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Preparation and infrared to visible upconversion luminescence of Yb_2O_3:Ho^{3+} nanocrystalline powders

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ABSTRACT

Yb_2O_3:Ho^{3+} nanocrystalline powders were synthesized through a solid state reaction method. X-ray diffraction analysis and field emission scanning electron microscopy were used to analyze the phase composition and morphology of the powders. Then under the 980 nm excitation of laser diode, the fluorescence of the crystals was studied via a fluorescence spectrometer. The green and red emissions centered on 551 and 668 nm were observed, and the green band dominated the emission spectrum. The effect of the concentration of Ho^{3+} on the upconversion luminescence intensity was discussed and the possible upconversion emission mechanism was explained. It indicates that like other metal oxide nanoparticles, Yb_2O_3 could also be a potential host material for doping to prepare the upconversion phosphor.

1. Introduction

The upconversion is a process that can emit higher energy photons after absorbing lower energy excitation photons via multiple absorption or energy transfer [1]. After the discovery of upconversion process in 1960s, over the past few decades, upconversion materials have been the focus of much research, mainly for their potential applications including laser [2], bio-imaging [3], novel displays [4], and solar cells [5]. The lanthanide is often used as dopant to research the upconversion. Part of upconversion studies have been concentrated on semiconductor such as Bi_2MoO_4 [6], In_2O_3 [7], ZnO [8], and TiO_2 [9]. However, there has also been a considerable amount of investigations on doped insulators, including halides [10,16], glass [11,12], and some oxides. Among the different types of upconversion host materials, oxides gradually become research focus due to their high chemical stability and low thermal expansion, although the phonon energy of them is slightly higher. Particularly, some rare earth oxides, such as Y_2O_3 [13], Lu_2O_3 [14], and Gd_2O_3 [15] have been selected as host materials for doping to investigate the upconversion.

It is well known that trivalent rare-earth ions such as Er^{3+}, Ho^{3+}, Pr^{3+} and Tm^{3+} are usually doped in host materials serving as activator ions for upconversion luminescence, because these ions possess affluent energy levels for radiative transition. However, Er^{3+} and Ho^{3+} have a small absorption cross-section near 980 nm, so they are often used together with Yb^{3+}, which has large absorption cross-section around 980 nm and commonly acts as sensitizer to promote efficient energy transfer [16,17]. Nevertheless, to our knowledge, there is no report about Yb_2O_3 served as host material doped with Er^{3+}/Ho^{3+} in order to study the upconversion properties.

In this paper, solid state reaction method was adopted to synthesize the Yb_2O_3:Ho^{3+} nanocrystalline powders. Then we investigated the upconversion luminescence at 980 nm excitation, and the possible mechanisms of the process were analyzed.

2. Experimental section

All reagents used are of analytical grade and used without any further purification. Ho doped Yb_2O_3 nanopowders were synthesized by high-temperature solid state reaction method. In a typical experiment, initially, appropriate amount of Ho_2O_3 powders were dispersed in 6 ml absolute ethyl alcohol in a beaker for 10 min with the aid of ultrasonic. After that, the suspension ethanol liquid was decanted into an agate mortar with 0.316 g Yb_2O_3. Then the mixture was ground for about 3 h. Afterwards, the Ho_2O_3-Yb_2O_3 powders were annealed in air at 1250 °C for 4.5 h in an alumina crucible. Thus, Yb_2O_3:Ho^{3+} crystalline powders were obtained. Yb_2O_3 doped with 1 wt%, 3 wt%, 5 wt%, 10 wt%, and 19 wt% holmium oxide nanocrystals were labeled as S1, S2, S3, S4, and S5 in sequence.

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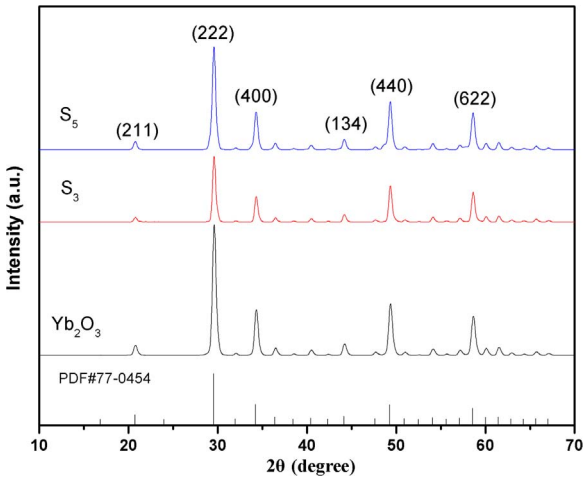

Fig. 1. XRD patterns of Yb_2O_3, S3, and S5.

Table 1
Variation of FWMH and 2θ for some crystal planes of different samples.

Samples	FWHM of (222) (deg)	2θ (deg)	(222)	(440)	(622)
Yb_2O_3	0.4661	–	29.6338	49.3637	58.6374
S3	0.3964	–	29.6162	49.3428	58.6100
S5	0.4310	–	29.5997	49.3278	58.5956

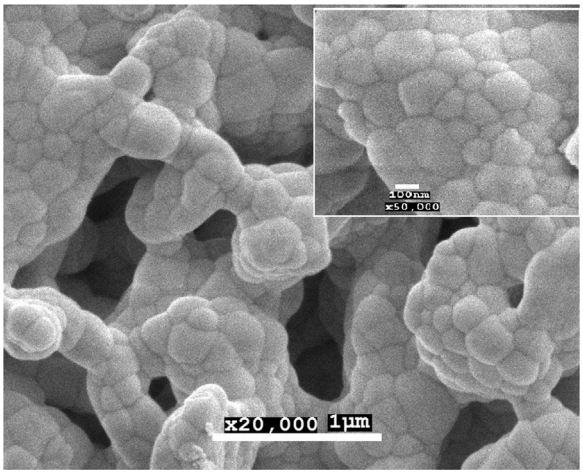

Fig. 2. FESEM image of S3 with Ho_2O_3 of 5 wt%.

