Crystal growth, spectroscopy and high-power diode-pumped CW laser operation of a new laser medium: Nd:Lu:YLF

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High optical quality $YLiF_4$ crystals (YLF) are well-known host materials for several rare-earth active-center ions, including neodymium (Nd), [1] due to the good optical, thermal and mechanical characteristics, and low photo-elastic coefficients. Moreover, it is birefringent, therefore minimizing the loss of the optical polarization due to induced stress or nonlinearities. The concentration of Nd^{3+} ions that substitute the Y^{3+} ions in Nd:YLF is of the order of 1%. Recently, a new laser material similar to Nd:YLF has been demonstrated: Nd:LuLF ($LuLiF_4$). This material presented same spectral and physical properties as Nd:YLF, with the advantages of easier growth, and broader Nd emission cross section, but with the disadvantage of lower Nd concentration, typically around 0.6%. [2] However, broadening the spectral gain width is a desirable enhancement for crystalline Nd laser media in order to obtain ultrashort pulse generation, especially when using passive mode locking techniques. [3] In this case, the minimum pulse width, obtained by balancing the nonlinear phase effects with net group-velocity dispersion, is inversely proportional to the square of the gain spectral linewidth. [4]

Lutetium is also a well-known size-compensating codopant for Nd:YAG ($Y_3Al_5O_{12}$), allowing the incorporation of greater amounts of Nd into YAG without significant degradation in optical quality, and broadening the emission lines by around 10 to 35%. [5] In this work we report the development of a new laser material, the Nd:Lu:YLF_4 (Nd:LuYLF), that presented a higher Nd concentration than in Nd:LuLF, same physical and spectroscopic parameters, and spectral broadening of the emission linewidth around 25%, compared with Nd:YLF.

Crystal growth and sample preparation

The binary rare earth and yttrium fluorides were prepared from pure oxide powders (99.99%, Aldrich and/or Johnson Matthey) by fluorination in a stream of argon gas (99.995%, White Martins) and HF gas (99.99%, Matheson Products) at 850 oC. The YLF and LuLF were synthesized, using the same procedure, in compositions of 49,5 mol% YF_3 : 50,5 mol% LiF, and 50 mol% LuF_3 : 50 mol% LiF, respectively. They were purified by zone refining process in a single pass. The stoichiometric part of the bars were used in the growth experiments. The single crystals were grown by Czochralski technique, both the zone refined YLF or LuLF plus the NdF_3 were melted in a platinum crucible under an argon atmosphere, after a heating treatment under vacuum. The crystal pulling rate was 1 mm h^{-1} and rotation rate was 25 rpm. During the process, the crystal diameter was controlled visually. For the growth of the LuYLF crystal, the melt composition was: 48,85 mol% YLF, 48.85 mol% LuLF. A concentration of 2.3 mol% NdF_3 was utilized for the growth of both crystals. The growing direction was parallel with the [100] crystallographic axis. Finally, the crystal suffered thermal treatment, under argon atmosphere, to eliminate internal stress. More details will be presented elsewhere. [6] The Nd concentration in the crystals, determined by optical absorption measurements, were 1,3 mol% for the Nd:YLF and 0,9 mol% for the Nd:LuYLF. The concentration of Lu was around 40 mol%, as determined by density measurements.

The optimum active medium length, for the longitudinally pumped Nd laser at $\lambda_P = 792$ nm, [7] is of the order of 1 cm, for Nd concentrations around of 1 mol%. Thus, the samples were extracted along the growth direction with dimensions of, approximately, (0.2 x 0.6 x 1) cm for Nd:YLF and (0.2 x 0.4 x 1) cm for Nd:LuYLF. The optical faces were prepared at Brewster angle, in the π polarization, and polished to a flatness of $\lambda/4$. The transmission of both samples at $\lambda = 1.05\mu m$ were better than 0.995(5).

Spectroscopic characterization

The infrared pumping source was a GaAlAs diode laser, model SDL-2382-P1, with 4 W CW at 792 nm. The diode laser beam at the sample position had a rectangular profile with dimensions of 60 μm x 300 μm.

The Nd polarized emission was analyzed using a 1m Spex monocromador and detected with a photomultiplier detector. The lifetime measurements were performed exciting the samples using a dye laser at 792 nm, pumped by a pulsed Nitrogen laser (10 ns). The emission was detected by an S-20 photomultiplier, and analyzed using a Box-Car Averager (PAR 4402).

The measured $^4F_{3/2}$ lifetime was 481μs for Nd:LuYLF and 476 μs for Nd:YLF. The emission and absorption spectra of Nd:LuYLF showed close similarity to those of Nd:YLF. The emission cross-section for the $^4F_{3/2} \rightarrow ^4I_{11/2}$ transition, for both polarizations (π and σ), were obtained through the emission spectrum and the measured lifetime, using the method of McCumber. [8] The results are presented in the Figure 1 for both polarizations. The determined emission cross-section peak, for the π polarization, was 2.7 x 10^{-19} cm^2, at 1.046 μm; for the σ polarization, it was 2.2 x 10^{-19} cm^2, at 1.052 μm. A detailed measurement of the fluorescent emission spectrum around 1047nm showed a broadening of 25% compared with the Nd:YLF emission spectrum, as shown in Figure 2. The Nd:LuYLF crystal presented a larger splitting of the laser manifolds due to the crystalline field, compared to Nd:YLF, and consequently a broadening of lines. It also indicates a more favorable thermal occupation of the upper and lower levels.

