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Martin Stalder

University of Central Florida

Bruce H. T. Chai

University of Central Florida

Michael Bass

University of Central Florida

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Flashlamp pumped Cr:LiSrAlF₆ laser

Martin Stalder, Bruce H. T. Chai,^{a)} and Michael Bass^{b)}

Center for Research in Electro-Optics and Lasers, University of Central Florida, 12424 Research Parkway, Orlando, Florida 32826

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Tunable, flashlamp-pumped laser properties are described for the crystal Cr:LiSrAlF₆ (Cr:LiSAF) in both long pulse and Q-switched modes of operation. Slope efficiencies of 5%, overall efficiency of 3%, and a tuning range from 780 to 1010 nm are reported.

Cr³⁺-doped LiSrAlF₆ (Cr:LiSAF) was shown to be a potentially interesting tunable laser using a four-level, phonon-terminated, laser process by Payne *et al.*^{1,2} They reported absorption and emission spectroscopic properties of Cr:LiSAF crystals as well as laser-pumped lasing. In this work tunable, flashlamp-pumped laser properties are described for Cr:LiSAF in both long pulse and Q-switched modes of operation. Slope efficiencies of 5%, overall efficiency of 3%, and a tuning range from 780 to 1010 nm are reported. The present work demonstrates that Cr:LiSAF is a practical, tunable laser which can be used in conventional laser systems. As a result many application of Cr:LiSAF are feasible.

Single crystals of Cr:LiSAF were grown by the Czochralski technique. LiSAF melts congruently at 790 °C with preferential evaporation of AlF₃ causing a continuous shift of composition during growth. Boules of Cr:LiSAF were successfully pulled up to 25 mm in diameter and 120 mm in length. Laser rods 6.35 mm in diameter and 100 mm long were cut from these boules. The melt was charged with up to 15% CrF₃. However, because there is also a slow decomposition of the CrF₃, the actual Cr concentration in the crystals is less than in the melt. All the Cr concentrations given in this work are those of the initial melt.

The as-grown LiSAF crystal is strongly faceted along the (0001) direction. This is primarily due to the distortion of the fluorine ion site by the large size of the Sr ion. This distortion also causes an unusual behavior of LiSAF in that the thermal expansion coefficient along the *c* axis (optical axis) is negative. This was noted in our growth experiments and subsequently confirmed by the direct measurement at Lawrence Livermore National Laboratory.³ This is the first fluoride crystal known to have such behavior.

Cr³⁺ in LiSAF substitutes for the Al³⁺ ion which is octahedrally coordinated with the fluorine ions, meaning that neighboring Cr³⁺ ions do not share the same fluorine. As a result, one expects very weak ion-ion interaction between the Cr³⁺ ions and possible impurity ions. This was confirmed by measurements which showed that the lifetime of the upper laser level ⁴T₂ depended only slightly on concentration, decreasing from 69 ± 3 μs in 2% doped material to 63 ± 3 μs in 15% doped material.

The laser rod was mounted in a close coupled, specular reflecting, elliptical pump chamber and excited with a 100-

mm-long, 4 mm bore xenon flashlamp. In this work the pump pulse duration was 120 μs and the laser was operated at 1 Hz. To prevent erosion of the laser crystal through interaction with de-ionized water in the cooling system a mixture of equal parts of water and ethylene glycol was needed as a coolant.

The laser rod end faces were polished flat and parallel and were not antireflection coated. The stable optical resonator was 31 cm long and consisted of a flat output mirror and a 20 m radius of curvature concave spherical 100% reflector. Though lasing with a flat 100% reflector was observed, higher output and more complete filling of the laser rod was achieved with the slightly curved reflector.

Figure 1 shows the long pulse output energy versus the total input energy to the flashlamp measured using a 4% Cr:LiSAF laser rod. This data was obtained with a 43% reflectivity output coupler which resulted in lasing centered at 845 nm in a band ≈ 10 nm wide. The output was polarized parallel to the crystal's *c* axis due to the higher emission cross section for light so polarized.¹ The measured slope efficiency of 5% and the overall efficiency of 3.1% measured at 86 J input energy are similar to the best results found with Alexandrite lasers.⁴

Lasing was first detected (i.e., the first relaxation oscillation spikes could be seen with a detector-oscilloscope combination) at an input energy called *E*_{th1} which was as little as 8 J using a 1.5% transmission output mirror. Another, higher threshold energy, *E*_{th2}, is found by extrapolating the linear region of the output versus input energy curve to zero output energy. Table I gives the threshold energies *E*_{th1} and *E*_{th2} and the slope efficiencies, *σ*, using output mirrors with different reflectivities, *R*. In Table I the effective reflectivity, *R'*, was obtained from *R* by correcting for the Fresnel reflection of the uncoated laser rod.⁵

In Fig. 2 the Findlay-Clay analysis⁵ was applied to determine losses in the laser resonator using the data in Table I. Using the threshold energy *E*_{th2}, 39% double pass losses have been found. These losses are the result of scattering, strain, inhomogeneous pumping, and absorption, as well as losses due to the decay of excited Cr ions which have a lifetime of 67 μs while pumping the rod with a 120 μs pump pulse. Transmission measurements have also been performed with a 850 nm (continuous wave) light source giving 29% double pass losses and 23% when correcting for the Fresnel reflections. When measuring the transmission, a large-area detector was used which means that scatter and strain losses leading to small angular deviation of the laser beam were not detected. Since the determination

^{a)}Also Department of Physics.

