Gain properties of $LiSrAlF_6:Cr^{3+}$

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Small-signal gain properties of Cr^{3+}-doped $LiSrAlF_6$ are measured for the first time to our knowledge. Maximum small-signal gain values of 0.17 cm^{-1} are reported for π-polarized light. For σ polarization, the gain is significantly suppressed by excited-state absorption.

A new avenue of opportunity in the field of intense ultrashort laser pulse technology has opened as a result of the development of a new laser material: Cr^{3+}-doped $LiSrAlF_6$. The generation of ultraintense, ultrashort laser pulses by using chirped-pulse amplification (CPA) has so far been demonstrated in Nd:glass,[1] alexandrite,[2] and, recently, $Al_2O_3:Ti^{3+}$,[3,4] in which intense 100-fs laser pulses have been generated. Cr^{3+}-doped $LiSrAlF_6$ has been shown to have a very large fluorescence bandwidth of 220 nm, a relatively large gain cross section of 5×10^{-20} cm^2, and a fluorescence lifetime of 67 μs.[5,6] The latter permits pumping of this material by flash lamps, an important consideration for a compact low-cost laser system. A $LiSrAlF_6:Cr^{3+}$ laser has already demonstrated a high slope efficiency for single-flash-lamp pumping.[7] Moreover the Cr concentration in the host material can be varied across a broad range (from 0% to 100%), which permits the optimization of gain conditions for given optical pump configurations, and progress is being made in fabricating large boules of these materials with the use of a modified Czochralski crystal growth process.[8]

These properties make $LiSrAlF_6:Cr^{3+}$ an attractive solid-state material for the generation of intense ultrashort laser pulses with the CPA technique.[9] Moreover a mode-locked $Al_2O_3:Ti^{3+}$ laser can be used as a convenient short-pulse source for a $LiSrAlF_6:Cr^{3+}$ system based on CPA. The Kerr lens mode-locked $Al_2O_3:Ti^{3+}$ laser[10] produces stable and clean optical pulses of <100-fs duration and can be operated at 850 nm, the peak gain wavelength of $LiSrAlF_6:Cr^{3+}$.

The use of $LiSrAlF_6:Cr^{3+}$ for high-power pulse amplification requires an accurate knowledge of its gain properties. Whereas absorption and emission spectroscopic properties and laser action have been published,[5,7] no detailed gain measurements have yet been reported to our knowledge. For this reason we have begun to investigate the small-signal gain properties of $LiSrAlF_6:Cr^{3+}$. Our first results, presented in this Letter, show that the high small-signal gain of this material for π-polarized light is encouraging for the use of this new laser material for high-intensity pulse generation. For σ-polarized light, however, the small-signal gain is strongly suppressed and indicates the presence of excited-state absorption (ESA).

The small-signal gain is measured by probing cylindrical rods with an $Al_2O_3:Ti^{3+}$ laser tunable in wavelength from 780 to 900 nm (Fig. 1). The rods under investigation are pumped at 6 Hz in a dual-flash-lamp laser head with an effective pump length of 56 mm and with energies of as much as 50 J. The rods investigated thus far had a diameter of 4 mm and a length of 65 mm. The flash-lamp pump duration is 130 μs, short enough to pump the sample efficiently and long enough to ensure the flash-lamp lifetime. The small-signal gain is determined by transmitting a small fraction of the $Al_2O_3:Ti^{3+}$ beam through the rod and measuring the amplification during flash-lamp operation. Special care is taken to eliminate the contribution of fluorescence from the $LiSrAlF_6:Cr^{3+}$ crystal to the transmitted signal by placing the detector far from the laser head and using two iris pinholes. The background fluorescence signal is measured in the absence of the probe beam and is then subtracted from the amplified signal. Within the accuracy of our experiment, we do not observe any pump-induced change of the transmitted beam, which may be due to optical bending, thermal focusing, or birefringence.

