apparent that the selection of either p or s polarized pump or probe light has a dramatic impact on the performance of the laser. The experiment employed to obtain the data of Fig. 3 is that where separate pump (266nm Nd:YAG) and probe (290nm dye) laser beams are arranged to impinge on the sample, and where the probe field is weak compared to that of the pump. This plot of laser gain versus the pump fluence reveals that the most favorable situation is one where the pump and probe are both π-polarized. For the cases where either the pump or the probe is σ-polarized we observe a very substantial decrease in the amount of gain that is achieved. These observations can be explained by suggesting that the loss process involves ESA at the pump and the probe wavelengths, and that the ESA is strongly σ-polarized. In this way, it is advantageous for the pump source and the laser to be π-polarized, to avoid the ESA losses. We conclude that it is feasible to overcome most of the ESA problem in the case of Ce:LiSAF by carefully choosing the polarizations of the laser beams.[1]

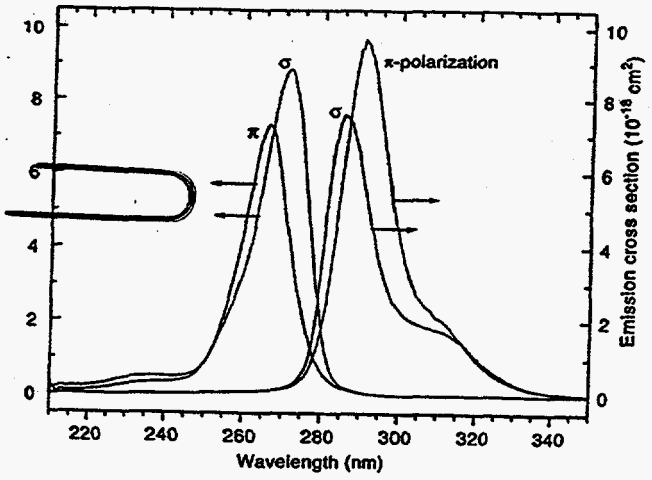

Fig.2: Absorption and emission spectra of Ce:LiSAF

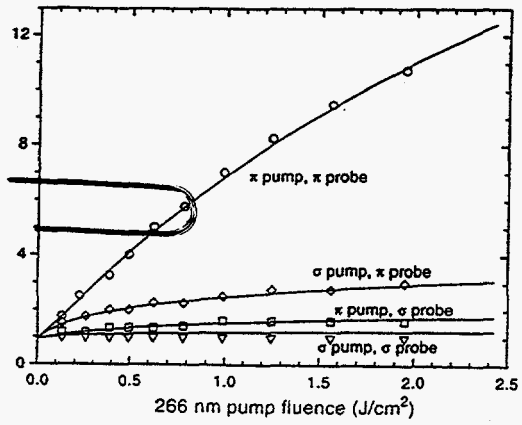

Fig.3: Pump-probe experiment of Ce:LiSAF, illustrating importance of the polarization in determining the level of gain that is present.

It is possible to understand the nature of the broad ESA feature that is under the absorption and emission bands. On the basis of previous studies of rare earth ions, the ESA has become understood to arise from a transition in which an electron is transferred from the Ce^{3+} ion to the conduction band of the $LiSrAlF_6$ host.[2] Upon examination of the host crystal it is noticed that the Sr^{2+} cations are arranged in planes perpendicular to the c axis of the lattice. Since the lowest conduction band component is most likely to be composed of the Sr ions, it may be suggested that the ESA is expected to be σ-polarized (or along the direction of the electron transfer step).