^{b)}Also Departments of Electrical Engineering and Physics.

FIG. 1. Output vs input energy for a 6.35×100 mm 4% Cr:LiSAF rod. A 43% reflectivity output mirror was used. The slope efficiency is 5% and the overall efficiency at 86 J input energy is 3.1%. Input energy is the energy stored in the power supply capacitors.

of loss using the Findlay–Clay technique is sensitive to scattering, strain, and lifetime effects, it is reasonable that it yields higher losses than the transmission measurement. Despite the high losses, excellent performance has been obtained. Improvements in material quality can be expected to yield even better results.

The slope of the curve in the Findlay–Clay plot in Fig. 2 is proportional to the gain of that part of the rod which is lasing.⁶ Figure 2 shows that the gain of the curve corresponding to the first laser spikes is higher than the gain determined from the linear extrapolated case. This difference is considered to be due to inhomogeneous pumping of the laser rod as confirmed by the near-field burn pattern on unexposed Polaroid film. Near threshold these patterns showed the region of lasing to be that part of the rod facing the flashlamp. For pumping with 86 J the data in Fig. 2 yield a small signal gain of 0.24 and 0.14 cm^{-1} for the strongly pumped region and the average of the laser rod, respectively.

The tuning range of Cr:LiSAF when flashlamp pumped with a fixed input energy was determined using a wavelength selector consisting of two intracavity 60° F2 Flint glass prisms. The angles between the prisms and the laser beam were close to the Brewster angle for the polar-

TABLE I. Threshold energies and slope efficiencies for different mirror reflectivities.

Output coupler reflectivity, R	Effective reflectivity R'	E_{th1} (J)	E_{th2} (J)	Slope efficiency σ (%)
0.985	0.99	8.0	15	0.43
0.83	0.88	10.8	18	2.9
0.69	0.77	12.5	22	3.9
0.50 ^a	0.61	17.5	35	5.0
0.43	0.55	18.5	32	5.0
0.00 ^b	0.028	71.0	...	...

^aThis reflectivity, for the ZnSe window, was estimated for a resonant reflector of index of refraction of 2.5 (see Ref. 6).

^bIn this case the output coupler was removed and the end surface of the laser rod acted as the output reflector. With an index of refraction of about 1.4 this corresponds to a mirror having a reflectivity of 2.8%.

FIG. 2. Findlay–Clay analysis for the 6.35×100 mm 4% Cr:LiSAF rod. (a) Analysis using E_{th1} corresponding to the energy at which the first relaxation oscillation spike was observed. (b) Analysis using E_{th2} corresponding to the energy obtained from the linear extrapolation of output vs input energy data at which the output energy is zero. The slopes of the two curves lead to gains of 0.24 and 0.14 cm^{-1} at 86 J of input energy.

ized beam. An uncoated, flat ZnSe window was chosen as the output coupler because the reflectivity was not very sensitive to wavelength over the range of interest. It was observed that different regions of the stressed rod lased at different wavelengths with a separation of up to 25 nm. Internal stresses resulted in a varying amount of wedge across the rod which, when combined with the prism tuner, resulted in this phenomenon. By inserting a 1.6-mm-diam aperture in the resonator, only one region could oscillate and it had a bandwidth ≤ 5 nm. The resulting tuning spectrum of flashlamp pumped Cr:LiSAF is given in Fig. 3. By pumping with an energy about four times the energy threshold given at 840 nm radiation from about 780 to 1010 nm were observed. The tuning range is limited by absorption into the upper laser level 4T_2 state at the shorter wavelengths and possible excited state absorption^{1,7} at the

FIG. 3. Tuning spectra for Cr:LiSAF. The laser crystal was pumped at energies of 2 $\times$ and 3 $\times$ the threshold for the first relaxation oscillation spikes at 840 nm for the resonator containing the two prisms. Measurements were carried out with a rear mirror centered at 800 nm (unfilled symbols) and 900 nm (filled symbols) and a flat uncoated ZnSe window as output coupler.

longer wavelengths. It is expected that with more homogeneous rods and a more selective tuner the same tuning curve and bandwidth can be obtained from the whole rod.

Initial *Q*-switched lasing experiments with Cr:LiSAF were performed with a rotating mirror. The resonator was lengthened to 80 cm and the 100% reflecting mirror was rotated at 250 Hz. Single *Q*-switched pulses of between 40 and 50 ns duration were generated. The experiment was terminated when 150 mJ output had been obtained because the damage thresholds of the material and intracavity optics were not known. Further the spatial distribution in the beam may have contained hot spots increasing the risk of damage to the system. The tuning range in this initial experiment with *Q*-switching was from 800 to 930 nm.

In summary, tunable long pulse and *Q*-switched laser operation has been demonstrated for a 4% Cr:LiSAF crystal using conventional flashlamp excitation. Despite high losses in the laser medium outstanding performance poten-

tials have been demonstrated. These include low-threshold, high slope efficiency, and wide tuning range. Improvements in material quality and proper matching of dopant concentration with the pump source and cavity can be expected to result in still better performance.

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