The dependence of the small-signal gain on the electrical-input energy density is shown in Fig. 2(a). The molecular Cr^{3+} concentration of this rod is 1.4% (CrF_3 replacing AlF_3). The incoming light is π polarized (parallel to the c axis), and the probe wavelength is tuned in the range from 800 to 900 nm through the maximum of the gain curve. For the maximum available pump energy of 48.6 J (69 $J\,cm^{-3}$), we measure a maximum small-signal gain of 0.125 cm^{-1}. The saturation in the curves in Fig. 2(a) is probably due to saturation of the flash-lamp pump energy. The wavelength dependence

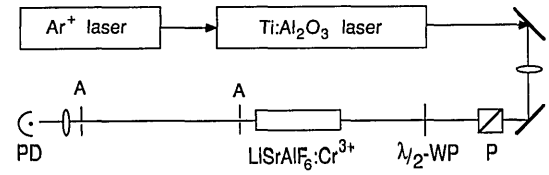

Fig. 1. Experimental setup. P, polarizer; WP, wave plate; A's, apertures; PD, photodiode.

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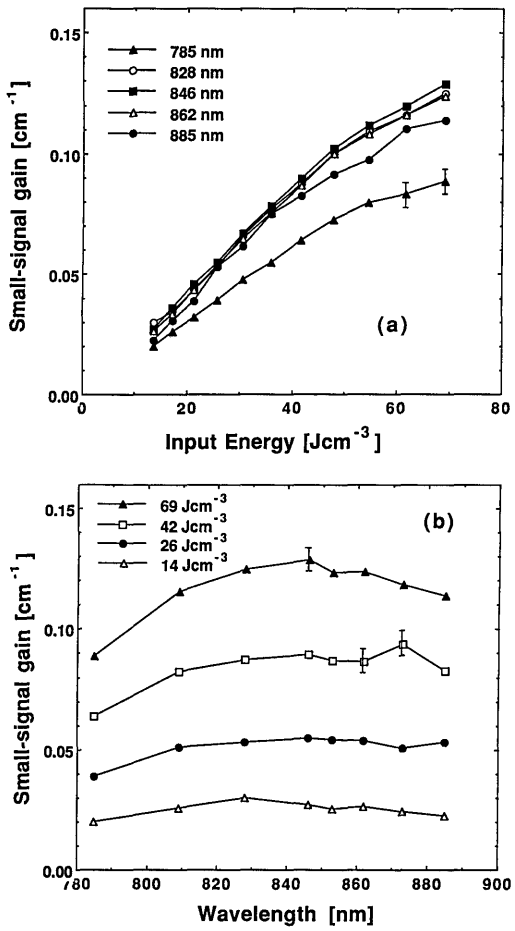

Fig. 2. Small-signal gain (a) versus electrical input energy and (b) versus wavelength in $LiSrAlF_6:Cr^{3+}$ for π-polarized light.

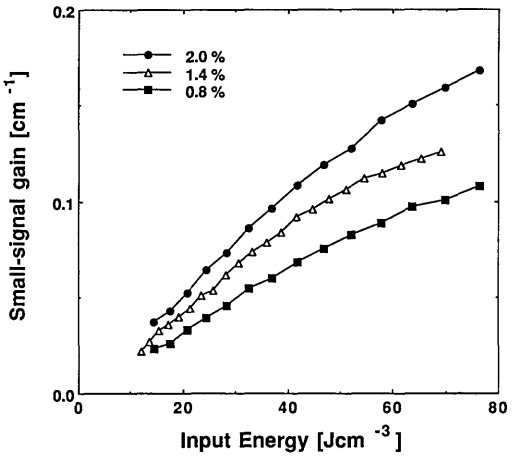

Fig. 3. Small-signal gain at 850 nm versus electrical input energy in $LiSrAlF_6:Cr^{3+}$ for π-polarized light for three different Cr^{3+} concentrations.

is better illustrated in Fig. 2(b), which shows the small-signal gain values plotted for several levels of electrical input energies. The maximum gain is found to be ~850 nm, which corresponds, within the accuracy of the experiment, to the maximum of the emission cross section of 845 nm.[5] The steeper decay of the gain toward lower wavelengths is due to the increasing edge of the broad absorption band centered at 640 nm. The latter is due to the $^4A_2$–$^4T_2$ transition, the transition from the ground state to the upper laser level.

