

## White-light generation and full-color in single-phase garnet-based phosphors



Mengmeng Shang<sup>a,\*</sup>, Jian Fan<sup>c</sup>, Yang Zhang<sup>a,b</sup>, Hongzhou Lian<sup>a</sup>, Jun Lin<sup>a,\*</sup>

<sup>a</sup> State Key Laboratory of Rare Earth Resource Utilization, Changchun Institute of Applied Chemistry, Chinese Academy of Sciences, Changchun 130022, PR China

<sup>b</sup> University of Chinese Academy of Sciences, Beijing 100049, PR China

<sup>c</sup> College of Chemical and Environmental Engineering, Shandong University of Science and Technology, Qingdao 266590, PR China

### ARTICLE INFO

#### Article history:

Received 18 November 2014

Received in revised form 18 December 2014

Accepted 2 January 2015

Available online 6 January 2015

#### Keywords:

Garnet structure

Single-phase phosphors

White light emission

Energy transfer

### ABSTRACT

Presently considerable interest in single-phase white-emitting for ultraviolet light-emitting diodes is stimulated. Here we report the effective single-phase phosphors based on YAG-garnet structure, which exhibit varied hues from blue through white and eventually orange by tuning the relative proportion of  $\text{Ce}^{3+}/\text{Mn}^{2+}$  excited at ultraviolet.

© 2015 Elsevier B.V. All rights reserved.

Currently, a typically blue LED with YAG: $\text{Ce}^{3+}$  yellow phosphor is considered undesirable for indoor use, as white light achieved in such WLEDs has a high correlated color temperature (CCT) greater than ~4000 K, and low color rendering index (CRI) value of 70–80 [1–5]. Due to this problem, ultraviolet (UV) LED (300–410 nm) chip coupled with phosphors were introduced and have been rapidly developed in the past decade. Due to their superior color uniformity with tunable CCT, high CRI, and tunable CIE chromaticity coordinates, these white LEDs have potential application in the back light field [6–12]. However, white LEDs with multiple emitting components can be problematic as the device is very complicated, and the color balance is difficult to control. Therefore, the development of single-phase white-emitting phosphors that can solve the reabsorption problems existing in multi-component phosphors is a very important prospect that requires prompt attention [13].

White light emission obtained by using the principle based on the energy transfer from sensitizer to activator in a single-phase host is one effective strategy and significant achievements have been made [14–18]. In our previous work, solid solution phosphors  $\text{Y}_3\text{Al}_5 - 2x\text{Mg}_x\text{Si}_x\text{O}_{12}:\text{Ce}^{3+}$  ( $\text{Mg}^{2+}$ – $\text{Si}^{4+}$  replacing  $\text{Al}_{(1)}^{3+}$ – $\text{Al}_{(2)}^{3+}$  in garnet- $\text{Y}_3\text{Al}_5\text{O}_{12}$  host lattice on octahedron and tetrahedral sites) not only show an obvious red shift under the excitation of blue light compared with YAG: $\text{Ce}^{3+}$  yellow phosphor, but also give an additional emission band in the range of 350–500 nm excited with UV light [19]. In this paper, we choose the hosts with the composition of  $\text{Y}_3\text{Al}_3\text{MgSiO}_{12}$  and  $\text{Y}_3\text{AlMg}_2\text{Si}_2\text{O}_{12}$  (viz.  $x = 1$  and 2,

which is abbreviated to 311-YAMSO and 122-YAMSO) as the research object and create color-tunable warm white light emission via energy transfer between  $\text{Ce}^{3+}$  and  $\text{Mn}^{2+}$ .

The 311/122-YAMSO: $\text{Ce}^{3+}$ ,  $\text{Mn}^{2+}$  samples were prepared by high-temperature solid-state reaction. The doping concentrations of  $\text{Ce}^{3+}$  and  $\text{Mn}^{2+}$  are 0.3–3 mol% of  $\text{Y}^{3+}$  and 0–30 mol% of  $\text{Mg}^{2+}$  in 311/122-YAMSO, respectively. Measurements on the power X-ray diffraction for all samples were performed to verify the phase purity and all the diffraction peaks can be well indexed to the standard data of  $\text{Y}_3\text{Al}_5\text{O}_{12}$  (JCPDS No. 78-1891). The XRD patterns of all samples are the same as each other and hence only part of the results is given in Fig. S1 (Supporting information), indicating that the 311/122-type YAMSO solid solutions with garnet structure have formed and the substitution of doping ions does not significantly influence the crystal structure.