3. Yb:S-FAP lasers.[3-7] We have pursued the identification of a new class of Yb-lasers. Since Yb generally lases at nearly the same wavelength as the much better characterized Nd-lasers, one would naturally inquire as to what the potential advantages of ytterbium are over the neodymium-based alternatives. The main benefits are derived from the longer emission lifetime (3-5x) and the reduced quantum defect (0.3-0.5x). Since the emission lifetime impacts the energy storage that is possible for the material, ytterbium-doped media are roughly capable of storing 3 to 5x more energy for a given pump rate. For situations where the delivered energy per pulse from the laser is a specified value, and where the cost of the pump is important (e.g. as for laser diodes), the Yb-doped media can present the greatest advantage over Nd. The quantum defect between the pump and laser photons is also noteworthy, since the smaller difference characteristic of Yb leads to reduced heating of the gain medium. On the other hand, lasers based on the ytterbium ion tend to operate as quasi-three-level systems so the level of ground state absorption at the laser wavelength is an important aspect of the laser physics. Furthermore, Yb-doped crystals and glasses tend to exhibit rather low cross sections, rendering it difficult to efficiently extract the stored energy from amplifiers (or alternatively, reach threshold in laser oscillators). We have sought to identify new laser materials that would emphasize the favorable characteristics of Yb while minimizing the potential problems.

We have studied a variety of Yb-doped crystals in order to identify those materials that appear most promising with respect to the ground state absorption and the gain cross section. In this vein the figure-of-merit plot pictured below in Fig. 4 was devised. Here the parameter indicated as I_{min} serves the role of quantifying the difficulty of overcoming the ground state absorption at the laser wavelength. (It is actually the product of the pump saturation intensity and the fraction of ions that must be inverted, to yield transparency at the laser wavelength.) A simple explanation of I_{min} is that it is the minimum absorbed pump intensity required to reach threshold in an otherwise lossless resonator (using a lightly doped sample). The objective of our search as defined by Fig. 4 is to identify the Yb-doped materials that possess the maximal gain cross section and the smallest value of I_{min}. Upon inspection of the crystals characterized and included in the figure, we see that the family having the apatite structure is the most promising laser material candidates, including $Ca_5(PO_4)_3F$, $Sr_5(PO_4)_3F$, mixtures of these two compounds, and the related vanadate derivatives.

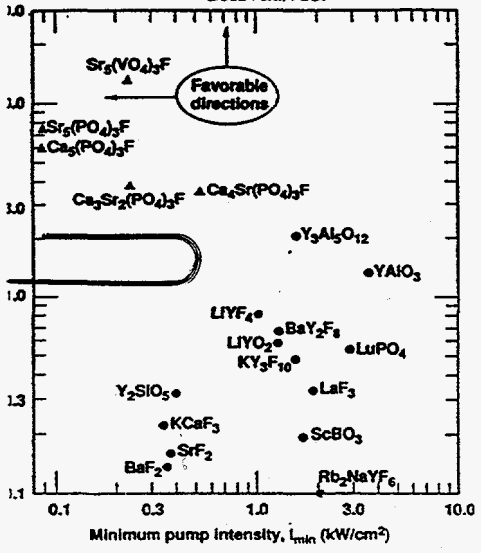

Fig.4: Figure-of-merit plot of the gain cross section against the minimum pump intensity.

The absorption and emission spectra of Yb in strontium fluorophosphate (Yb:S-FAP) are drawn in Fig.5. Here the pump line is seen to be at 900nm, while the laser line is at 1047nm. It's interesting to see that the absorption level at 1047nm is less than 5% of the gain cross section; this useful situation is reflected by the low value of I_{min} seen in the figure-of-merit plot of Fig.4. The spectra of Fig. 5 are striking in terms of the sharpness of the pump and laser lines, in that they appear to be subject to very little phonon broadening compared to nearly all other Yb spectra in crystalline media.

