Spectroscopy of Yb^{3+}-doped multicomponent alkaline earth metal fluorides for ultrafast lasers

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Abstract. The optical spectroscopy of an ytterbium-doped multicomponent alkaline earth metal fluoride crystal Yb^{3+},Y^{3+}:(Ca,Sr)F_2 was studied with the goal of developing novel gain media for ultrafast mode-locked oscillators. The contribution of phonon-assisted (Stokes) emission to extending the gain profile beyond the range of electronic transitions was evidenced.

1 Introduction

Alkaline earth metal fluorides MF_2 (where M stands for Ca, Sr, Ba or their mixture), are attractive laser host materials for doping with Ytterbium ions (Yb^{3+}) for ultrafast lasers operating around 1 μm. This is due to the rare-earth ion clustering inducing inhomogeneous spectral line broadening resulting in "glassy-like" spectral gain profiles supporting sub-100 fs pulse generation while preserving good thermal properties of the host matrix. Apart from solid-solution compounds (M_1,M_2)F_2, codoping with optically inactive "buffer" ions such as Y^{3+}, Gd^{3+} or La^{3+} could modify the cluster composition and the crystal-field around the active ions, contributing to the emission bandwidth engineering via an additional degree of disorder. In this work, we report on a detailed spectroscopic study of a multicomponent fluorite-type crystal, Yb,Y:(Ca,Sr)F_2, for applications in ultrashort-pulse 1-μm mode-locked lasers.

2 Results and discussion

The Yb^{3+},Y^{3+}-codoped calcium-strontium fluoride solid-solution crystal was grown by the temperature gradient technique under Argon atmosphere. The actual doping level were 2.86 at.% Yb^{3+} and 6.05 at.% Y^{3+}, and the Ca^{2+}/Sr^{2+} ratio was 1:1. Raman spectroscopy revealed the compositional disorder, by comparing the spectra of the parents (Yb:CaF_2, Yb:SrF_2) and singly-doped solid-solution (Yb:(Ca,Sr)F_2), Fig. 1(a). The energy of the Raman peak is

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300 cm^{-1} (the T_{2g} mode). The absorption and stimulated-emission cross-sections, σ_{abs} and σ_{SE}, respectively, for the $^2F_{7/2} \leftrightarrow {}^2F_{5/2}$ transition of Yb^{3+} ions were determined, Fig. 1(b). For the zero-phonon line used for pumping, σ_{abs} is 0.56×10^{-20} cm^2 at 975 nm (absorption bandwidth: 6.6 nm). At the expected laser wavelength, σ_{SE} amounts to 0.11×10^{-20} cm^2 at ~1040 nm. The luminescence decay of Yb^{3+} ions is singly exponential (suggesting a single type of rare-earth Yb-Y clusters) yielding a measured lifetime τ_{lum} of 2.76 ms, Fig. 1(c). The spectral gain profiles, $\sigma_{gain} = \beta\cdot\sigma_{SE} - (1-\beta)\,\sigma_{abs}$, were calculated for the quasi-3-level Yb-laser scheme, for various population inversion ratios β, Fig. 1(d). A special attention was paid to observing the long-wavelength phonon sideband of the emission profile extending up to 1.3 µm which contributes to the pulse shortening in Kerr-lens mode-locked oscillators. For $\beta = 0.10$, the gain bandwidth (FWHM) amounts to 36 nm. Low-temperature (LT, ~12 K) absorption and emission spectroscopy was employed to determine the crystal-field splitting for Yb^{3+} manifolds: $^2F_{7/2}$ = (0, 789, 444, 589) cm^{-1} and $^2F_{5/2}$ = (10252, 10348, 10833) cm^{-1}, Fig. 1(e,f).

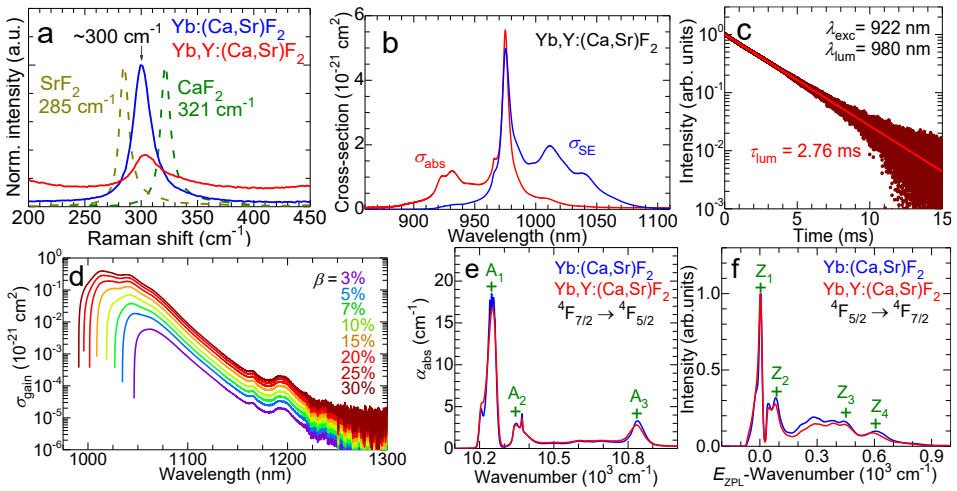

Fig. 1. Spectroscopy of multicomponent alkali earth metal fluoride Yb,Y:(Ca,Sr)F_2 crystal: (a) Raman spectrum; (b) absorption (σ_{abs}) and stimulated emission (σ_{SE}) cross-sections; (c) luminescence decay curve, τ_{lum} – luminescence lifetime; (d) gain cross-section (σ_{gain}) spectral profiles plotted in a semi-log scale; (e,f) low-temperature (LT, 12 K) absorption (e) and emission (f) spectra in Yb^{3+} ions, + mark the electronic transitions assigned to rare-earth cluster sites.

3 Conclusion

Yb^{3+}-doped multicomponent MF_2-type crystals (combining the solid-solution composition with "buffer" ion codoping) hold great promise for pulse shortening in 1-µm mode-locked lasers. They retain a consistent geometry of Yb^{3+} sites with respect to singly-doped crystals, while introducing an additional degree of structural disorder that contributes to extension of spectral bandwidths, potentially supporting laser gain up to ~1.2 µm.

References

1. Z. Zhang, Z.-Q. Li, P. Loiko, H.-J. Zeng, G. Zhang, Z.-L. Lin, S. Normani, A. Braud, F. Ma, X. Mateos, H.-C. Liang, V. Petrov, D. Jiang, L. Su, and W. Chen, Opt. Lett. **50**(6), 1835-1838 (2025).