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Short communication

High-purity red up-conversion emission of $Ba_5Zn_4Y_8O_{21}$:Er^{3+},Yb^{3+} phosphor excited by 1550 nm laser diode

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ABSTRACT

A novel $Ba_5Zn_4Y_8O_{21}$:Er^{3+},Yb^{3+} up-conversion phosphor was synthesized via solid-state reaction method. The up-conversion luminescence properties of the phosphor excited by 1550 nm laser diode were investigated. Results showed that the characteristic emission peaks of Er^{3+} were located at 530, 550, 675, and 810 nm, which correspond to the transitions of $^2H_{11/2} \rightarrow {}^4I_{15/2}$, $^4S_{3/2} \rightarrow {}^4I_{15/2}$, $^4F_{9/2} \rightarrow {}^4I_{15/2}$, and $^4I_{9/2} \rightarrow {}^4I_{15/2}$, respectively. The up-conversion mechanisms were systematically studied through concentration and laser power dependence of the up-converted emissions. An evident tuning effect of Yb^{3+} on the luminescence of the phosphor was observed. An efficient red emission with high purity was obtained through the $Er^{3+} \rightarrow Yb^{3+} \rightarrow Er^{3+}$ energy transfer process, and the intensity ratio of the red and green emissions could reach 62.28. Therefore, $Ba_5Zn_4Y_8O_{21}$:Er^{3+},Yb^{3+} can be an excellent up-conversion red emitter.

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1. Introduction

Rare-earth-doped up-conversion luminescent (UCL) materials have received considerable attention because of their widespread applications in industrial fields, such as phosphors, up-conversion lasers, and solar cells [1–3]. These materials usually exhibit low luminescence efficiency because of their special transition mechanisms. Therefore, various methods have been attempted to obtain highly efficient UCL materials.

A host material has a significant effect on UCL efficiency. Fluoride with relatively low phonon energy (~355 cm^{-1}) can usually achieve high UCL efficiency, and this characteristic of fluoride has become a popular research topic in biological fluorescence labeling [3,4]. However, these UCL materials usually present poor chemical and thermal properties that limit their application. Oxide materials are usually stable chemically and thermally. Therefore, highly efficient and stable oxide materials can adequately meet the demand for the applications of solar cells and fluorescent powder. In recent years, a series of studies has been performed on the structural and optical properties of oxide UCL materials. In 2010, BaY_2ZnO_5:Er^{3+},Yb^{3+} and BaY_2ZnO_5:Ho^{3+},Yb^{3+} UCL materials with UCL yields of up to 5% and 2.6% respectively were prepared by Etchart and his coworkers [5,6]. Subsequently, the studies on efficient UCL of BaY_2ZnO_5, $BaGa_2ZnO_5$, $Ba_5Gd_8Zn_4O_{21}$, and other zinc acid salts were reported [7,8]. Aside from UCL efficiency, color purity is another factor affecting the application of UCL materials. A series of recent works focused on achieving materials with high-purity red, green, or blue up-conversion emission [9,10].

In the present work, a novel phosphor, namely, $Ba_5Zn_4Y_8O_{21}$:Er^{3+}/Yb^3, was synthesized via solid-state reaction method. Our previous studies confirmed that the $^4I_{13/2}$ level of Er^{3+} has a large absorption cross section (6×10^{-20} cm^{-2}) for ~1500 nm photons. Therefore, research on the UCL properties of the phosphors was performed using a 1550 nm (so-called eye-safe wavelength) laser diode (LD). The tuning effect of the Yb^{3+} ion on the UCL spectrum was also studied to obtain the high purity of red emission.

2. Experimental

$Ba_5Zn_4Y_8O_{21}$:Er^{3+}/Yb^{3+} phosphors were synthesized via the solid-state reaction method with starting materials of $BaCO_3$, ZnO, Y_2O_3, Er_2O_3, and Yb_2O_3. According to the stoichiometric ratio, the starting materials were weighed, mixed, and ground in an agate mortar for 30 min. The obtained mixtures were placed into crucibles and calcined at 1200 °C for 4 h at an ambient atmosphere. After the furnace naturally cooled to room temperature, the obtained phosphors were fully ground again for the subsequent crystal phase and UCL measurements. The doping concentrations

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in this experiment are a fixed Er^{3+} concentration of 5% and a Yb^{3+} concentration of 3%–20%. For comparison, the $NaYF_4$:Er^{3+},Yb^{3+} phosphor was synthesized using the method described in Ref. [11] which is generally accepted to provide the most efficient UCL.