Fig. 1 displays the crystal structure and the photoluminescence excitation (PLE) and emission (PL) spectra of  $\text{Ce}^{3+}$  singly doped 311/122-YAMSO powder. The crystal structure of garnet for 311/122-YAMSO is cubic in space group  $Ia\bar{3}d$ , which has been validated by our previous study. In the 311/122-YAMSO solid solutions, doping ions  $\text{Ce}^{3+}$  and  $\text{Mn}^{2+}$  would substitute for  $\text{Y}^{3+}$  and  $\text{Mg}^{2+}$  sites with dodecahedral and octahedral coordination, respectively. As shown in Fig. 1, the luminescence spectra of 311/122-YAMSO: $\text{Ce}^{3+}$  excited with UV irradiation at room temperature exhibit two broad emission band in the range of 350–480 nm and 480–650 nm, respectively. It is well known that there is only one luminescence center in YAG: $\text{Ce}^{3+}$  phosphor and the additional emission band locating at higher energy has been demonstrated to be an additional site occupied by  $\text{Ce}^{3+}$  introduced by the incorporation of  $\text{Mg}^{2+}$ – $\text{Si}^{4+}$  ion pairs into YAG crystal structure. In

\* Corresponding authors.

E-mail addresses: [mmshang@ciac.ac.cn](mailto:mmshang@ciac.ac.cn) (M. Shang), [jlin@ciac.ac.cn](mailto:jlin@ciac.ac.cn) (J. Lin).

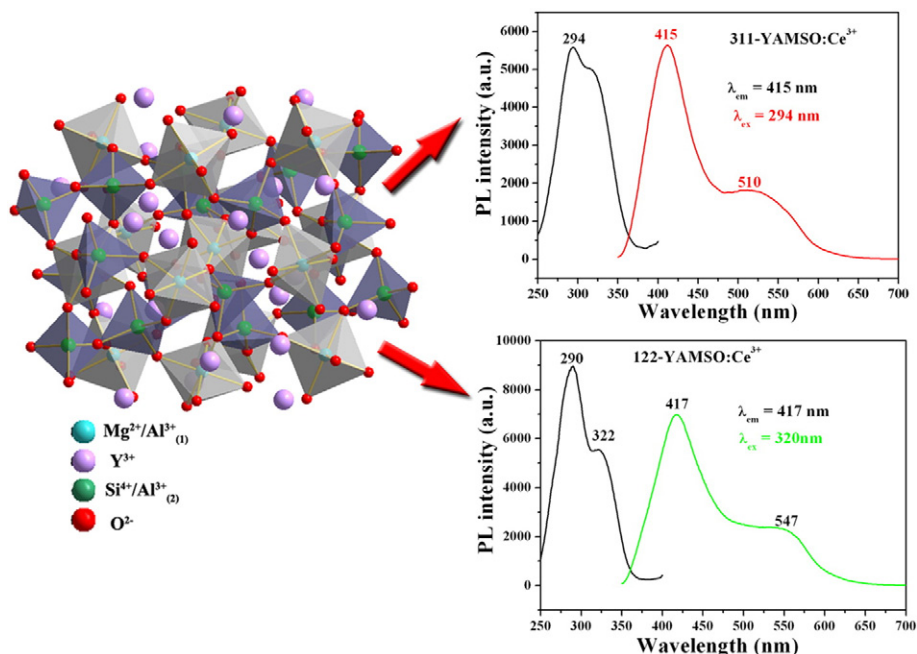


Fig. 1. The crystal structure and the PLE and PL spectra of  $\text{Ce}^{3+}$  singly doped 311-YAMSO and 122-YAMSO garnet powders.

our experiment, the optimum doping concentration for  $\text{Ce}^{3+}$  is about 2 mol% in the 311/122-YAMSO host and the CIE chromaticity coordinates calculated from the PL spectra locate at the blue region.

As we know,  $\text{Mn}^{2+}$  ion activated phosphors can give an orange-red emission when  $\text{Mn}^{2+}$  ions lie in octahedral surrounding, but the emission intensity and luminescence efficiency are weaker due to the forbidden d–d transition [20]. Moreover,  $\text{Ce}^{3+}$  ion is a good sensitizer for  $\text{Mn}^{2+}$  and the effective energy transfer between them can tune the luminescence color [21]. Fig. 2 presents the PL spectra and schematic illustration of the energy transfer process of 311/122-YAMSO: $\text{Ce}^{3+}$ ,

