Effect of the doping level on the 2.8-µm laser performance of Er^{3+}:CaF_2 crystals

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Abstract: The effect of doping level (0.1-10 at.%) on mid-infrared laser performance of Er:CaF_2 crystals was investigated. The Er:CaF_2 laser delivered 1.17 W at 2798 nm with 35.8% slope efficiency and was tuned across 2690-2830 nm.

1. Introduction

The development of lasers emitting around 3 µm has recently received tremendous interest due to their applications in medicine, molecular spectroscopy, and pumping of mid-infrared coherent light sources. Erbium ions (Er^{3+}) offer one of the most straightforward solutions to address this spectral range according to their $^4I_{11/2} \rightarrow {}^4I_{13/2}$ transition. Er^{3+}-doped materials can be pumped at 0.98 µm, e.g., by InGaAs diode lasers or high-brightness Yb-fiber lasers (YFLs), and continuous-wave (CW) operation is enabled by an energy-transfer upconversion process (ETU), $^4I_{13/2} + {}^4I_{13/2} \rightarrow {}^4I_{9/2} + {}^4I_{15/2}$, whose efficiency is boosted by providing high Er^{3+} doping levels (tens of at.%). Among laser gain media for 2.8-µm Erbium lasers, Er^{3+}-doped calcium fluoride (CaF_2) crystals appear intriguing [1] owing to their unique rare-earth ion clustering behavior [2]. The ion clustering simultaneously enhances the energy-transfer processes among the dopant ions and provides a significant inhomogeneous spectral line broadening. The latter is of primary importance for applications in mode-locked lasers, as demonstrated for Yb^{3+}:CaF_2 at ~1 µm [3].

In the present work, we aimed to reveal the effect of rare-earth ion clustering on the mid-infrared laser performance of Er^{3+}:CaF_2 crystals via a comprehensive spectroscopic and laser study.

2. Optical spectroscopy

The single-crystals of Er^{3+}:CaF_2 with a broad range of doping levels C_{Er} between 0.05 and 10 at.% were grown by the Bridgman-Stockbarger technique using graphite crucibles. To avoid oxygen pollution, the growth chamber was sealed to vacuum (<10^{-5} mbar) and refilled with a mixture of Ar + CF_4 gases.

The absorption and emission spectra, Fig. 1(a,b), evidenced important changes in the mechanisms of Er^{3+} ion incorporation in the fluorite matrix: at a very low C_{Er} of 0.05 at.%, predominantly isolated ions residing in several sites (C_{3v}, C_{4v}, O_h) were observed and already at 0.1 at.%, a strong tendency for ion clustering was noted, resulting in a profound inhomogeneous spectral line broadening. The complexity of clusters further increased with the doping level. For 5 at.% Er^{3+}:CaF_2, the maximum absorption cross-section σ_{abs} for the $^4I_{15/2} \rightarrow {}^4I_{11/2}$ pump transition was 0.21×10^{-20} cm^2 at 981.3 nm (FWHM: 21 nm). For the $^4I_{11/2} \rightarrow {}^4I_{13/2}$ Er^{3+} transition giving rise to mid-infrared emission, at the expected laser wavelength, the stimulated-emission (SE) cross-section σ_{SE} was 0.40×10^{-20} cm^2 at ~2798 nm. The luminescence lifetimes τ_{lum} of the $^4I_{11/2}$ and $^4I_{13/2}$ manifolds strongly varied with the doping level, revealing a favorable ratio around C_{Er} of 3 – 5 at.%. For 5 at.% Er^{3+}:CaF_2, τ_{lum} = 4.83 ms ($^4I_{11/2}$) and 2.49 ms ($^4I_{13/2}$).

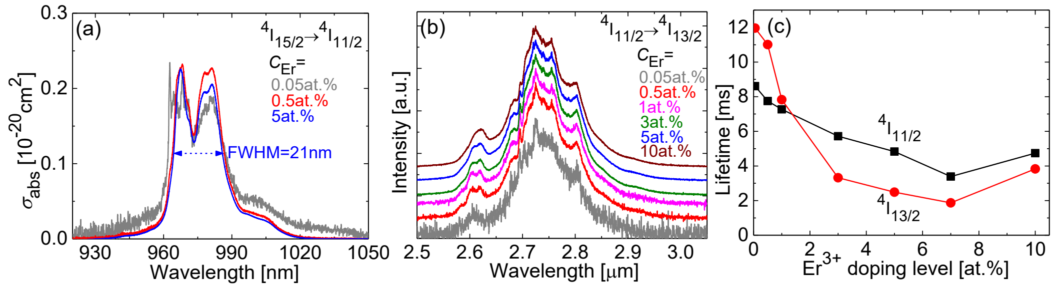

Fig. 1. Optical spectroscopy of Er^{3+} ions in CaF_2 crystals: (a) absorption cross-section, σ_{abs}, spectra for the $^4I_{15/2} \rightarrow {}^4I_{11/2}$ transition; (b) luminescence spectra, the $^4I_{11/2} \rightarrow {}^4I_{13/2}$ transition, λ_{exc} = 976 nm; (c) mean luminescence lifetimes of the $^4I_{11/2}$ and $^4I_{13/2}$ manifolds as a function of Er^{3+} doping level.

