



HAL
open science

Highly-Doped Er:LiYF₄ Waveguiding Epitaxial Films for 2.7 μm Laser Sources

Liza Basyrova, Pavel Loiko, Gurvan Brasse, Abdelmjid Benayad, Jean-Louis Doualan, Alain Braud, Patrice Camy

► **To cite this version:**

Liza Basyrova, Pavel Loiko, Gurvan Brasse, Abdelmjid Benayad, Jean-Louis Doualan, et al.. Highly-Doped Er:LiYF₄ Waveguiding Epitaxial Films for 2.7 μm Laser Sources. CLEO: Science and Innovations, 2021, San Jose, United States. pp.STh1E.5, 10.1364/CLEO_SI.2021.STh1E.5 . hal-03368751

HAL Id: hal-03368751

<https://hal.science/hal-03368751v1>

Submitted on 7 Oct 2021

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Highly-Doped Er:LiYF₄ Waveguiding Epitaxial Films for ~2.7 μm Laser Sources

Liza Basyrova*, Pavel Loiko, Gurvan Brasse, Abdelmjid Benayad, Jean-Louis Doualan, Alain Braud, and Patrice Camy

Centre de recherche sur les Ions, les Matériaux et la Photonique (CIMAP), UMR 6252 CEA-CNRS-ENSICAEN, Université de Caen Normandie, 6 Boulevard du Maréchal Juin, 14050 Caen Cedex 4, France

*liza.basyrova@ensicaen.fr

Abstract: Single-crystalline oriented highly-doped 9.6 at.% Er³⁺:LiYF₄ waveguiding thin films (30–50 μm) are grown by Liquid Phase Epitaxy. Their polarized spectroscopic properties are studied. The maximum stimulated-emission cross-section is 1.61×10^{-20} cm² at 2720 nm for π -polarization. © 2020 The Author(s)

Erbium (Er³⁺) ions are well-known due to their mid-IR emission at ~2.7 μm owing to the $^4I_{11/2} \rightarrow ^4I_{13/2}$ transition. ~2.7 μm Er lasers are of practical importance for medicine, infrared countermeasures, spectroscopy and pumping of mid-infrared OPOs. The difficulty in developing ~2.7 μm Er lasers is the self-terminating nature of the $^4I_{11/2} \rightarrow ^4I_{13/2}$ transition. It is typically overcome by using high Er doping levels [1]. Among the gain media for mid-IR Er lasers, fluoride crystals have attracted a lot of attention. For example, Er³⁺-doped yttrium lithium fluoride (LiYF₄) crystal was successfully employed in ~2.7 μm lasers [2].

Waveguide lasers emitting in the near- and mid-IR are of interest for the development of compact ultrafast oscillators [3]. Few results concerning ~2.7 μm waveguide lasers have been already reported [4]. Rare-earth-doped LiYF₄ is well-known for fabrication of active waveguides by Liquid Phase Epitaxy (LPE) [5]. However, so far, Er³⁺:LiYF₄ active layers were not fabricated. In the present work, we aimed to grow highly Er³⁺-doped LiYF₄ single-crystalline oriented waveguiding films and to study their spectroscopic properties relevant for ~2.7 μm laser operation.

Er³⁺:LiYF₄ thin-films were grown on (001)-oriented bulk undoped LiYF₄ substrates by LPE using a batch with a composition of 73 mol% LiF – 27 mol% (YF₃ + ErF₃); LiF was used as a solvent also entering into the composition of the solute. The growth temperature was 729.5 – 730.5 °C and the growth duration was 45 min. The Er³⁺ doping level in the flux was 11 at.% and the segregation coefficient for Er³⁺ ions K_{Er} was 0.88. The thickness of the films was in the range 30 – 50 μm (growth rate: 0.7–1.1 μm/min). The structure (cubic, sp. gr. $C_{4h}^6 - I4_1/a$) and orientation of the films were confirmed by single-crystal XRD. The film composition was studied by EDX.

Figure 1(a) shows an optical image of a polished end-facet of the Er³⁺:LiYF₄/LiYF₄ epitaxy. The film / substrate interface is clear, there are no cracks nor secondary phases in the film. The film thickness, $t = 38 \pm 2$ μm, is nearly constant over the substrate area deep in the flux. The morphology of the as-grown top surface reveals smooth “hills and valleys” landscape and crystallized residual LiF solvent which is typical for LPE films, Fig. 1(b).