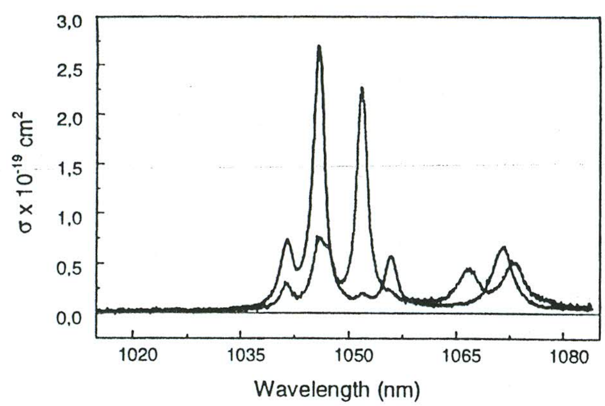

Figure 1: Emission cross-section of Nd:LuYLF, for the π (continuous line) and σ (dashed line) polarizations.

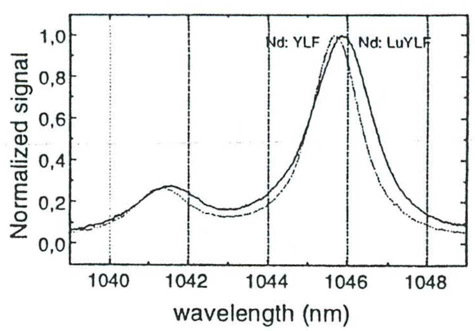

Figure 2: Comparison between Nd:YLF and Nd:LuYLF emission cross-sections for the π polarization.

Diode-pumped laser performance

The laser was pumped by a fiber lensed 20 W diode array emitting at 792 nm. The beam, with emitting dimensions of w_x = 1 cm parallel to the bar and $w_y \approx$ 0.2 mm perpendicular to the bar, was reconfigured into two columns of twelve beams each using a two mirror beam shaper [9]. The reconfigured beam had dimensions and quality factors of w_x = 200 μm, M_x^2 = 120 and w_y = 120 μm, M_y^2 = 56 at the focus where the crystal is placed. The laser resonator consisted of a 1-meter radius concave mirror, high reflector for 1.05 μm, with 94 % transmission for the pump beam, and flat output couplers with varied transmission at the laser wavelength. The total cavity length was 5 cm. The complete experimental setup is shown in Figure 3.

With the diode emitting 20 W of output power we had a total peak pump power incident on the crystal of 14 W due to losses in the beam shaper, lenses and the back mirror of the resonator. The duty cycle was only 10% due to a non-optimized crystal heatsink.

Measurements of the output power as a function of the output reflectivity were performed, as shown in figure 4. By fitting the following expression from the laser oscillator model: [10] $P_{OUT} = K.(1-R).[G/(L-\ln(R))-1]$, it was determined the equivalent non-saturated gain, G, the cavity losses, L, and $K = AI_s/2$, where A is the beam area at the active medium, and I_s the emission saturation intensity. The determined parameters are also plotted in the figure (inset). The observed lower laser output power of Nd:LuYLF is attributed to the lower Nd concentration in this crystal.

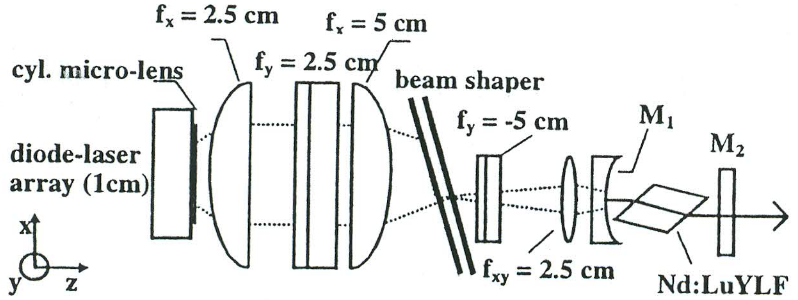

Figure 3: Setup of the Tm:Ho:YLF laser utilizing a two mirror beam shaper in the pump arrangement.

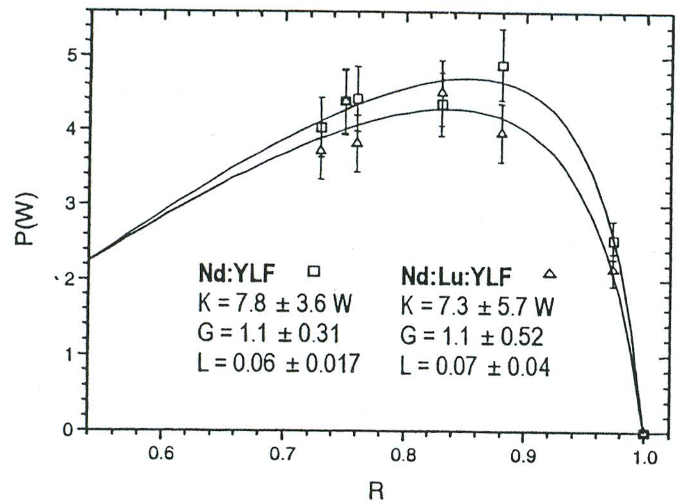

Figure 5: Output power as a function of the output reflectivity for the developed Nd:YLF and Nd:LuYLF lasers.

The only observed difference between Nd:YLF and Nd:LuYLF was the emission linewidth. No other significant differences were found between the spectroscopic and CW laser parameters of both crystals. Thus, Nd:Lu:YLF can be used for mode locking purposes with advantages over Nd:YLF. A complete short-pulse operation of this system, as well as characterization for different Lu concentrations, is currently under investigation.

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