LASERS AND OPTICAL INSTRUMENTATION BROADENING OF THE Nd EMISSION LINEWIDTH IN A NEW LASER HOST, LuYLF

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In the study of new crystalline host materials for Nd ions, aiming the development of Nd lasers with innovative properties, a new material was grown and characterized, the Nd:Lu:YLiF₄. The Lu and Y concentrations were varied from 0 to 100%, in a complementary way, and the Nd starting concentration was fixed in 2%. It was verified that the Nd ${}^{4}F_{3/2} \rightarrow {}^{4}I_{11/2}$ polarized emission has shown a linewidth broadening that depends on the Lu concentration. The ${}^{4}F_{3/2}$ luminescent lifetime and the stimulated emission cross-section were obtained for each sample, presenting values close to those from the well-known Nd:YLF crystal. With this results it was possible to determine the best concentration of Lu in order to have the maximum broadening and Nd concentration.

Introduction

Solid-state lasers have several advantages over gas or liquid-based lasers, first regarding the active center concentration, but also due to the intrinsic toughness of solid-state materials. When pumped by diode-lasers, an all solid-state compact laser arrangement can be obtained, with high efficiency and optical quality ^[1]. The most used solid-state laser medium is the Nd:YAG (neodymium-doped Y₃Al₅O₁₂), due to the combination of good optical, thermal and mechanical characteristics. However, there are several crystal hosts where the Nd laser action can be obtained, some as efficient as Nd:YAG (like Nd:YLF for instance). These lasers support many different kinds of operation. The continuous laser action (CW) is possible due to combination of the efficient pumping capability, reasonable values of the upper laser level lifetime and emission cross-section, and four-level laser cycle. The large energy storage capacity allows giant pulse operation, and the emission bandwidth allows picosecond-range ultrashort pulse generation and amplification.

Since high-power CW diode-lasers are now commercially available and can be efficiently used as Nd-ion pump sources in the 800 nm region, spectroscopic studies of Nd-doped insulator materials are renewed ^[2]. The Nd: YLiF₄ (Nd: YLF), currently used in a large variety of applications ^[3], presents a low laser threshold and high average output power. Despite it is well known as an efficient crystalline Nd laser for ultrashort pulse generation, the ~1.3 nm FWHM emission linewidth limits the pulsewidth to 1 ps. Recently, a new fluoride laser material has been demonstrated: Nd:LuLiF₄ (Nd:LuLF) ^[4]. This material presents almost the same spectral and physical properties as Nd:YLF laser crystals, and one significant difference: the broader spectral width. It is easier to grow, but can incorporate smaller Nd ion concentration (around 0.6% at maximum, whereas in Nd:YLF it is typically 1.5%), due to the lower segregation coefficient compared with YLF. Another inconvenient is the higher costs of lutetium compounds.

We have studied the codoping of lutetium in Nd:YLF crystals in order to obtain significant line broadening and enhanced Nd concentration, also aiming to lower the production costs compared with those of Nd:LuLF. We verified that, by using 50% of lutetium and 50% of yttrium, the obtained laser crystal presented a higher Nd concentration than in Nd:LuLF, the same physical and spectroscopic parameters (with a slightly higher peak emission cross-section), and a significant spectral broadening of the emission linewidth (25%), compared with Nd:YLF. We named this laser material as Nd:LuYLF.

Experimental Methods

The binary rare earth and yttrium fluorides were prepared from pure oxide powders (99.99%) by fluorination in a stream of argon gas and HF gas at 850° C. The same synthesis procedure was used for YLF and LuLF starting materials. For YLiF₄, it was used a composition of 49,5 mol% of YF₃ and 50,5 mol% of LiF; for LuLF, 50 mol% of LuF₃ and 50 mol% of LiF. They were purified by a single pass zone-refining process. The single crystals were grown by Czochralski technique, under argon gas atmosphere. The crystal pulling rate was 1 mm/h and rotation rate was 25 rpm. For all crystals, the nominal starting concentrations of Y, Lu and Nd are summarized in Table 1. The final concentration of Lu was around 50 mol% in sample 5 (Nd:LuYLF). The growing direction was parallel with the [100] crystallographic axis. The final Nd concentrations were determined by different measurements, and are around 1,3 mol% for Nd:YLF, 0,9 mol% for Nd:LuYLF, and less than 0.6 mol% for Nd:LuLF.

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Sample	Name	Y (mol%)	Lu (mol%)	Nd (mol%)
1	YLF	97.70	0	2.30
2		88.20	9.10	2.70
3		67.64	30.00	2.36
4		50.00	47.25	2.75
5	LuYLF	48.85	48.85	2.30
6	LuLF	0	97.70	2.30

Table 1. Proportional starting concentrations of Y, Lu and Nd in the grown crystals.

Lifetime measurements were performed at room temperature, by exciting the spectroscopic samples with a dye laser, pumped by a pulsed nitrogen laser (10 ns). The polarized Nd emission was analyzed using a 1-m Spex spectrometer, an S-1 photo-multiplier and a boxcar averager. The measured ${}^{4}F_{3/2}$ lifetimes were 481 ms and 476 ms for Nd:LuYLF and Nd:YLF, respectively.

The emission and absorption spectra of both Nd:LuYLF and Nd:LuLF are similar to those of Nd:YLF. In this case, we have used a 4W GaAlAs diode laser (SDL2382P1) for the pumping excitation at 792 nm. The emission cross-sections for the ${}^{4}F_{3/2} \rightarrow {}^{4}I_{11/2}$ transitions (π and σ polarizations), were obtained by using the method of McCumber ⁽⁵⁾.

Obtained Results

The absorption spectra have shown a close resemblance with that of Nd:YLF. Thus, the most suited wavelengths for diode-laser pumping are 797 nm and 792 nm. The measured ${}^{4}F_{3/2}$ level lifetimes were around 500 µs, as shown in Figure 1.

The obtained emission cross-sections for Nd:LuYLF are shown in Figure 2, for both polarizations. The peak emission cross-section measured for the three main crystals under study are presented in Table 2.

A detailed measurement of the fluorescent emission spectrum around 1047 nm (like the spectra presented in Figure 3) allowed the determination of the spectral linewidths presented in Figure 4 (filled circles). The continuous line is an empirical fit. We readily notice that Nd:LuYLF has almost the same emission linewidth as Nd:LuLF, broadened approximately by 25% compared with that of the Nd:YLF sample, under this high intensity diode laser pumping.



Figure 1. Measured ${}^{4}F_{3/2}$ level lifetime for the studied samples.

Sample	π polarization (cm ²)	σ polarization (cm ²)
YLF	2.23 x 10 ⁻¹⁹	1.67 x 10 ⁻¹⁹
LuLF	1.65 x 10 ⁻¹⁹	1.29 x 10 ⁻¹⁹
LuYLF	2.70 x 10 ⁻¹⁹	2.25 x 10 ⁻¹⁹





Figure 2. Obtained emission cross-sections for Nd:LuYLF, for both polarizations.



Figure 3. Comparative linewidth broadening between Nd:LuYLF and Nd:YLF.

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Figure 4. Measured spectral linewidths at the 1047 nm emission (π) , for the studied samples with different Lu concentrations.

Conclusions

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Except by the emission linewidth broadening, no significant differences were measured among spectroscopic or CW laser parameters of Nd:LuYLF and Nd:YLF. Thus, Nd:LuYLF can be used for mode locking purposes with advantages over Nd:YLF. A complete short-pulse operation of this system is currently under investigation.

Acknowledgements

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