



Short Communication

Inducing tunable host luminescence in Zn_2GeO_4 tetrahedral materials via doping Cr^{3+} Qiongyu Bai^{a,b}, Panlai Li^{c,*}, Zhijun Wang^c, Shuchao Xu^c, Ting Li^c, Zhiping Yang^c, Zheng Xu^{a,b}^a Key Laboratory of Luminescence and Optical Information (Beijing Jiaotong University), Ministry of Education, Beijing 100044, China^b Institute of Optoelectronics Technology, Beijing Jiaotong University, Beijing 100044, China^c College of Physics Science & Technology, Hebei Key Lab of Optic-Electronic Information and Materials, Hebei University, Baoding 071002, China

ARTICLE INFO

Article history:

Received 16 August 2017

Received in revised form 18 March 2018

Accepted 23 March 2018

Available online 26 March 2018

Keywords:

Luminescence

Phosphors

Defect

 Cr^{3+}

ABSTRACT

Zn_2GeO_4 consisting of tetrahedron, and it is a self-luminescent material due to the presence of the native defects and shows a bluish white emission excited by ultraviolet. Although Cr^{3+} doped in a tetrahedron generally cannot show luminescence, in this research, new defects are formed as Cr^{3+} doped in Zn_2GeO_4 , hence a green emission band can be obtained. Meanwhile, the intensity of host emission is also decreased. Therefore, $\text{Zn}_2\text{GeO}_4:\text{Cr}^{3+}$ are synthesized using a high-temperature solid-phase method. Thermoluminescence (TL) and luminescence decay curves are used to investigate the variation of native defects. The emission colour can be tuned from bluish white to green when Cr^{3+} doped in Zn_2GeO_4 . This result has guidance for controlling the native emission of self-luminescent material.

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1. Introduction

Zn_2GeO_4 has a wide-band gap (4.68 eV), herein Zn^{2+} and Ge^{4+} coordinates with four O^{2-} combine to form tetrahedral. The ZnO_4 and GeO_4 tetrahedral connect with each other by means of corner sharing. Rare earth ions or transition metal ions doped Zn_2GeO_4 present excellent luminescent properties [1–10]. For example, $\text{Zn}_2\text{GeO}_4:\text{Eu}^{3+}$ nanocrystal can show multi-colour, which can be used for white light-emitting diodes [11]. Zinc germanate doped with manganese ($\text{Zn}_2\text{GeO}_4:\text{Mn}^{2+}$) is a native defect material which has been widely investigated as a new green phosphor for field emission displays (FEDs) [12]. Table 1 shows the luminescence properties of various activators doped Zn_2GeO_4 [8–12], in which host shows an excellent emission. Zn_2GeO_4 as a self-luminescent material has been widely research due to many advantages, such as energy-saving and convenient scheme [13]. Many strategies are researched to control the native defects, further control the luminescence. The most common of these is changing the sintering atmosphere. Liu et al. controlled the number of native defects in different atmosphere [3]. However, the experimental conditions of this method are difficult to control. Therefore, it is necessary to research a method to control the luminescence of Zn_2GeO_4 .

In our previous study, the native defects can be controlled by the rare earth ions (RE) substituting for the Zn^{2+} or Ge^{4+} in Zn_2GeO_4 [14,15]. The native defects are greater declined as RE substitute Ge^{4+} in

$\text{Zn}_2\text{GeO}_4:\text{RE}$. Take Ce^{3+} doped Zn_2GeO_4 as an example, the quenching concentration of host emission for $\text{Zn}_2\text{Ge}_{1-x}\text{O}_4:x\text{Ce}^{3+}$ ($x = 0.01$) is smaller than that of $\text{Zn}_2-x\text{GeO}_4:x\text{Ce}^{3+}$ ($x = 0.02$), which may be related to the defect types produced by substitution in Zn_2GeO_4 host.

In recent years, rare earth or transition metal ions doped phosphors, especially Cr^{3+} ions, have received much attention because of their excellent luminescent properties [16–18]. Generally, phosphor shows red or near infrared emission as Cr^{3+} occupied an octahedral site, which may be applied to the vivo biological imaging [19–23]. $\text{Zn}_3\text{Ga}_2\text{Sn}_1\text{O}_8:\text{Cr}^{3+}$ exhibits a bright near infrared emission at 696 nm, and it shows a long persistent luminescence over 300 h [24]. Jin et al. reported that Cr^{3+} -doped $\text{Mg}_4\text{Ga}_8\text{Ge}_2\text{O}_{20}$ shows a near infrared long persistent luminescence, which can be used in vivo bio-imaging [25]. However, if Cr^{3+} ion occupied a tetrahedral, it would not present photoluminescence properties. Therefore, Cr^{3+} ion is selected to control the native defects of Zn_2GeO_4 , due to that there is no characteristic emission of Cr^{3+} to affect the host emission. Because there are two tetrahedrons in this host, transition metal Cr^{3+} ions doped $\text{Zn}_2-x\text{GeO}_4:\text{Cr}^{3+}$ and $\text{Zn}_2\text{Ge}_{1-x}\text{O}_4:\text{Cr}^{3+}$ are synthesized, and the effect of doping ions on native emission is investigated. A green emission is observed due to the obvious variations of traps when Cr^{3+} ions doped in Zn_2GeO_4 .

2. Experimental

2.1. Sample Preparation

A series of phosphors with composition $\text{Zn}_{2-x}\text{GeO}_4:x\text{Cr}^{3+}$ and $\text{Zn}_2\text{Ge}_{1-x}\text{O}_4:x\text{Cr}^{3+}$ were synthesized by a high-temperature solid-

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Table 1
Host and activators luminescence of different activators in Zn_2GeO_4 .

