Liquid phase epitaxy of rare earth doped $LiYF_4$ layers for the elaboration of compact laser planar waveguides

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Introduction:

The miniaturization of optical devices generates a growing interest, especially in the laser domain, in order to develop more and more compact photonic devices for their integration in various systems. The targeted applications increase considerably in various domains like the detection of atmospheric pollutants, biomedical applications, Lidar technologies, RGB display or quantum information. Laser planar waveguides have thus been elaborated, especially Tm^{3+} doped or Pr^{3+} doped $LiYF_4$ (YLF) thick layers epitaxially grown on undoped YLF substrates, to get original emissions in the visible domain or in the mid-infrared spectral range.

Description of the experimental process:

Two crystal growth techniques have been used successively to obtain crystalline optical waveguides : the Czochralski technique, first, to synthesize pure $LiYF_4$ substrates, then the liquid phase epitaxy (LPE) technique for the homoepitaxial growth of Tm^{3+} and Pr^{3+} doped $LiYF_4$ monocristalline layers on the substrates.

A fluorination step of the oxides raw materials required for the preparation of the melts at the origin of the substrates and the epilayers, is needed to get pure fluoride precursors. For that purpose, the NH_4HF_2 chemical precursor is used as fluorinating agent and the resulting chemical equation of the reaction is:

$$RE_2O_3 + 6NH_4HF_2 \xrightarrow{\text{Stirring + heating at } 240^\circ C} 2REF_3 + 6NH_4F + \overset{\nearrow}{3H_2O}$$

The so prepared material is then annealed at 650°C during few hours to eliminate the excess of NH_4HF and the residual water.

The bulk crystals obtained by using the Czochralski technique (boules of about 3 cm in diameter and 6 cm long) are cut perpendicularly to the (001) growth axis and prepared in the form of plates with a thickness of about 2 mm and rectangular faces of about 1x3 cm^2 to be used as substrates. Then, the rectangular faces on which the crystal growth occurs during the LPE process are very carefully and finely polished, in order to present a roughness of the order of a few nm and a flatness of few fringes. Such a step is crucial to obtain layers with a good crystalline quality and to limit the internal optical losses as much as possible.

The molar composition of the LPE bath is determined from the phase diagram of the system $LiF\text{-}YF_3$ [1] and is composed as follows: 73% of LiF and 27% of a mixture composed of YF_3 and of a few percents of GdF_3.GdF_3 is used to ensure the necessary refractive index contrast between epitaxial layer and substrate and also because Gd^{3+} ions only absorb light in the near UV, thus in a spectral region which does not interfere with the mid-infrared and visible optical transitions of the Tm^{3+} and Pr^{3+} dopants considered in the present work.

The substrates were thus immerged in the LPE bath at a temperature slightly lower than the saturation temperature, to initiate the epitaxial process, and the growth occurs on both sides of the substrates. The thickness of the layers is a function of the immersion duration and can be varied from a few microns to a hundred of microns. Moreover, the purity of the growth atmosphere, which has to be absolutely oxygen free, is also a key parameter for the crystal growth of fluoride layers. Once the epitaxial layers have been grown with the required thickness, the samples are cooled down to room temperature very slowly to avoid cracks and structural defects. They are then polished on the both faces to remove the residual deposit of LiF and also to adjust the thickness of the guiding layers.

To finalize the preparation of the waveguide samples before testing them in a laser cavity, it was also necessary to polish their input and output faces to have good pump and laser input and output coupling efficiencies and to ensure a good parallelism of the faces, thus optimal lasing conditions.

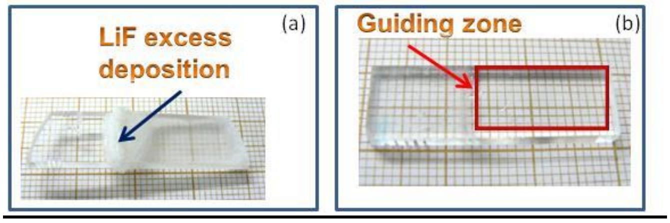

Figure 1: (a) Sample just after the LPE growing process, (b) sample prepared as planar waveguide.

Characterizations of the epitaxial layers and laser performances of the planar waveguides:

The obtained layers were all initially characterized by controlling the nature of the crystallized phase by using X-Ray Diffraction, then by determining the real dopant concentrations in the deposited layers by using absorption spectroscopy and EDX analysis. The refractive index contrast between the layers and the substrates, which is an important parameter for the final laser device, was also measured by using the M-Lines technique.

The planar waveguides were then tested optically to estimate the internal optical losses. Assuming nearly 100% coupling efficiency and subtracting for the Fresnel reflection losses (4%) occurring at the end faces of the waveguides, upper limits for the internal optical losses of 0.64 and 0.80 ± 0.03 dB⁄cm were obtained for the $LiY_{0.935}Gd_{0.05}Pr_{0.015}F_4$ and $LiY_{0.885}Gd_{0.05}Lu_{0.05}Pr_{0.015}F_4$ layers, and optical losses of 0.11 dB/cm and 0.26 dB/cm were obtained for the $LiY_{0.907}Gd_{0.024}Tm_{0.069}F_4$ and $LiY_{0.887}Gd_{0.024}Tm_{0.089}F_4$ layers, respectively. These losses are thus extremely low, attesting of the excellent crystalline quality of the epitaxial layers.

The good quality of the layers has been confirmed with the laser measurements. We have indeed demonstrated the first epitaxially grown Tm^{3+}-doped crystalline fluoride waveguide laser operating at 1.87 µm with an efficiency of 76% and a record output power of 560 mW [2]. we have also demonstrated, what we believe to be the first epitaxially grown Pr^{3+}-doped fluoride planar waveguide laser operating in the red (639.4 nm) and orange (604.2 nm) spectral range with output powers up to about 25 and 12 mW, respectively [3].

By adjusting the Tm^{3+} and Pr^{3+} ion concentrations in the layers, by adjusting the width and the length of the waveguides to the pump and laser conditions, and by structuring these planar waveguides to form linear waveguides, it is now expected to reach even better laser performance (lower laser threshold and higher overall laser efficiencies), which definitely open the way to the fabrication of compact, integrated, high-power, and highly efficient devices for various applications. More details and results will be given at the conference.

References:

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