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Performances of Cr:LiSrAlF₆ and Cr:LiSrGaF₆ for continuous-wave diode-pumped Q-switched operation

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We have developed a Cr:LiSrGaF₆ laser that produces, in Q-switched operation, four times as much energy (12 μJ at 10 kHz) as a Cr:LiSrAlF₆ laser under the same conditions of pumping (four red diodes emitting 400 mW). By using a theoretical model for small-signal gain calculations, we show that these better performances are mainly attributable to the fact that thermal quenching of fluorescence is lower in Cr:LiSrGaF₆ than in Cr:LiSrAlF₆. © 1997 Optical Society of America

Of the colquirite family of crystals, Cr:LiSrAlF₆ (Cr:LiSAF) and Cr:LiSrGaF₆ (Cr:LiSGaF) are the most used. They have nearly the same absorption band in the red and the same emission band between 800 and 1000 nm. They have a comparable fluorescence lifetime (67 μs for Cr:LiSAF and 88 μs for Cr:LiSGaF), which makes them convenient for the development of a Q-switched cw diode-pumped laser operating at a high repetition rate (greater than 10 kHz). They also have similar emission cross sections (4.8×10^{-20} cm² for Cr:LiSAF, and 3.3×10^{-20} cm² for Cr:LiSGaF). The same intrinsic slope efficiencies (approximately 50%) have been demonstrated in cw operation.¹ However, some differences are well established: Cr:LiSGaF has lower scattering losses¹ and better power-handling capability, particularly for cw operation.² The latter property is attributed to a lower thermal expansion coefficient and to a lower anisotropy of thermal expansion.^{2,3} In this Letter we propose to highlight a comparison between these two crystals in the Q-switched regime. We show that, in fact, thermal quenching of fluorescence is the dominant mechanism and that this effect explains the different performances of the two crystals.

The first goal is to find which crystal is best suited to the development of a tunable nanosecond source pumped in cw by red laser diodes. The second goal is to find the physical origins of the difference between the performances of the two lasers. To accomplish these goals we present experimental measurements and a theoretical modeling of the small-signal gain in Cr:LiSGaF.

Optimizing the energy per pulse in Q-switched operation requires that we maximize the pump power to obtain a large excited-state population. It also requires that we maximize the small-signal gain to have a good extraction of the stored energy. For these reasons, we used high-brightness pump diodes providing 400 mW at 670 nm with an emitting area of $1 \mu\text{m} \times 100 \mu\text{m}$. We also used a longitudinal pumping configuration in which two diodes were coupled by polarization on each side of the crystal (Fig. 1). Thus the pump power available reached 1.6 W, which is the maximum possible with this type of diode in this pumping scheme. The laser cavity consisted of three mirrors defined by the plane face of the crys-

tal (M₁), a highly reflective concave mirror with a radius of curvature of 100 mm (M₂), and a plane output coupler (M₃). The Cr:LiSGaF and the Cr:LiSAF crystals have the same geometry (plano-Brewster) and the same doping level (3%). The central path length of the crystals was 3 mm. The pumping optics were similar to those previously described.⁴ We imaged the emitting zone inside the crystal by using the following system: The beam was first collimated by a high-numerical-aperture objective O₁ (focal length, 15 mm); then afocal systems reshaped the beam, and a second objective O₂ (focal length, 100 mm) focused it inside the crystal. On the plane face of the crystal we used two afocal systems to make the pump beam more circular than the stripe of the diode. The pump spot size is $80 \mu\text{m} \times 30 \mu\text{m}$ at the focus point, and it remained below $200 \mu\text{m} \times 40 \mu\text{m}$ over a length of 2 mm on the propagation axis. This length corresponded to the confocal parameter in the diffraction-limited direction, perpendicular to the junction of the diode. On the Brewster face, only one afocal was necessary because we took advantage of the 1.4 magnification imposed by the air-crystal diopter.⁴ The magnification of the different afocal systems was chosen to maximize the small-signal gain for each crystal.⁴ To achieve Q-switched operation, we added an acousto-optic modulator inside the cavity. The output coupling of the mirror M₃ was chosen to optimize the output energy. For Cr:LiSAF, we used a coupling of 2%; for Cr:LiSGaF, a coupling of 5%. We also used an output coupler with a transmission of 2% for Cr:LiSGaF to compare the performances of the two crystals in exactly the same cavity.