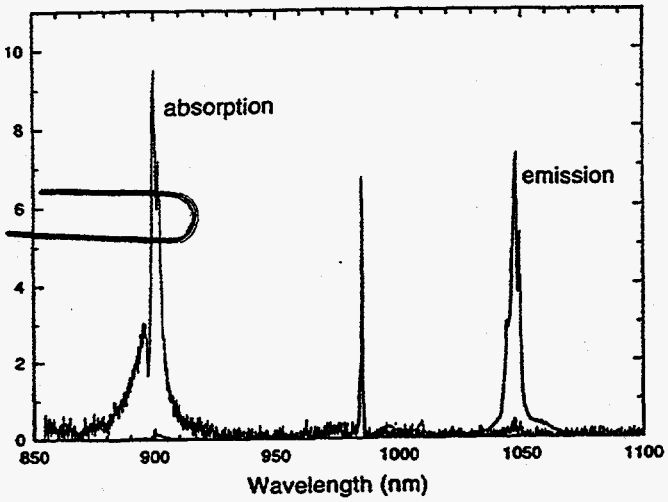

Fig.5: Absorption and emission spectra of Yb:S-FAP.

We have explored the laser performance of Yb:S-FAP and its compositional analogs using laser pumping (Ti:sapphire) and have measured efficiencies as high as 70%. Having scientifically confirmed the utility of the Yb:apatite lasers, we next developed a laser diode array pumped Yb:S-FAP oscillator and operated the 3x3x30mm rectangular element at over 13 Watts of output power, as shown below in Fig. 6. To construct this laser we fabricated a diode array that produces 3 kWatts of peak power, and then attached microlenses to the laser diode facets to collimate the light in the fast axis. A lens duct was employed to concentrate the light and direct it into the laser rod so that it was captured (by total-internal reflection). The laser rod was held between two water-cooled copper blocks to manage the waste heat that was generated. The overall electrical efficiency of the system is more than 10%. We believe that it should be possible to attain about twice this power and efficiency by securing higher quality Yb:S-FAP laser rods.

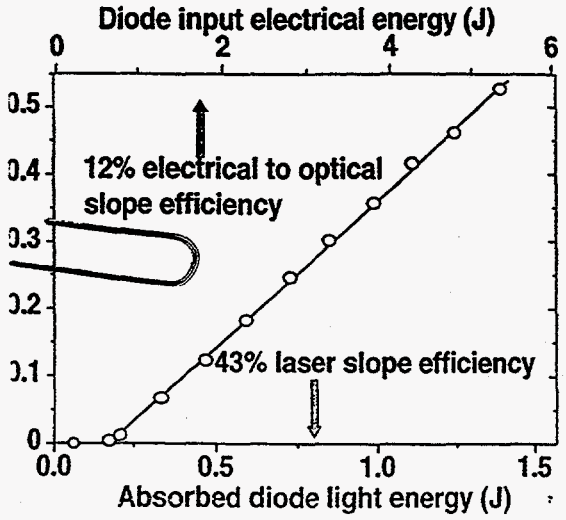

Fig.6: Laser performance of a laser diode array pumped Yb:S-FAP oscillator.

4. Cr:ZnSe lasers.[8] One of the challenging problems in laser science has been the development of systems that can generate tunable laser light in the mid-infrared spectral region. In order to accomplish this objective, optical parametric oscillators (OPOs) have been pursued. OPOs operate on the basis of nonlinear interactions, and therefore exhibit substantial sensitivity in their operating characteristics. Although these systems have proved to be useful and versatile, some technical circumstances call for the availability of direct tunable mid-infrared radiation from a diode-pumped solid state laser, since the system is expected to be more robust. Unfortunately no efficient lasers existed to permit direct generation of tunable infrared laser radiation. The main impediment to identifying mid-infrared tunable laser materials had been the predominating influence of nonradiative decay in most optical materials, which leads to a low emission quantum yield and high threshold of laser operation. We have uncovered a new class of laser materials that have a high quantum yield in the mid-infrared, based on Cr^{2+}-doped ZnSe crystals (and other II-VI hosts).

The absorption and emission spectra of Cr:ZnSe are displayed in Fig. 7, where it is seen that the features are quite broad. We can pump the Cr^{2+} ion at about 1800nm, and tunable radiation should be available from 2200-3000nm. The broadness of the spectra is a consequence of the strong interactions of the nearest-neighbor selenide anions with the 3d-3d transitions of chromium. Since the concept of employing divalent chromium as the laser ion as well as using ZnSe as the host medium were both previously untested, we sought to perform a scientific demonstration exercise where it was conclusively shown that Cr:ZnSe could offer gain and reach threshold in a laser cavity. To accomplish this, we used the "Cobra" laser from Schwartz Electro-Optics to pump a crystal in a 20cm confocal oscillator, and achieved laser action as pictured below in Fig.8. The untuned output from the laser was at 2350nm, as would be expected from the emission peak position shown in Fig.7.

