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Ultraviolet and visible emissions of Er^{3+} in monoclinic $KYb(WO_4)_2$ single crystals

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Abstract

We observed ultraviolet and visible up-conversion signals with maximum intensity at 383 nm, 407 nm, 457 nm, 474 nm, 525 nm, 552 nm, 652 nm, 696 nm, 772 nm, 801 nm, 814 nm and 848 nm from the 4f states of erbium-doped $KYb(WO_4)_2$ single crystals after pumping at 981 nm (10 194 cm^{-1}) at room temperature (RT) and low temperature (10 K). These emissions were generated after simultaneous excitation of erbium and ytterbium at 981 nm (Yb^{3+} acts as a sensitizer of Er^{3+}). We discuss the up-conversion mechanism in which three and two photons were involved in the generation of the ultraviolet and visible emissions.
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1. Introduction

Up-conversion phenomena are used to develop short-wavelength solid-state lasers. These lasers have many technical applications including data storage, laser printing and full color laser display. Multiphoton processes have been observed in Er^{3+} because of its rich energy level scheme, leading to up-conversion emissions under different excitation wavelengths in many Er^{3+}-doped hosts such as fluorides [1], silicates [2], chlorides [3], oxides [4]. Up-conversion laser operation was observed in several hosts such as $LiYF_4$ (YLF) [5] and $Y_3Al_5O_{12}$ (YAG) [6]. The most common infrared-to-ultraviolet or visible conversion mechanisms are sequential three or two excitation photons (STEP), cooperative up-conversion and excited-state absorption (ESA) [7]. In the case of the Er^{3+}, Yb^{3+} pair, the conversion is due to STEP via cross-relaxation. Yb^{3+} ions are widely known as sensitizers of erbium emissions due to the high absorption cross-section and the possibility of energy transfer between ions via cross-relaxation.

Stoichiometric Yb^{3+} tungstate crystal, $KYb(WO_4)_2$ (hereafter KYbW) is a promising laser crystal and laser host material. The low temperature phase of $KYb(WO_4)_2$ is a biaxial crystal with inversion centre. The three orthogonal principal optical axes are labelled N_g, N_m and N_p. In monoclinic crystals, one of the principal axes (in this case N_p) is always parallel to the C_2 symmetry axis that coincides with the crystallographic *b* axis. The other two principal axes (in this case, N_g and N_m) therefore lie in the *a*–*c* plane because the *b* axis is orthogonal to this plane. Structural, crystal growth data, optical characterization and laser operation of KYbW are found in previous studies [8–10]. We also doped KYbW crystals with Er^{3+} ions at several dopant concentrations to characterize the potential laser transitions. Crystal growth data and optical characterization of the visible green and infrared 1.5 μm emissions of erbium in KYbW:Er have been published in [11,12]. The most important features of the KYbW host doped with Er^{3+} are, firstly, that the high quantity of Yb^{3+} ions also with the very large absorption cross-section can efficiently transfer the pump energy to Er^{3+} thus increasing the emission intensity in the UV–vis region, secondly, that due to the similar nature of the ions, concerning charge and size, it makes possible easy substitution in the host without introducing high changes in the crystal quality, and finally, that the transitions (absorption and emission) cross-sections in such

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tungstates are very large, so that the requirements of the pump beam quality is reduced, favoring the pump with diodes. The diodes based on InGaAs structures are very suitable to pump Yb-doped tungstates because of the match between the emission wavelength of the diode and the maximum of absorption of Yb (∼980 nm).

The aim of this paper is to characterize the emission erbium spectroscopy in the 300–870 nm (33 333–11 494 cm^{-1}) spectral range after sensitization of erbium in KYbW crystals. We propose an energy diagram for the up-conversion process related to the generation of the ultraviolet and visible emissions on the basis of the logarithmic relationship between the intensity of the emissions and the pump power. Finally, we measured the lifetimes of energy levels of erbium at room temperature (RT).

2. Experimental

For the up-conversion study, we used a pulsed BMI Optical Parametric Oscillator (OPO) pumped by the third harmonic of a seeded BMI SAGA YAG:Nd laser. Fluorescence was dispersed through a Jobin Yvon-Spex HR460 monochromator and detected by a Hamamatsu R928 photomultiplier. We analyzed the signal using a lock-in EG&G 7265DSP amplifier. Low-temperature luminescence was done at 10 K using an Oxford closed-cycle helium CCC1104 cryostat. We measured the lifetimes using the averaging facilities of a computer-controlled Tektronix digital TDS-714 oscilloscope.

The sample used in this study was high-optical-quality $KYb(WO_4)_2$ single crystal doped with erbium at 0.5 mol% of Er_2O_3 substituting Yb_2O_3 in the solution whose dimensions were 3 mm × 3 mm × 3.49 mm, for N_g, N_p and N_m principal optical axes, respectively. This concentration, measured by electron probe microanalysis (EPMA), was 3.9×10^{19} ions/cm^3 in the crystal. We used this concentration because it provided enough signal-to-noise ratio without saturating the detector. This crystal, among other erbium concentrations crystals was grown by the top-seeded-solution-growth method (TSSG) [11].

To prepare the samples for the spectroscopic studies, we cut the grown crystals parallel to their principal optical axes using a goniometer and a Struers Accutom-50 diamond saw and polished them with a Logytech PM5 polisher with an oscillatory arm that allowed us to accurately rotate and pressurise the samples depending on the hardness of the material.