Figure 2 shows the energy per pulse at a repetition rate of 10 kHz as a function of the pump power (the

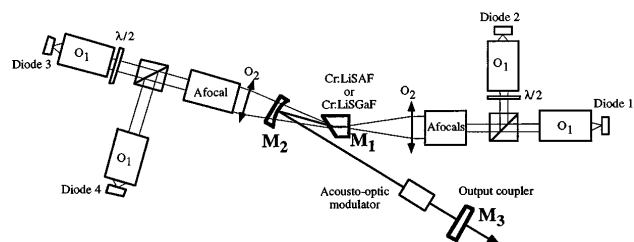


Fig. 1. Experimental setup.

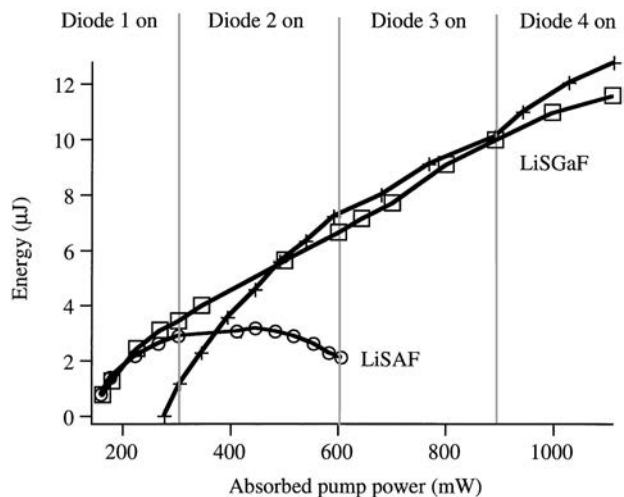


Fig. 2. Energy per pulse for Cr:LiSAF and Cr:LiSGaF at a 10-kHz repetition rate. The circles and the squares correspond to an output coupling of 2% for Cr:LiSAF and Cr:LiSGaF, respectively. The plus signs correspond to an output coupling 5% for Cr:LiSGaF.

diodes were successively switched on). We observed a roll-off in the Cr:LiSAF output energy with greater than 400 mW of absorbed pump power, whereas no decrease occurred for the Cr:LiSGaF laser. At the maximum, the energy reached 12 μJ for Cr:LiSGaF for an absorbed pump power of 1.1 W (pulse duration, 250 ns) and only 3 μJ for Cr:LiSAF for a pump power of 400 mW (pulse duration, 400 ns).

To gain an understanding of the better performances obtained with the Cr:LiSGaF laser, we measured the small-signal gain in the two crystals by using the method described in Ref. 5. Figure 3 shows that, as expected, the small-signal gain had the same behavior as the energy per pulse as a function of pump power: It decreased for Cr:LiSAF but not for Cr:LiSGaF. Moreover, it was higher for Cr:LiSGaF when the pump power was higher than 350 mW.

We have shown^{5,6} that the gain roll-off in Cr:LiSAF is caused by upconversion⁷ and by thermal quenching of fluorescence.⁸ To accomplish this, we performed theoretical calculations of the small-signal gain and compared it with the gain measured experimentally by pumping the crystal with a krypton laser. We used this pump laser instead of diodes because it was easier to perform the calculations with a circular Gaussian pump profile, which is similar to the krypton beam profile.

We used the same method in an effort to understand why the gain was higher with Cr:LiSGaF. To do so, we replaced the four diodes with a krypton laser and pumped the crystal with a 100-mm focal-length lens. To calculate the gain theoretically, different physical data of Cr:LiSGaF were necessary. Unfortunately, for this crystal the thermal conductivity and the thermal quenching of fluorescence are unknown. Moreover, the upconversion parameter was measured to be between 10^{-16} and $6 \times 10^{-16} \text{ cm}^3 \text{ s}^{-1}$ for a 3% doped Cr:LiSGaF crystal.⁷ To find the upconversion parameter more precisely, we operated the laser in quasi-cw (repetition rate, 50 Hz, duty cycle, 1/6). In this case,

one could neglect the thermal heating of the crystal and hence neglect the decrease of lifetime induced by the temperature increase. We fitted the Cr:LiSGaF experimental gain in this regime with our model (Fig. 4) by adjusting the upconversion parameter. We found a value of $6.5 \times 10^{-16} \text{ cm}^3 \text{ s}^{-1}$, close to the value obtained in Ref. 7. This value is comparable with the Cr:LiSAF value for a doping of 3%.⁹

