

Polarization-Dependent Enhancement of Population Inversion and of Green Upconversion in Er:LiNbO₃ by Yb Codoping

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Abstract—We have investigated the polarization dependence of both the population inversion of the $^4I_{13/2}$ state and the green upconversion to the $^4S_{3/2}$ state in Er:LiNbO₃ when codoped with Yb and pumped around 980 nm. The presence of Yb³⁺ enhances the population of the $^4I_{13/2}$ state of the Er³⁺ ions when pumped at σ -polarization (>2.5 times) and at π -polarization (>5.5 times), and introduces a broad useful pump band around 940–960 nm at σ -polarization, relaxing fabrication tolerances on semiconductor pump sources. An enhancement of green upconversion is also observed by the Yb codoping. Lifetime measurements indicate that, as in glass substrates, nonradiative energy transfer is responsible for these enhancements.

Index Terms—Erbium materials/devices, infrared spectroscopy, lithium materials/devices, optical amplifiers, ytterbium materials/devices.

THE SUCCESS of Er-doped fiber amplifiers (EDFA) and lasers has spawned the development of both planar glass waveguide [1] and LiNbO₃-based [2], [3] versions. The latter substrate is of particular interest because it provides the capability of integrating stimulated emission and second order nonlinear optical elements. In addition, because LiNbO₃ is a crystalline host, it produces less inhomogeneous broadening, and hence higher absorption and emission cross sections than amorphous glass substrates. Model calculations and experiments with both Er-doped fiber amplifiers [4] and Er-doped LiNbO₃ waveguide amplifiers [5] have shown that a higher efficiency can be obtained at the 980-nm pump wavelength than using 1480 nm. In particular, we have reported a differential gain of 1.6 dB/cm over a 1.7-cm-long waveguide with a coupled pump power of 80 mW at 980 nm in an Er:LiNbO₃ waveguide [6]—a result comparable to that at 1480 nm [2] but requiring only half the pump power. The narrow (<3 nm) absorption bandwidth at 980 nm and the weak absorption along the π -polarization, however, necessitate the careful selection and tuning of pump diode lasers. It has been shown that codoping Er³⁺ with Yb³⁺ increases the useful absorbed pump power at 980 nm in fiber lasers [7]. In this letter, we report the polarization-dependent enhancement of the $^4I_{13/2}$ state population in Er:LiNbO₃ codoped with Yb³⁺ when pumped

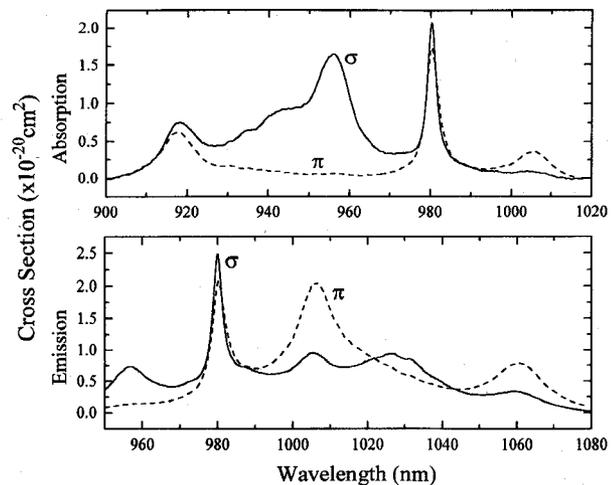


Fig. 1. Measured room temperature absorption and calculated emission cross sections of the $^2F_{5/2} \rightarrow ^2F_{7/2}$ transition of Yb-doped LiNbO₃ at σ (solid line) and π (dashed line) polarizations.

around 980 nm. We also find an increase in upconversion to the green (~ 560 nm) in the presence of Yb³⁺.

