



Full length article

The color tunability and mechanism of upconversion emissions in Yb³⁺ and Er³⁺ co-doped Y₂Ce₂O₇ phosphors

Weixiong You^{a,*}, Jiapeng Li^a, Youfusheng Wu^a, Fengqin Lai^b, Xiufang Yang^a^a School of Material Science and Engineering, Jiangxi University of Science and Technology, Ganzhou 341000, PR China^b School of Mechanical Science and Electrical Engineering, Jiangxi University of Science and Technology, Ganzhou 341000, PR China

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ABSTRACT

Y₂Ce₂O₇ powders doped with different Yb³⁺ and Er³⁺ concentrations were synthesized via the sol-gel method. The structure and upconversion properties were investigated. The results indicated that a single phase was obtained and that the structure was cubic. The green and red upconversion emissions were detected with excitation at 980 nm, and the emission intensities could be tuned by the Yb³⁺ and Er³⁺ concentrations. The mechanism of the upconversion process was evaluated on the basis of the emission intensities and lifetimes of the ⁴S_{3/2}, ⁴F_{9/2} and ⁴I_{13/2} levels. The results demonstrated that cross-relaxation was responsible for the dependence of green and red upconversion emissions on the dopant concentration.

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1. Introduction

Upconversion is an anti-Stokes process in which a substance absorbs two or more low-energy photons and emits a higher-energy photon [1]. The upconversion of luminescent materials can be used in many fields, including optical temperature sensing, laser cooling and cell imaging [2–4]. Recently, explorations of the application of upconversion in solar cells and white-light emission have attracted much attention [5–7]. Lanthanide rare-earth (RE) ions are suitable candidates for the upconversion process because of their abundant energy levels and narrow emission spectral lines [8]; The Er³⁺ ion is the most efficient and popular RE ion for this application. To enhance the efficiency of upconversion emission of Er³⁺ ions, the Yb³⁺ ions are often co-doped into materials doped with Er³⁺ as a sensitizer. Furthermore, the fluorescent color of upconversion can be tuned by the concentration of the Yb³⁺ ion [9,10], which is useful for fluorescent labels or biological images.

Host materials with low phonon energy are necessary to achieve efficient upconversion emissions. Such materials enable the multi-phonon relaxation rate to be reduced and the lifetime of excited levels to be increased [11]. Fortunately, Re₂Ce₂O₇ with a pyrochlore structure or a defect fluorite-type structure has a low phonon energy (approximately 464 cm⁻¹ [12]) and excellent chemical stability [13] and is therefore a potential upconversion material. However, the literature contains few reports of the photoluminescence properties of RE-doped Re₂Ce₂O₇ compounds. In this paper, we focus on the upconversion properties of Y₂Ce₂O₇ phosphors co-doped with Yb³⁺ and Er³⁺. The mechanism of upconversion emission and the effects of the dopant concentrations on the color tunability are studied.

* Corresponding author.

E-mail address: you.wx@126.com (W. You).