We also measured the small-signal gain of $LiSrAlF_6:Cr^{3+}$ for 0.8 and 2% Cr^{3+} concentrations. The result obtained with all three rods is shown in Fig. 3. The incoming light is again π polarized, and the probe wavelength is tuned to 850 nm, the maximum of the gain curve. For the 2% sample, we measure a small-signal gain value of 0.17 cm^{-1} at a pump energy of 78 Jcm^{-3}. The measured single-pass loss at 850 nm for the three samples was 7.1%, 15.3%, and 13.7% for the 0.8%, 1.4%, and 2% samples, respectively. These numbers also include the reflection losses inasmuch as the rods were not antireflection coated.

A strong dependence of the small-signal gain on polarization was observed. The measured small-signal gain for the σ polarization is shown in Figs. 4(a) and 4(b). In measuring the small-signal gain values for this polarization, a second polarizer was inserted after the amplifier to block detection of any light coupled into the π polarization. The small-signal gain value for σ polarization is an order of magnitude smaller at 820 nm than for the π polarization and decreases linearly with increasing wavelength. Moreover, the gain becomes negative for wavelengths >845 nm. This result is initially

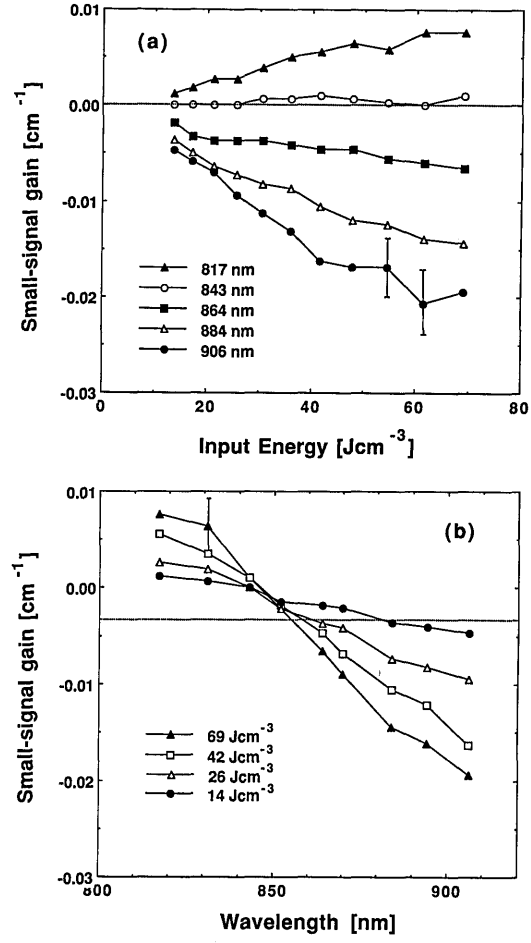

Fig. 4. Small-signal gain (a) versus electrical input energy and (b) versus wavelength in $LiSrAlF_6:Cr^{3+}$ for σ-polarized light.

surprising because the emission cross section for σ polarization is only three times smaller than for π-polarized light and has the same wavelength dependence.[5] From the results above we conclude that a serious pump-induced loss mechanism is present in $LiSrAlF_6:Cr^{3+}$.