Bulk-doped Er:LiNbO₃, Yb:LiNbO₃, and Er:Yb:LiNbO₃ samples of different concentrations were provided by Tianjin University, China. The 1.53- μ m absorption and emission cross sections and amplification properties of the Er³⁺ $^4I_{13/2} - ^4I_{15/2}$ transition in LiNbO₃ can be found in [8]. For the Yb-only system, the absorption and spontaneous emission spectra (using a 955-nm pump) of the $^2F_{5/2} - ^2F_{7/2}$ transition (~ 980 nm) were measured from a 1.5 mol% Yb:LiNbO₃ bulk sample. The Stark split energy levels of these two manifolds were obtained from these room temperature spectra: 0, 264, 467, and 779 cm^{-1} for the $^2F_{7/2}$ state, and 10204, 10460, and 10893 cm^{-1} for the $^2F_{5/2}$ state. These numbers were used to derive the absorption and emission cross sections (Fig. 1) of Yb:LiNbO₃ using McCumber theory [8]. As seen in Figs. 1 and 2, the Yb³⁺ spectra overlap the Er³⁺ $^4I_{11/2} - ^4I_{15/2}$ transition, as in glass and other hosts. The peak wavelength of Yb³⁺ is nearly coincident with that of the Er³⁺ (at 980 nm), enhancing energy exchange process.

The sum of the absorption coefficients (Fig. 2) around 980 nm of Er-only (1.0 mol%) and Yb-only (1.6 mol%) is nearly identical to that of LiNbO₃ doped with 1.0 mol% Er³⁺ and 1.6 mol% Yb³⁺, indicating that the optical properties of the Er³⁺ and Yb³⁺ ions are distinct from one another in the

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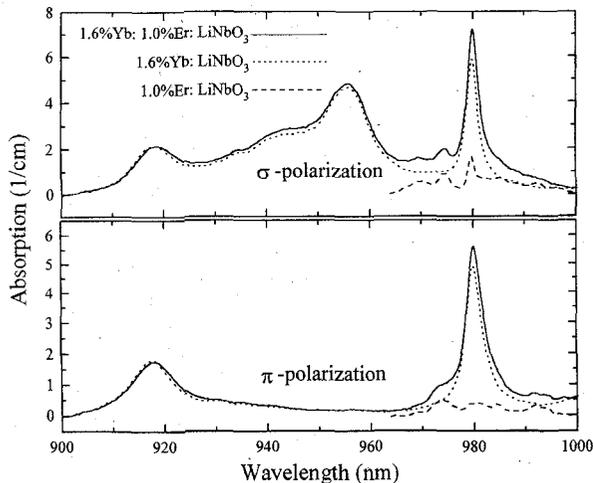


Fig. 2. The absorption spectra of 1.0 mol%Er:LiNbO₃, 1.6 mol%Yb:LiNbO₃, and codoped 1.0 mol%Er:1.6 mol%Yb:LiNbO₃ samples measured at σ - and π -polarizations.

codoped samples. As further evidence of this, we found that the Er³⁺ absorption cross sections for the $^4I_{15/2} \rightarrow ^4I_{13/2}$ (~ 1530 nm), $^4I_{15/2} \rightarrow ^4I_{9/2}$ (~ 800 nm), and $^4I_{15/2} \rightarrow ^4F_{9/2}$ (~ 660 nm) transitions, as well as the radiative lifetime of the $^4I_{13/2} \rightarrow ^4I_{15/2}$ fluorescence, remain unchanged in the presence of the Yb³⁺ codopant. For purposes of model calculations, we therefore assume, as a first approximation, that the optical properties of Er³⁺ are unaffected by the Yb³⁺ doping.