Next we measured the lifetime of Cr:LiSGaF as a function of temperature. $\tau(T)$ can be given by the formula⁸

$$\tau(T)^{-1} = \tau_R^{-1} + \left[\tau_{NR}^0 \exp\left(\frac{\Delta E}{kT}\right) \right]^{-1}, \quad (1)$$

where τ_R is the radiative lifetime; T , the temperature (K); k , the Boltzmann constant; ΔE , the activation energy; and τ_{NR}^0 , a parameter.⁸ From our measurement of the lifetime, we found the following values for the thermal quenching parameters ΔE and τ_{NR}^0 in Cr:LiSGaF: $\tau_{NR}^0 = 6.9 \times 10^{-14} \text{ s}$ and $\Delta E = 5155 \text{ cm}^{-1}$. For Cr:LiSAF crystals, these values are $\tau_{NR}^0 = 2.4 \times 10^{-14} \text{ s}$ and $\Delta E = 5125 \text{ cm}^{-1}$.⁸ To compare thermal quenching in the two crystals, we plotted

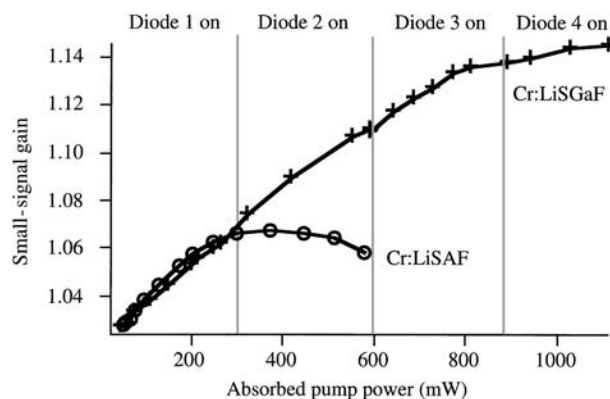


Fig. 3. Small-signal gain as a function of absorbed pump power.

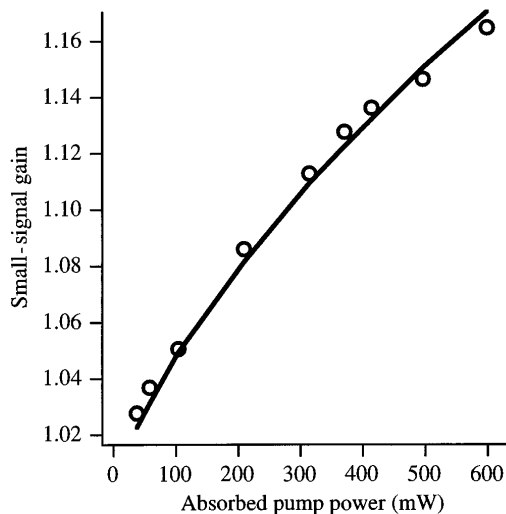


Fig. 4. Small-signal gain obtained by pumping Cr:LiSGaF with a krypton laser in quasi-cw. The circles correspond to experimental measurements and the curve to the theoretical calculations.

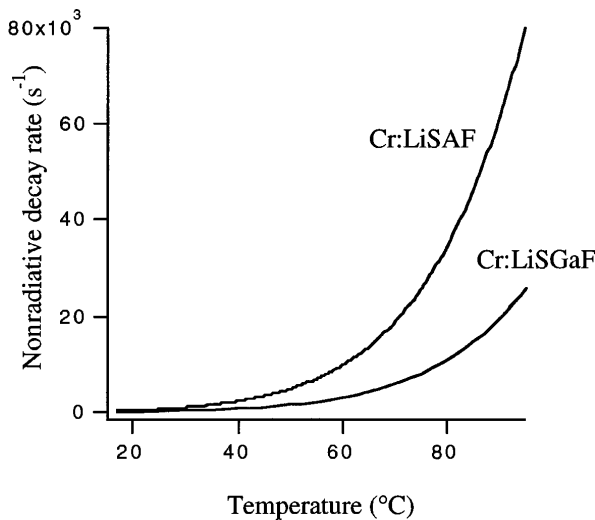


Fig. 5. Nonradiative decay rate D_{NR} as a function of temperature for Cr:LiSAF and Cr:LiSGaF.