A SHIMADZU X-ray diffractometer-6000 with Cu Kα radiation was used to analyze the phase of the samples. The tube voltage, tube electric current, and step velocity were 40 kV, 30 mA, and 4°/min, respectively. A Hitachi F-4500 fluorescence spectrophotometer equipped with 980 and 1550 nm LD was used to measure the UCL spectra. The test conditions were adjusted to be minimal, i.e., the emission slit and photoelectric multiplier voltage were 1 nm and 400 V respectively. The laser power was measured using a Beijing LPE-1A type power meter.

3. Results and discussion

Fig. 1 presents the X-ray diffraction (XRD) spectra of the phosphors calcined at 1200 °C for 4 h with fixed 5% Er^{3+} and different Yb^{3+} concentrations. Compared with those of standard PDF cards, the intensity and distribution of the diffraction peaks of the samples are similar to those of $Ba_5Zn_4Er_8O_{21}$ (JCPDS card number: #51-1687), and only a minimal amplitude of the peak shift (~0.2°) toward the small angle direction can be observed. The difference between the ionic radii of Y^{3+} (0.090 nm) and Er^{3+} (0.089 nm) is only 0.001 nm. Thus, we reasonably infer that the as-prepared samples should be $Ba_5Zn_4Y_8O_{21}$, and the aforementioned peak shift can be attributed to this small radius difference. The diffraction peaks are sharp, indicating that the pure $Ba_5Zn_4Er_8O_{21}$ with high crystallinity is synthesized.

Fig. 2 shows the UCL spectra of the as-prepared $Ba_5Zn_4Er_8O_{21}$:5%Er^{3+},x%Yb^{3+} phosphors excited by 1550 nm LD, with an excitation density of 416 mW/cm^2. Three characteristic emissions of Er^{3+} ion can be observed. The considerably strong red emission near 675 nm is generated by the $^4F_{9/2} \rightarrow {}^4I_{15/2}$ transition, and the extremely weak green emissions near 530–550 nm are generated by the $^2H_{11/2}/^4S_{3/2} \rightarrow {}^4I_{15/2}$ transitions. A weak near-infrared emission located at ~810 nm, which corresponds to the $^4I_{9/2} \rightarrow {}^4I_{15/2}$ transition, can also be observed. Compared with that of the Er^{3+} single-doped phosphor (Yb^{3+} content = 0), the green emission of the phosphor co-doped with Er^{3+} and Yb^{3+} ions is greatly suppressed. An obvious enhancement in the intensity of red emission of the phosphor is observed simultaneously. As a result, a strong and high-purity red emission is obtained. The green emission is completely quenched, particularly when the Yb^{3+} concentration is increased to 15%. The inset of Fig. 2 shows the intensity ratio of the red and green emissions (I_R/I_G) of the phosphor as a function of Yb^{3+} concentration. The I_R/I_G value sharply increases with Yb^{3+} concentration and reaches a maximum of 61.94 at 10 mol%.

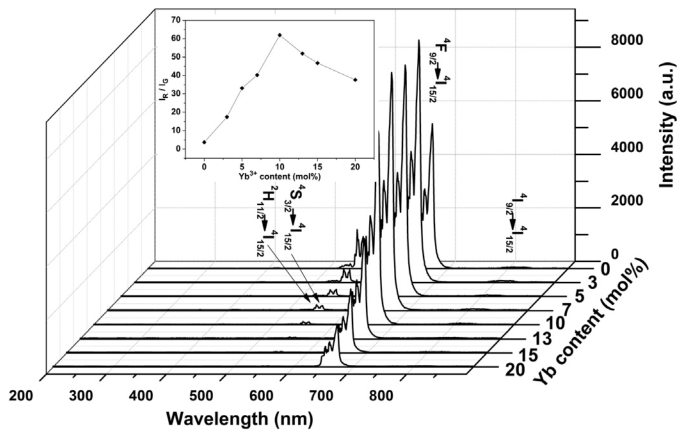

Fig. 2. Up-conversion spectra of the $Ba_5Zn_4Y_8O_{21}$: Er^{3+},Yb^{3+} phosphor with fixed 5% Er^{3+} and different Yb^{3+} concentrations excited by 1550 nm LD. (inset) The I_R/I_G value of the phosphor as a function of Yb^{3+} concentration (power density: 416 mW/cm^2; photomultiplier voltage: 400 V).