As in the case of codoped glass fibers, we expect a portion of the light energy absorbed by Yb³⁺ to be transferred to Er³⁺. In order to verify this, we compared the excitation spectra of the Er-only and the codoped samples by monitoring the Er 1531-nm fluorescence intensity while scanning the pumping wavelength. The samples (500- μ m thick) were pumped by a tunable Ti:sapphire laser focused to a spot ~ 0.2 mm (~ 950 W/cm²). Fig. 3 shows that the 1531-nm fluorescence is enhanced by 2.5 and 5.5 times in the σ - and π -polarizations, respectively, in the presence of Yb³⁺. That this enhancement is a direct energy transfer process from Yb³⁺ to Er³⁺ (rather than emission and re-absorption) is seen in the decrease in the measured lifetime of the $^2F_{5/2}$ manifold of Yb³⁺ (measured at 1010 nm) from 580 μ s in 1.6 mol%Yb:LiNbO₃ to 360 μ s in 1.6 mol%Yb:1.0 mol%Er:LiNbO₃. This energy transfer populates the $^4I_{11/2}$ manifold of Er³⁺ (lifetime 210 μ s), providing a parallel relaxation route out of the $^2F_{5/2}$ of Yb³⁺ and nearly halving the $^2F_{5/2}$ lifetime. Note that the lifetime of Er³⁺ (210 μ s) is of the same order as that of Yb³⁺ (580 μ s), suggesting that energy back-transfer from Er³⁺ to Yb³⁺ is also possible in the codoped system. The presence of Yb³⁺ does not alter the Er³⁺ lifetime of the 1531 nm fluorescence (~ 4.3 ms), indicating that no significant lifetime quenching of the $^4I_{11/3}$ manifold is induced by the Yb³⁺ codoping (up to 1.6 mol%).

We note that the large increase in the absorption coefficient of codoped LiNbO₃ along the π -polarization (whose absorption is originally very weak in Er-only samples, Fig. 2) creates the possibility of using unpolarized pump sources, such as high

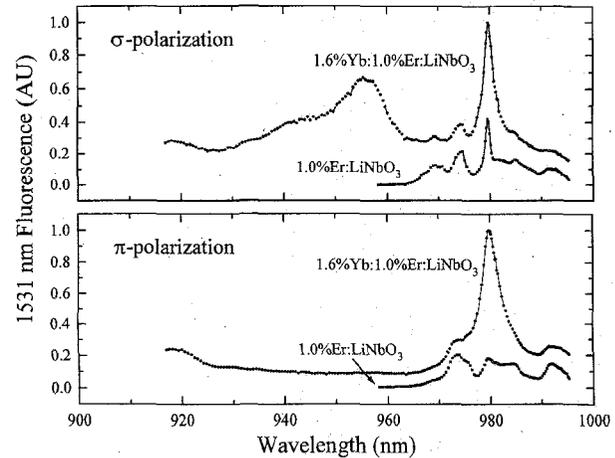


Fig. 3. The excitation spectra of a singly doped 1.0 mol%Er:LiNbO₃ and a codoped 1.0 mol%Er:1.6 mol%Yb:LiNbO₃ samples measured at σ - and π -polarizations.

power LED's. Another important feature of the σ -polarization spectrum is the addition of new broad excitation wavelengths from 940 nm to 960 nm, which greatly relieves the wavelength tolerance for diode laser pump sources.

We can use Figs. 2 and 3 to calculate the effective energy transfer efficiency of the Er/Yb codoped system. Using σ -polarized 980-nm pump light, for example, the pump light absorbed by the codoped sample is 4.5 times that of the Er-only doped sample (Fig. 2), and the emitted 1531-nm fluorescence is increased 2.5 times (Fig. 3). Therefore, the energy transfer efficiency is approximately $2.5/4.5 = 56\%$. The total quantum efficiency for the codoped system is therefore about $0.56\eta_o$, where η_o is the quantum efficiency of the Er-only system. The reduction of the total quantum efficiency is mostly due to the radiative loss of the Yb³⁺ via $^2F_{5/2} \rightarrow ^2F_{7/2}$ transition. The increased absorption, however, may be worth the penalty of a lower effective quantum efficiency because integrated-optic devices provide only a limited length over which to perform amplification, and only a small fraction of the pump power is absorbed by the Er³⁺ ions. We can, therefore, increase the degree of 980 nm pump-power absorption, and therefore the population inversion and differential gain of Er³⁺, by adding Yb³⁺.

When the Er³⁺ ions are pumped to the $^4I_{11/2}$ state, this state can upconvert to the $^4S_{3/2}$ state and produce green fluorescence [9]. This process may be caused by the cooperative upconversion (CUC) between two closely spaced, excited Er³⁺ ions in the $^4I_{11/2}$ state, or by the excited state absorption (ESA) of a single excited Er³⁺ ion. We found this upconversion process in LiNbO₃ is also significantly enhanced in the codoped sample, as seen in a plot of the upconversion intensity versus 980-nm pump power (inset of Fig. 4). Since the upconversion is a two-photon process, the pump-power dependence of the green fluorescence in the Yb:Er codoped sample, like the Er-only sample, is quadratic in nature (i.e., curves with slope $\cong 2$).