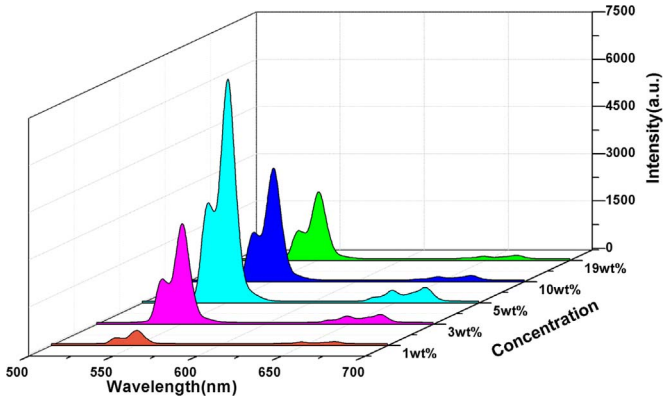

Fig. 3. Emission spectra of Yb_2O_3:Ho^{3+} with different concentrations of Ho_2O_3.

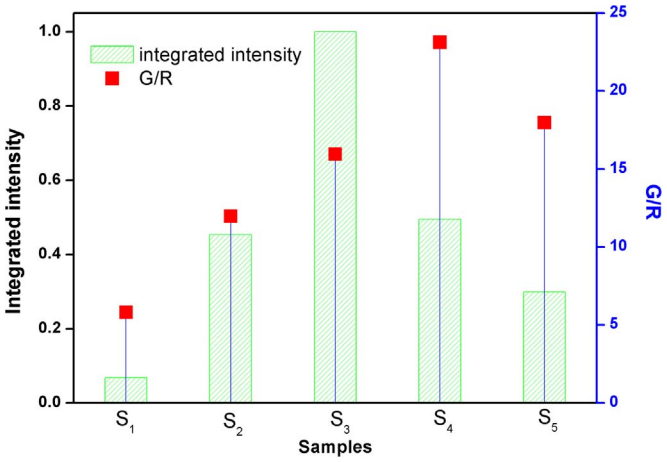

Fig. 4. Integrated upconversion intensity and green/red ratio as a function of the Ho^{3+}(or Yb^{3+}) ions concentrations.

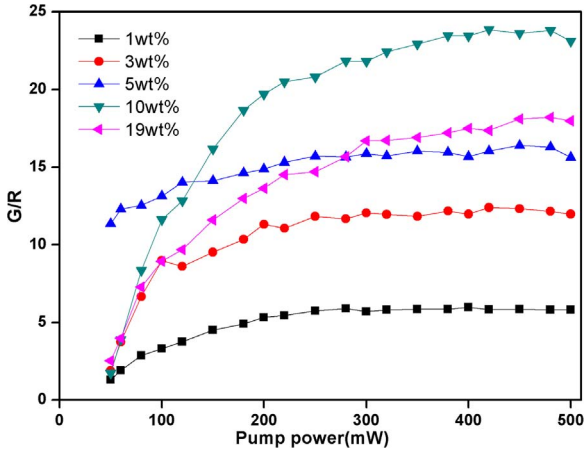

Fig. 5. Power dependence of the green/red emissions ratio.

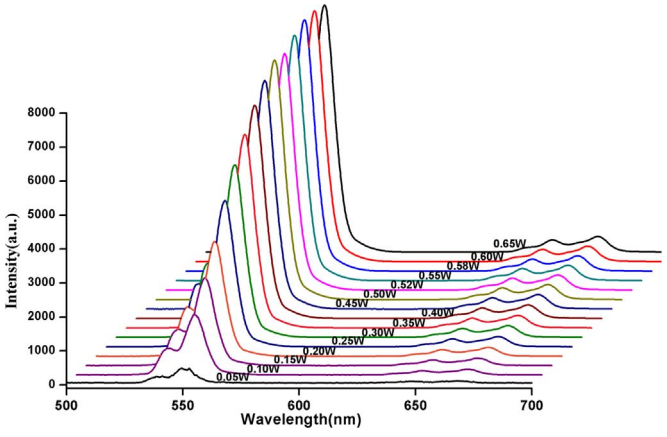

Fig. 6. The fluorescence intensity of S3 under diverse pump power.

The phase composition of the powders was identified by X-ray diffraction analysis (XRD-7000, SHIMADZU, Japan) with Cu K_α radiation (λ=0.15406 nm, 40 kV, 20 mA). The morphologies and particle size were observed by a field-emission scanning electron microscope (FE-SEM, JSM-7800F, JEOL, Japan). The as-prepared samples were filled into a sample cell with a diameter of 10 mm and a thickness of 2 mm. Under the excitation by a continuous wave diode laser tuned to 980 nm, the up-conversion luminescence spectra were recorded by a fluorescence spectrometer (F-7000, Hitachi, Japan) controlled with a PC at room temperature. The different characterized