Sample	Host		Activators		
	Excitation (Ex) (nm)	Emission (Em) (nm)	Excitation (Ex) (nm)	Emission (Em) (nm)	Ex peak (nm)
$\text{Zn}_2\text{GeO}_4:\text{Bi}^{3+}$ [9]	200–300	350–600	200–350	350–600	265
$\text{Zn}_2\text{GeO}_4:\text{Eu}^{3+}$ [7]	250–300	400–650	240–320	575–700	265
$(\text{ZnMg})_2\text{GeO}_4:\text{Mn}^{2+}$ [8]	200–300	350–600	200–300	500–600	243
$\text{Zn}_2\text{GeO}_4:\text{Tb}^{3+}, \text{Eu}^{3+}$ [10]	200–300	350–600	200–550	350–650	270
$\text{Zn}_2\text{GeO}_4:\text{Ce}^{3+}$ [11]	200–300	350–600	200–350	350–700	270

state method. The starting materials, including reagent grade ZnO , GeO_2 (99.99%), Cr_2O_3 (99.99%), were weighed in stoichiometric proportion, mixed homogeneously and ground by an agate mortar. The resultant

mixtures were transferred into a furnace and heated up to 850°C for 2 h in air, and then cooled down to room temperature naturally with the furnace. After intermediately ground to improve the homogeneity,

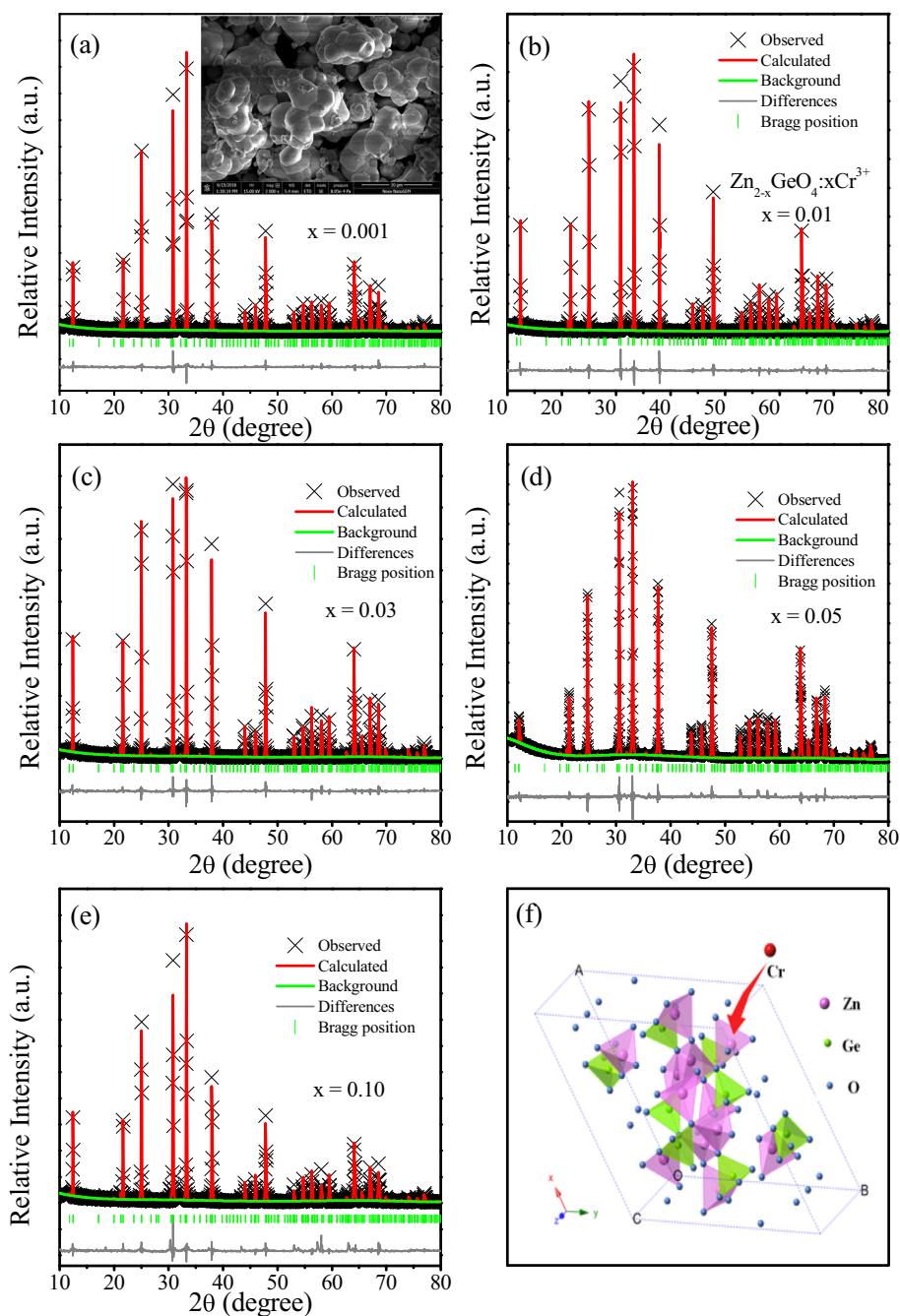


Fig. 1. (a–e) XRD profiles for Rietveld refinement results of $\text{Zn}_{2-x}\text{GeO}_4:x\text{Cr}^{3+}$ ($x = 0.001, 0.01, 0.03, 0.05, 0.10$) and SEM of $\text{Zn}_2\text{GeO}_4:0.001\text{Cr}^{3+}$. (f) The crystal structure of Zn_2GeO_4 .

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