In order to determine the mechanism of the upconversion, we measured the decay curve of the green fluorescence with a fast photo-multiplier tube (Fig. 4). If the ESA is the dominant

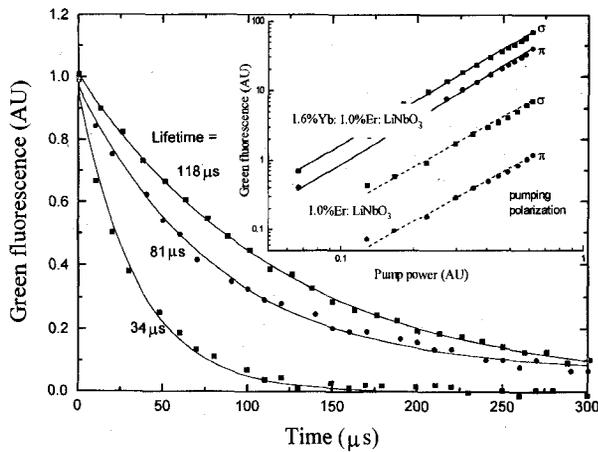


Fig. 4. The measured green upconversion fluorescence decay (dots) of Er:LiNbO₃ (1.0 mol%) codoped with different concentrations of Yb³⁺ pumped at 980 nm. From top to bottom: 1.6 mol% Yb³⁺, 0.6 mol% Yb³⁺, and no codoping. The solid lines are the single-exponential fits. Inset shows the quadratic pump-power dependence of green light of singly and codoped samples pumped along two polarizations.

ing effect, the green light should show a single exponential decay with the $^4S_{3/2}$ state characteristic lifetime ($\sim 30 \mu\text{s}$ [10]). If CUC dominates, on the other hand, the decay will show a longer lifetime because the presence of the $^4I_{11/2}$ state population (lifetime $\sim 210 \mu\text{s}$) continues to cooperatively upconvert to the $^4S_{3/2}$ state even after the pump light is turned off, extending the lifetime of the green fluorescence. CUC usually becomes more important than ESA for high sample concentrations because the ions are more closely spaced, increasing the energy transfer probability. Previous results [10] have shown that only ESA is significant in a 0.16 mol% Er:LiNbO₃ sample. For our sample with much higher concentration (1.0 mol%), the lifetime result (Fig. 4) for the Er-only sample ($34 \mu\text{s}$) shows that ESA still dominates. When doped with Yb, however, we can see that the green light lifetime is significantly increased, to as much as $118 \mu\text{s}$. Since it is unlikely that the Yb codoping will dramatically increase the possibility of Er-Er pair CUC, we believe that the increased green upconversion is most likely due to CUC in the Yb-Er pair.

In a waveguide structure where larger pumping intensities can be achieved, large upconversion efficiency is possible due to the quadratic dependence, which suggests the possibility of making a coherent green-light source based on this material. It should be noted that the green upconverted light is an

important source of photorefractive damage in Er:LiNbO₃ waveguide amplifiers, and using ZnO waveguide in Mg-doped LiNbO₃ [5] substrates may permit one to fabricate a stable green waveguide laser device.

In summary, we have measured and calculated the absorption and emission cross sections of Yb:LiNbO₃. We note that, in addition to our studies, these data are also applicable to modeling Yb:LiNbO₃ waveguide lasers, first demonstrated by Jones *et al.* [11]. We have also observed that the Er:LiNbO₃ (1.0 mol%) 1531 nm fluorescence is enhanced 5.5 (2.5) times for the π - (σ -) polarization by incorporating 1.6 mol% Yb and pumping at 980 nm. The broad-absorption band (940 nm to 960 nm) produced by Yb codoping should make it possible to pump Er with semiconductor light sources having relaxed fabrication tolerances. Green upconversion is also enhanced 33 (10) times in the π - (σ -) polarization when pumped at this wavelength.

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