3. Laser performance

The laser performance of a set of Er^{3+}:CaF_2 crystals (C_{Er} = 0.1 - 10 at.%) was studied. The cylindrical laser elements (Φ8 mm, thickness: 6-8 mm) were mounted in a passively cooled Cu-holder and placed in a hemispherical cavity formed by a flat dichroic pump mirror (PM) coated for HT at 0.98 µm and HR at 2.6-3 µm and a concave (radius of curvature: RoC = -100 mm) output coupler (OC) with a transmission T_{OC} of 1.7% at the laser wavelength, Fig. 2(a). The cavity length was ~99 mm. The pump source was a commercial Yb-fiber laser (YFL) emitting up to 6 W at 976 nm with a near diffraction limited beam quality ($M^2 \approx 1$). The pump radiation was focused into the crystal through the PM using an AR coated achromatic lens (f = 75 mm). The single-pass pump absorption monotonously increased with the doping level, from 5% (0.1 at.% Er) to 90% (10 at.% Er). A long pass filter for was used to remove the non-absorbed pump after the OC. Figure 2(b) depicts a typical far-field beam profile of the 2.8 µm Er:CaF_2 laser.

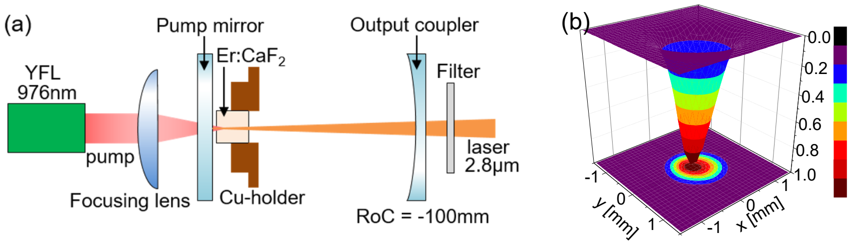

Fig. 2. (a) Layout of the Er:CaF_2 laser pumped by a Yb-fiber laser (YFL); (b) a typical far-field profile of the laser beam.

The continuous wave (CW) laser operation was achieved for the entire range of the studied Er^{3+} doping levels, Fig. 3(a,b). The 5 at.% Er^{3+}:CaF_2 laser generated a maximum output power of 1.17 W at 2798 nm with the highest slope efficiency η of 35.8% (versus the absorbed pump power) and a very low laser threshold P_{th} of 14 mW. The laser slope efficiency exceeded the Stokes limit owing to efficient energy-transfer upconversion (ETU), $^4I_{13/2}$ + $^4I_{13/2} \rightarrow {}^4I_{9/2}$ + $^4I_{15/2}$, refilling the upper laser level. The typical spectra of laser emission are shown in Fig. 3(b). Multiple emission lines were observed, 2746, 2750 and 2758 nm (for small C_{Er} of 0.1 – 1 at.%) and 2798 and 2809 nm (for higher doping levels). Figure 3(c) illustrates the dependence of the laser slope efficiency and laser threshold on C_{Er}. For both parameters, a range of optimum Er^{3+} doping levels of 3 – 5 at.% was observed. This behavior can be explained as follows: first, an increase of C_{Er} promotes Er^{3+} ion clustering which enhances ETU and leads to a favorable $^4I_{11/2}$ to $^4I_{13/2}$ lifetime ratio notably improving the laser performance. At too high doping levels, the unwanted energy migration to defects and impurities becomes the major limiting factor.

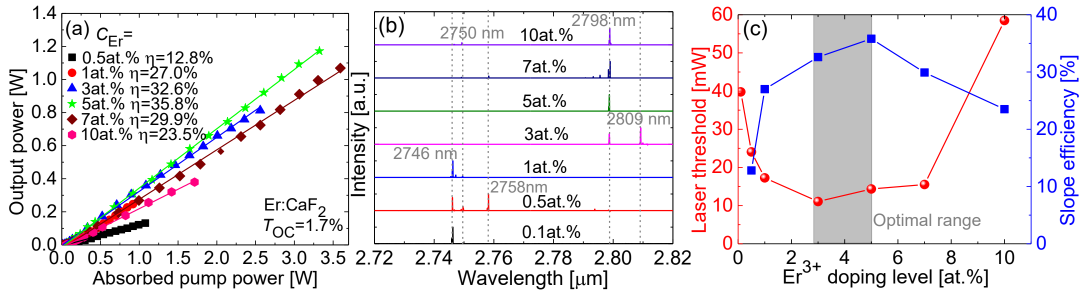

Fig. 3. Effect of the Er^{3+} doping level on the 2.8-µm performance of Er:CaF_2 lasers pumped by an Yb-fiber laser at 976 nm: (a) input-output dependences, η – slope efficiency; (b) typical spectra of laser emission measured well above the laser threshold; (c) evolution of the laser threshold and slope efficiency vs. the Er^{3+} doping level. T_{OC} = 1.7% (0.5 - 10 at.% Er), 0.33% (0.1 at.% Er).

The broadband emission properties of Er^{3+} ions in CaF_2 were exploited by studying the wavelength tunabilty. For this, a birefringent filter (a 0.8 mm-thick MgF_2 plate) was inserted into the laser cavity at the Brewster's angle. The 5 at.% Er^{3+}:CaF_2 laser was continuously tunable between 2690 and 2830 nm (tuning range: 140 nm).

4. Conclusion

To conclude, optimization of the Er^{3+} doping level is a key solution for unlocking power-scalable, low-threshold and highly efficient laser operation of Er^{3+}:CaF_2 crystals at 2.8 µm. We report on a Watt-level Er^{3+}:CaF_2 laser pumped by a high-brightness 976-nm Yb-fiber laser with a slope efficiency (~36%) exceeding the Stokes limit and a very low laser threshold (14 mW). We also demonstrate CW laser operation for very low doping levels of 0.1-0.5 at.% highlighting the role of ion clustering in boosting the ETU efficiency in Er^{3+}-doped fluorite crystals. Further power scaling is envisioned under diode pumping at 0.98 µm or 1.45 µm.

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