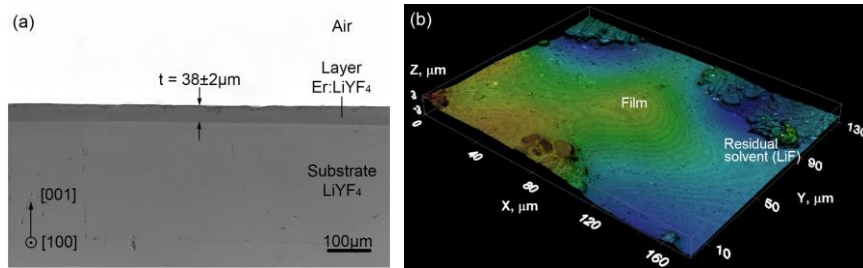


Fig. 1. (a) Optical microscope image of the polished end-facet of the Er³⁺:LiYF₄ / LiYF₄ epitaxy after cleaning the residual solvent; (b) Confocal microscope image (interferometric regime, reflection mode, $\lambda = 405$ nm) of the as-grown top surface of the film.

The absorption cross-section, σ_{abs} , spectra for transitions of Er³⁺ ions from the ground-state $^4I_{15/2}$ to the excited-states $^4I_{13/2}$, $^4I_{11/2}$ and $^4I_{9/2}$ are shown in Fig. 2(a) for the available σ -polarization. For the most common pump transition ($^4I_{15/2} \rightarrow ^4I_{11/2}$), $\sigma_{abs} = 0.46 \times 10^{-20}$ cm² at 972.3 nm and the absorption bandwidth (FWHM) is 6.2 nm. For the $^4I_{15/2} \rightarrow ^4I_{9/2}$ one, σ_{abs} is much lower, $\sim 0.05 \times 10^{-20}$ cm² at 781 nm, but the absorption band is broader.

The stimulated-emission (SE) cross-sections, σ_{SE} , for the $^4I_{11/2} \rightarrow ^4I_{13/2}$ transition in emission were calculated using the Füchtbauer–Ladenburg (F-L) equation, Fig. 2(b), based on the polarized luminescence spectra and using a radiative lifetime $\tau_{rad} = 6.7$ ms and a luminescence branching ratio $\beta = 15\%$ determined from the Judd-Ofelt theory. As the

considered transition has both electric-dipole (ED) and magnetic-dipole (MD) contributions, three polarization states were considered: σ ($\mathbf{E} \perp \mathbf{c}, \mathbf{k} \perp \mathbf{c}$), α ($\mathbf{E} \perp \mathbf{c}, \mathbf{k} \parallel \mathbf{c}$) and π ($\mathbf{E} \parallel \mathbf{c}, \mathbf{k} \perp \mathbf{c}$) - LiYF₄ is optically uniaxial with the optical axis parallel to the \mathbf{c} -axis. The maximum σ_{SE} is $1.61 \times 10^{-20} \text{ cm}^2$ at 2720 nm and at longer wavelengths where the laser operation is expected (owing to the ESA), it is $0.69 \times 10^{-20} \text{ cm}^2$ at 2812 nm (both values correspond to π -polarization).

The crystal-field splitting of the multiplets involved in the $\sim 2.7 \mu\text{m}$ laser transition ($^4I_{11/2}$ and $^4I_{13/2}$) is revealed using LT (12 K) spectroscopy, Fig. 2(c).

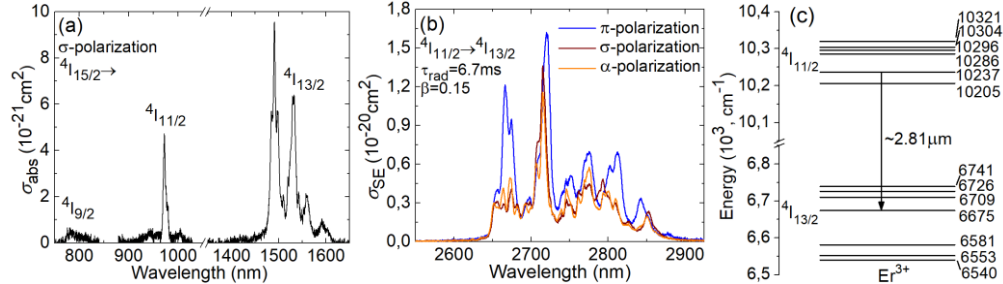


Fig. 2. Spectroscopy of 9.6 at.% Er³⁺:LiYF₄ LPE films: (a) absorption cross-sections, σ_{abs} , for the $^4I_{15/2} \rightarrow ^4I_{9/2}$, $^4I_{11/2}$ and $^4I_{13/2}$ transitions (σ -polarization), (b) stimulated-emission, σ_{SE} , cross-sections for the $^4I_{11/2} \rightarrow ^4I_{13/2}$ transition, light polarizations: π , σ and α ; (c) crystal-field splitting for the $^4I_{11/2}$ and $^4I_{13/2}$ multiplets of Er³⁺ ions resolved at 12 K.