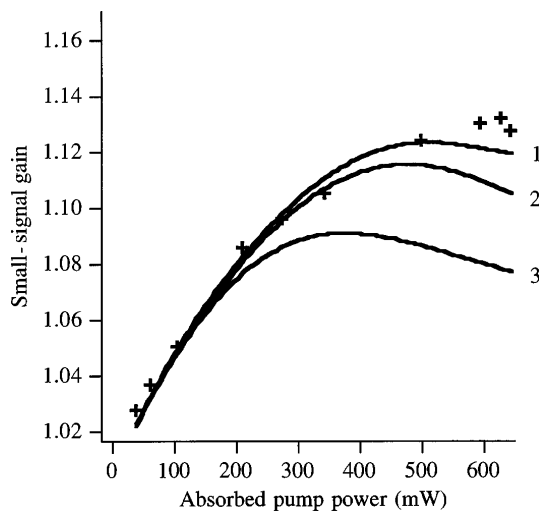


Fig. 6. Small-signal gain obtained by pumping Cr:LiSGaF with a krypton laser in cw. The plus signs denote experimental measurements. Curves 1–3 indicate the theoretical gain, as mentioned in the text.

the nonradiative decay rate D_{NR} as a function of temperature (Fig. 5). If we follow Eq. (1), D_{NR} is given by the formula⁸

$$D_{NR} = \left[\tau_{NR}^0 \exp\left(\frac{\Delta E}{kT}\right) \right]^{-1}. \quad (2)$$

Figure 5 shows that the quenching is much more important in the case of Cr:LiSAF.

Next we measured the Cr:LiSGaF small-signal gain in cw. We observed a roll-off at approximately 600 mW (Fig. 6). This effect did not occur in the case of diode pumping even though the absorbed pump power was higher (1.1 W). This result was due to the pumping configuration: In the case of the diodes, the pump power was better distributed because the crystal was pumped on both sides.

To model the small-signal gain in Cr:LiSGaF, we used our measured quenching parameters ΔE and τ_{NR}^0 , and we adjusted the thermal conductivity to fit the experimental points. We found curve 1 of Fig. 6 for a thermal conductivity of $K = 3.5 \text{ W m}^{-1} \text{ }^\circ\text{C}^{-1}$. This value is comparable with the thermal conductivity of Cr:LiSAF given at $K = 3 \text{ W m}^{-1} \text{ }^\circ\text{C}^{-1}$.⁹ In fact, all the Cr:LiSGaF parameters studied in this Letter are more or less similar to the Cr:LiSAF parameters, except for the quenching parameter τ_{NR}^0 , which induced a much higher nonradiative decay rate in the case of Cr:LiSAF.

To prove that thermal quenching is the origin of the different small-signal gain in Cr:LiSAF and in Cr:LiSGaF, we modified the parameters of the gain modeling for Cr:LiSGaF as follows: First, we set the thermal conductivity to $K = 3 \text{ W m}^{-1} \text{ }^\circ\text{C}^{-1}$, and we obtained curve 2 of Fig. 6. This curve is relatively near curve 1, showing that the better thermal conductivity of Cr:LiSGaF cannot explain the difference between the two crystals in terms of gain as shown in Fig. 3. Second, we used $\tau_{NR}^0 = 2.4 \times 10^{-14} \text{ s}$ instead of $\tau_{NR}^0 = 6.9 \times 10^{-14} \text{ s}$, and we obtained curve 3. This curve is very different from curve 1: The gain is lower, and the roll-off occurs at a lower pump power (350 mW instead of 500 mW).

In conclusion, we have obtained four times as much energy per pulse with the Cr:LiSGaF laser as obtained with the Cr:LiSAF laser. The main origin of the difference is the thermal quenching of fluorescence, which is much lower in Cr:LiSGaF. This effect induced a roll-off of the gain and a roll-off of the energy in Cr:LiSAF, which did not occur in Cr:LiSGaF for the pump power available (1.1 W). This study proves that the better behavior of Cr:LiSGaF for high pump power cannot be attributed only to a lower thermal expansion coefficient and to a lower anisotropy of thermal expansion, as was done elsewhere.^{2,3}

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