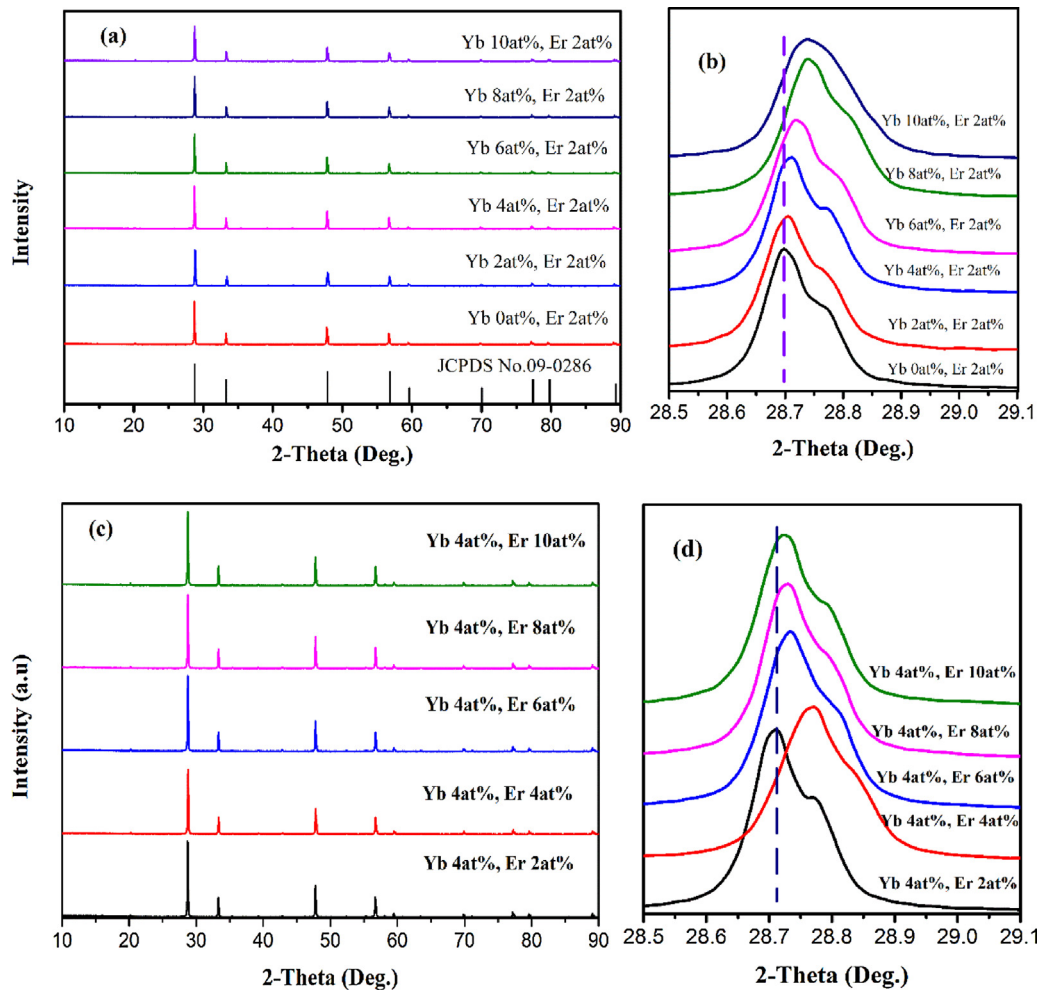


Fig. 1. (a) The X-ray diffraction patterns of $Y_2Ce_2O_7:xYb, 2at\%Er$ samples with different Yb^{3+} concentration; (b) (111) diffraction peaks of $Y_2Ce_2O_7:xYb, 2at\%Er$ samples with different Yb^{3+} concentration; (c) The X-ray diffraction patterns of $Y_2Ce_2O_7:4atYb, yEr$ samples with different Er^{3+} concentration; (d) (111) diffraction peaks of $Y_2Ce_2O_7:4at\%Yb, yEr$ samples with different Er^{3+} concentration;.

2. Experiment

$Y_2Ce_2O_7$ samples co-doped with Yb^{3+} and Er^{3+} were prepared by the sol-gel method. Y_2O_3 (99.99%), Yb_2O_3 (99.99%), Er_2O_3 (99.99%) and CeO_2 (99.99%) were used as starting materials. According to the formula, $(Yb_xEr_yY_{1-x-y})_2Ce_2O_7$ ($y = 0.02, x = 0, 0.02, 0.04, 0.06, 0.08, 0.1$ and $x = 0.04, y = 0.02, 0.04, 0.06, 0.08, 0.1$), stoichiometric amounts of the starting materials were dissolved in nitric acid, and citric acid was then added into the solution as the chelating agent. The molar ratio of metal ions and citric acid was 1:2.5. H_2O_2 was added when CeO_2 is dissolved using nitric acid. The mixture was stirred at approximately $90^\circ C$ for 4 h until a sol was formed. The sol was transformed into a sticky gel by evaporating the sol over several hours. The gel was dried at approximately $100^\circ C$ in air. After being fully ground, the dried powders were sintered at $1200^\circ C$ in a muffle furnace in air atmosphere for approximately 4 h, and white powder samples were obtained.

X-ray diffraction (XRD) patterns of the samples in the range of $10^\circ \leq 2\theta \leq 90^\circ$ were recorded on a Bruker D8 Advance X-ray diffractometer with high-intensity $CuK\alpha$ radiation ($\lambda = 1.54178 \text{ \AA}$). The upconversion emission spectra were detected using a spectrophotometer (FL980, Edinburgh) with a laser diode (LD) as the pump source; the excitement wavelength was 980 nm. The fluorescence decay curves, at a wavelength of 547, 677 and 1535 nm, were also excited at 980 nm by an optical parametric oscillator (OPO) in the same instrument. The signal was detected using an NIR PMT (R5509, Hamamatsu). All measurements were carried out at room temperature, and the spectra were corrected.

3. Results and discussion

Fig. 1(a)–(d) shows the XRD patterns of samples doped with different concentrations of Yb^{3+} and Er^{3+} . From the figure, the diffraction peaks of $Y_2Ce_2O_7$ agree with those of the standard card (JCPDS 09-0286), which indicates that the crystal

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