3. Results and discussion

Fig. 1 shows the emission spectra obtained under excitation in the $^2F_{5/2}$ (Yb) and $^4I_{11/2}$ (Er) energy levels (981 nm). We analyzed the 300–870 nm (33 333–11 494 cm^{-1}) range and found that the highest intensity line corresponded to the transition from the $^4S_{3/2}$ level to the ground (emission with maximum intensity at 552 nm). We detected the other signals with maximum intensity at 383 nm, 407 nm, 457 nm, 474 nm, 525 nm, 652 nm, 696 nm, 772 nm, 801 nm, 814 nm and 848 nm at RT. In the spectrum the signals appear in arbitrary units and are rescaled. We assigned these emissions to different transitions (labels of Fig. 1) with the help of low-temperature emission spectra and agree well with the difference in energy between the Stark levels of each energy level. The maximum number of Stark levels is $(2J + 1)/2$, expected by the crystalline field, due to the odd number of electrons of Er^{3+}, and due to the low symmetry site in which Er^{3+} is located (C_2) in KYbW host.

Special care was taken to assign the signals around 474 nm because of the presence of thulium contamination as labelled in Fig. 1. Fig. 2 clarifies the overlapping between the emissions of

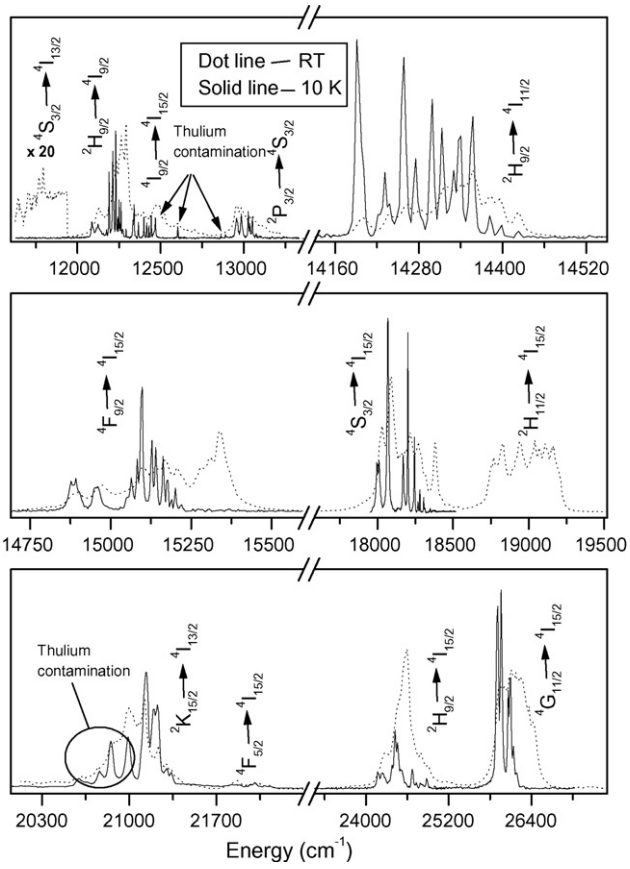

Fig. 1. Room temperature and 10 K emissions of erbium in KYbW in the 300–870 nm range (33 333–11 494 cm^{-1} range) after pumping at 981 nm. The scale is in arbitrary units and the spectra are rescaled.

the two ions at low temperature. The thulium signals arrive to 21 140 cm^{-1} (with maximum at 21 133 cm^{-1}) as mentioned in ref. [13] and the signal intensity at these energy value is lower than the rest of signals attributed to thulium. In our spectrum, the signal at 21 133 cm^{-1} is the most intense peak. Moreover, some other signals appeared at energies higher than 21 133 cm^{-1}, just up to 21 350 cm^{-1} which are, in principle, not attributed to thulium. These peaks are located at 21 133 cm^{-1}, 21 197 cm^{-1}, 21 220 cm^{-1}, 21 296 cm^{-1} and 21 336 cm^{-1} which are in absolute agreement with the difference between the Stark levels of $^2K_{15/2}$ and $^4I_{13/2}$ energy levels of erbium. Examples are: 27 735 cm^{-1} ($^2K_{15/2}$) and 6515 cm^{-1} ($^4I_{13/2}$) that generated the

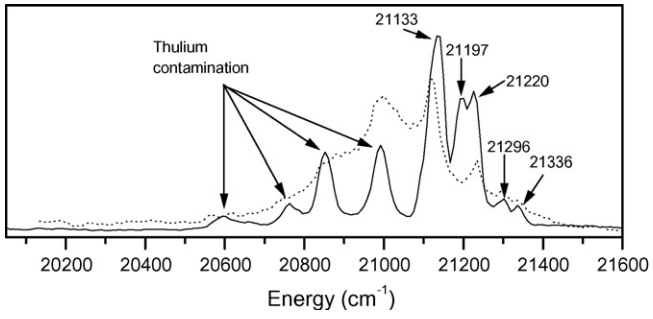

Fig. 2. Overlap between the $^1G_4 \rightarrow {}^3H_6$ transition of thulium and $^2K_{15/2} \rightarrow {}^4I_{13/2}$ transition of erbium and peaks assignment.

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