The $NaYF_4$:Er^{3+},Yb^{3+} phosphor with optimum doping concentration is prepared via the solid-state reaction method to characterize the UCL property of the as-prepared $Ba_5Zn_4Y_8O_{21}$ phosphor. The UCL spectrum of the phosphor excited by 980 nm LD is used as a reference, as shown in Fig. 3. The red emission intensity of $Ba_5Zn_4Y_8O_{21}$:5%Er^{3+},3%Yb^{3+} phosphor excited by 1550 nm LD is the same as the green emission intensity of $NaYF_4$ excited by 980 nm LD under the same excitation power density (416 mW/cm^2). The dependence of the UCL emissions of $Ba_5Zn_4Y_8O_{21}$:5%Er^{3+}, 20%Yb^{3+} phosphor on the laser power density is also evaluated to further characterize the laser power dependence of the color purity (see the inset of Fig. 3). Despite the increase of power

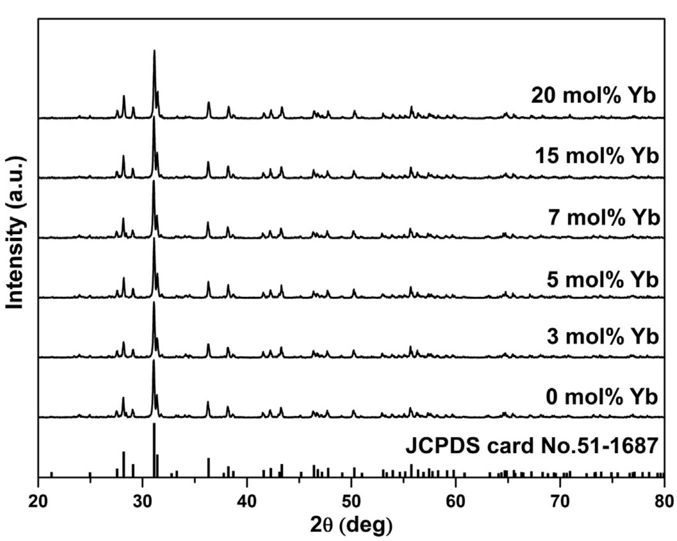

Fig. 1. XRD spectra of $Ba_5Zn_4Y_8O_{21}$:5%Er^{3+}, *x*%Yb^{3+} phosphor annealed at 1200 °C for 4 h.

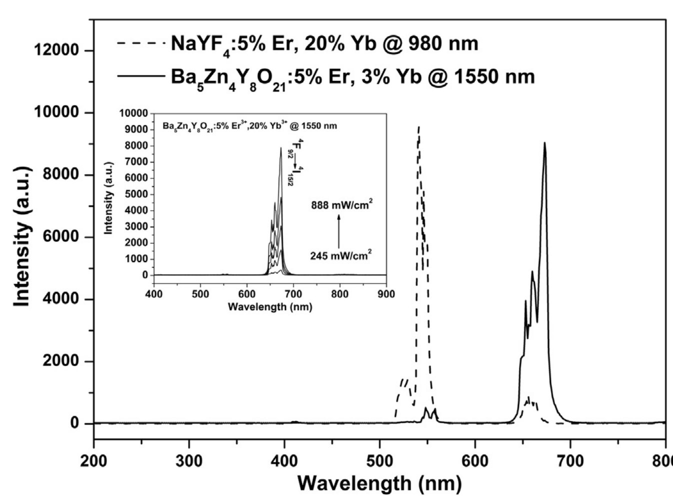

Fig. 3. Up-conversion spectra of $Ba_5Zn_4Y_8O_{21}$: 5%Er^{3+},3%Yb^{3+} phosphor excited by 1550 nm LD and $NaYF_4$: 5%Er^{3+},20%Yb^{3+} phosphor excited by 980 nm LD (power density: 416 mW/cm^2). (inset) Up-conversion spectra of $Ba_5Zn_4Y_8O_{21}$:5%Er^{3+},20% Yb^{3+} phosphor excited by 1550 nm LD with different power densities.

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