We believe that this loss mechanism is due to ESA.[5] ESA in $LiSrAlF_6:Cr^{3+}$ has not been investigated to our knowledge, but a detailed study of ESA in $LiCaAlF_6:Cr^{3+}$, a laser material that has a similar structure to that of $LiSrAlF_6:Cr^{3+}$, shows a broad ESA band centered at 997 nm.[11] This band in $LiCaAlF_6:Cr^{3+}$ is far enough from the fluorescence maximum at 760 nm, and the ESA cross section is small enough, to have no serious effect on the gain of this material. The emission peak for $LiSrAlF_6:Cr^{3+}$ occurs at 845 nm. This difference in the emission wavelength for the two materials is related to the extent of lattice relaxation that occurs in the $^4T_2$ excited state. As a result, the ESA peak for $LiSrAlF_6:Cr^{3+}$ is probably shifted toward lower wavelengths (compared with the ESA peak of $LiCaAlF_6:Cr^{3+}$), thereby resulting in a greater ESA loss.[5] This is supported by the fact that the pump-dependent loss found in our experiment increases as the wavelength increases from 800 to 900 nm. The gain crosses the zero line at ~845 nm [see Fig. 4(b)]. At this wavelength the ESA cross section σ_{ESA} therefore equals the emission cross section. From the knowledge of the latter at this wavelength,[5] the ESA cross section is $\sigma_{ESA} = \sigma_{21} = 1.6 \times 10^{-20}$ cm^2.

Although ESA suppresses the gain of $LiSrAlF_6:Cr^{3+}$ for σ-polarized light, there is no evidence of strong ESA for π-polarized light. This would result in a steep decrease of the gain values toward higher wavelengths. Such behavior clearly is not observed. However, ESA may lead to some reduction in the small-signal gain for π polarization. Theoretically, predictions[11] suggest that the ESA cross section is unpolarized for $LiCaAlF_6:Cr^{3+}$. Because $LiSrAlF_6:Cr^{3+}$ is a similar crystal, we expect that ESA in $LiSrAlF_6:Cr^{3+}$ is independent of polarization.[12] Under this assumption, and using the value of σ_{ESA} evaluated above, the emission cross section σ_{21} of 4.8×10^{-20} cm^2 for π polarization at 845 nm (Ref. 5) reduces by ~33% to an effective gain cross section σ_{eff} $(=\sigma_{21} - \sigma_{ESA})$ of 3.2×10^{-20} cm^2. This result is in good agreement with a recent publication[5] in which the intrinsic slope efficiency of a $LiSrAlF_6:Cr^{3+}$ laser was found to be ~30% lower than the quantum-defect efficiency.

In summary, we have reported what is to our knowledge the first small-signal gain values of $LiSrAlF_6:Cr^{3+}$. Our results demonstrate for π polarization a maximum small-signal gain of 0.17 cm^{-1} at an electrical input energy of 78 J cm^{-3} for a Cr concentration of 2%. Using this value and the effective gain cross section evaluated above of 3.2×10^{-20} cm^2, we obtain an inversion density $n = g/\sigma_{eff}$ of 5.3×10^{18} cm^{-3} (g is the measured small-signal gain). This corresponds to an inversion of 3.5% of the Cr ions and to a stored energy density of 1.24 J cm^{-3}. Assuming a homogeneous pumping, we calculate with these numbers an overall pump efficiency (stored energy versus electrical input) of 1.6%. The high gain and the extremely broad emission band render $LiSrAlF_6:Cr^{3+}$ ideal for the amplification of ultrashort laser pulses. We have already demonstrated a femtosecond chirped-pulse regenerative amplifier using $LiSrAlF_6:Cr^{3+}$.[13] In addition, we have shown that the gain of $LiSrAlF_6:Cr^{3+}$ is reduced by ESA, giving an estimate of 1.6×10^{-20} cm^2 for the unpolarized ESA cross section at a wavelength of 845 nm. This results in a 30% reduction of the gain for π-polarized light. For σ-polarized light, the gain is completely suppressed by ESA. The latter result, however, does not affect the use of $LiSrAlF_6:Cr^{3+}$ in an amplifier system but has some consequences for the design of multipass amplifying systems.

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