The luminescence lifetimes τ_{lum} of different multiplets of Er³⁺ ions (from $^4I_{13/2}$ to $^4S_{3/2} + ^2H_{11/2}$) were systematically studied, Fig. 3. For the levels involved in the $\sim 2.7 \mu\text{m}$ laser transition, $\tau_{\text{lum}}(^4I_{11/2}) = 2.71 \text{ ms}$ and $\tau_{\text{lum}}(^4I_{13/2}) = 8.36 \text{ ms}$.

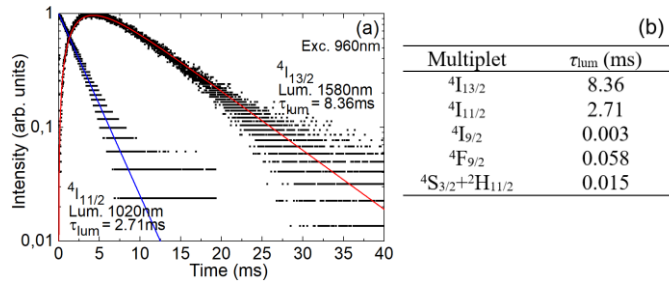


Fig. 3. (a) Luminescence decay curves for emissions from the $^4I_{13/2}$ and $^4I_{11/2}$ states of Er³⁺ ions in the 9.6 at.% Er³⁺:LiYF₄ epitaxial film, $\lambda_{\text{exc}} = 960 \text{ nm}$; (b) summary of the measured luminescence lifetimes τ_{lum} for Er³⁺ multiplets.

The fabricated films exhibit a positive refractive index contrast, $\Delta n = n_{\text{film}} - n_{\text{substrate}} \approx 2 \times 10^{-3}$ owing to the effect of Er³⁺ dopant ions. The waveguiding properties of the films are confirmed by coupling the pump radiation at 972 nm from a Ti:sapphire laser. The waveguide laser experiments are in progress.

To conclude, highly-doped single-crystalline oriented Er³⁺:LiYF₄ thin-films (thickness: up to 50 μm) of high optical quality are grown on bulk LiYF₄ substrates by LPE. They exhibit intense mid-IR emission at $\sim 2.7 \mu\text{m}$ and weak quenching of the $^4I_{11/2}$ lifetime, as well as waveguiding properties. Er³⁺:LiYF₄ epitaxial films are promising for mid-IR waveguide lasers. Further work will focus on laser experiments and adjustment of the Er³⁺ doping level in order to optimize energy-transfer processes populating the upper laser level.

References

- [1] R. C. Stoneman and L. Esterowitz, "Efficient resonantly pumped 2.8- μm Er³⁺:GSGG laser," *Opt. Lett.* **17**(11), 816-818 (1992).
- [2] S. Hubert, D. Meichenin, B.W. Zhou, and F. Auzel, "Emission properties, oscillator strengths and laser parameters of Er³⁺ in LiYF₄ at 2.7 μm ," *J. Lumin.* **50**(1), 7-15 (1991).
- [3] C. Khurmi, N. B. Hébert, W. Q. Zhang, S. Afshar V., G. Chen, J. Genest, T. M. Monro, and D. G. Lancaster, "Ultrafast pulse generation in a mode-locked Erbium chip waveguide laser," *Opt. Express* **24**(24), 27177-27183 (2016).
- [4] D. G. Lancaster, S. Gross, H. Ebdorff-Heidepriem, M. J. Withford, T. M. Monro, and S. D. Jackson, "Efficient 2.9 μm fluorozirconate glass waveguide chip laser," *Opt. Lett.* **38**(14), 2588-2591 (2013).
- [5] P. Loiko, R. Soulard, G. Brasse, J.-L. Doualan, B. Guichardaz, A. Braud, A. Tyazhev, A. Hideur, and P. Camy, "Watt-level Tm:LiYF₄ channel waveguide laser produced by diamond saw dicing," *Opt. Express* **26**(19), 24653-24662 (2018).