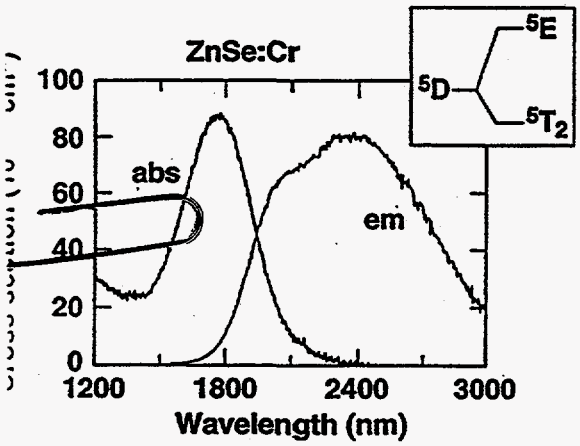

Fig.7: Absorption and emission spectra of Cr:ZnSe, revealing the pump band at 1800nm and the emission band peak at 2350nm.

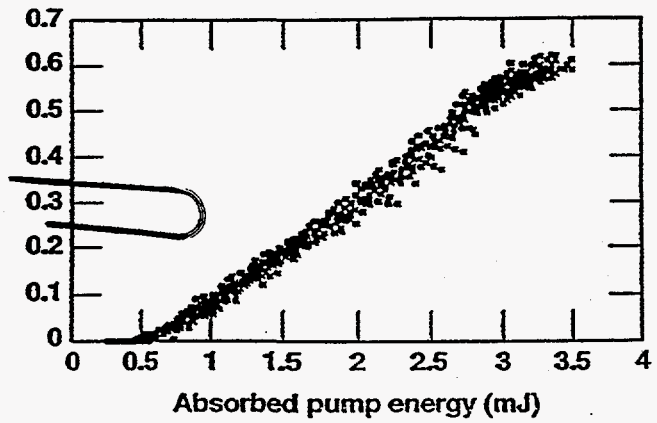

Fig. 8: Scientific demonstration of laser action for Cr:ZnSe pumped at 1700nm and generating laser light at 2350nm.

From the data displayed in Figs. 7 and 8, we can conclude that a new class of lasers has been identified, which is exceptionally promising for several reasons. First, this laser material and its chemical analogs are the first systems found to emit efficiently in the mid-infrared with a broad spectral band. Second, the ZnSe host is thermomechanically robust and should serve adequately in many types of laser systems. Third, a substantial commercial element already exists regarding the manufacture of ZnSe optics. Finally, the preferred pump wavelength of about 1800nm it accessible on the basis of known laser diode structures. We believe that the Cr:ZnSe laser demonstration is the beginning of a new approach to generating mid-infrared laser light.

5. Summary. We have discussed three novel laser materials first lased at Lawrence Livermore National Laboratory which offer access to the ultraviolet, infrared, and mid-infrared spectral ranges. The laser physics of each of these materials was addressed at a fundamental level in order to provide optimal performance. For the case of the 280-320nm tunable Ce:LiSAF laser, excited state absorption at both the pump and laser wavelengths was determined to be important, although we found that it was possible to mitigate its impact by employing π-polarized light. Yb:S-FAP operates at 1047nm (which is similar to the well-known Nd-lasers), but provides a substantially longer storage time. The special advantage of Yb:S-FAP among potential Yb-lasers is related to its low level of ground state absorption loss at the laser wavelength. The Cr:ZnSe laser represents a new approach to directly generating tunable mid-infrared (2200-3000nm) radiation, on the basis of a material that emits with a high quantum yield.

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