$\text{Mn}^{2+}$  phosphors. As shown in Fig. 2a and c, 311/122-YAMSO: $\text{Mn}^{2+}$  exhibit one emission band at  $\sim 580$  nm, which corresponds to the  ${}^4\text{T}_1({}^4\text{G}) \rightarrow {}^6\text{A}_1({}^6\text{S})$  transition of  $\text{Mn}^{2+}$ . In addition, the excitation spectra of  $\text{Mn}^{2+}$  are well overlapped with the emission spectra of  $\text{Ce}^{3+}$ . So there would be an effective energy transfer in  $\text{Ce}^{3+}$  and  $\text{Mn}^{2+}$  co-doped 311/122-YAMSO phosphors. The PL spectra (Fig. 2d and e) of 311/122-YAMSO: $\text{Ce}^{3+}$ ,  $\text{Mn}^{2+}$  show two emission bands situated at 420 and 580 nm, which are attributed to the emission from  $\text{Ce}^{3+}$  and  $\text{Mn}^{2+}$ , respectively. Compared with  $\text{Mn}^{2+}$  singly doped samples, the emission band of  $\text{Mn}^{2+}$  emits efficiently by introducing the

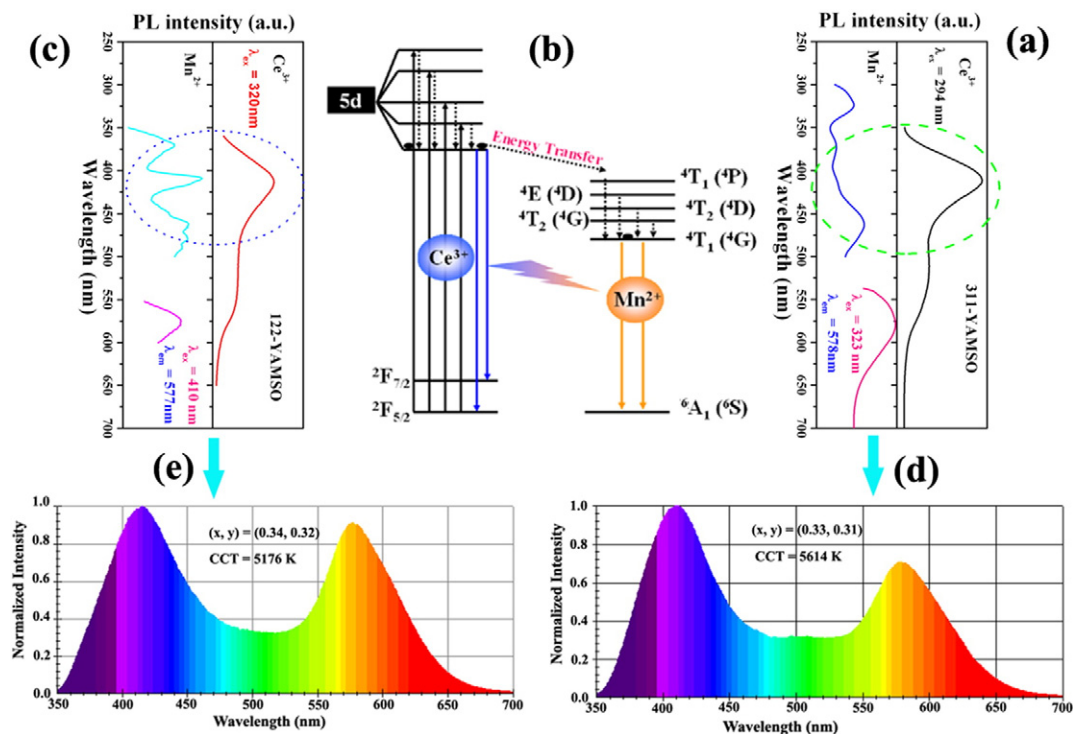


Fig. 2. The PL and PLE spectra of  $\text{Ce}^{3+}$  and  $\text{Mn}^{2+}$  singly doped 311-YAMSO (a) and 122-YAMSO (c) powders; (b) schematic illustration of energy transfer process for  $\text{Ce}^{3+}$  and  $\text{Mn}^{2+}$  co-doped 311/122-YAMSO samples; (d, e) PL spectra of 311-YAMSO: $\text{Ce}^{3+}$ ,  $\text{Mn}^{2+}$  ( $\lambda_{\text{ex}} = 294$  nm) and 122-YAMSO: $\text{Ce}^{3+}$ ,  $\text{Mn}^{2+}$  samples ( $\lambda_{\text{ex}} = 327$  nm).

Download English Version:

<https://daneshyari.com/en/article/1305489>

Download Persian Version:

<https://daneshyari.com/article/1305489>

[Daneshyari.com](https://daneshyari.com)