



Tunable emission color of $Gd_2(MoO_4)_3:Yb^{3+}$, Ho^{3+} , Tm^{3+} phosphors via different excitation condition



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ABSTRACT

The methods of modulating emitting color are greatly important for up-conversion phosphors to broaden their applications, ranging from information storage to surface display. In this paper, the $Ho^{3+}/Tm^{3+}/Yb^{3+}$ tri-doped $Gd_2(MoO_4)_3$ phosphors are synthesized by typical sol-gel method. It is found that the phosphors can radiate intense blue (475 nm), green (542 nm) and red (660 nm) up-conversion luminescence under 980 nm or 915 nm laser excitation. Especially, the emission color of the phosphors can be tuned from red to lavender by adjusting the excitation power of 980 nm or 915 nm continuous lasers. In addition, the up-conversion luminescence is also investigated under the excitation of 980 nm pulse laser. The emission color varies from green to blue and green to yellow by altering the pulse width and frequency, respectively. Combining the above methods, multi-color and white light are realized, indicating the potential application in dynamic colorful display.

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1. Introduction

Lanthanide ion (Ln^{3+}) doped materials, converting near-infrared light into visible and ultraviolet light, have a wide range of applications, such as photo-catalysis [1], lightings [2], solar energy conversion [3], vitro/vivo imaging [4,5]. In recent years, Ln^{3+} doped materials have been extensively explored in three-dimensional displays [6,7] and colorful displays [8]. Many efforts have been made to modify the color of rare earth ion doped phosphors. Most researchers adjust the color of the up-conversion phosphor through changing the doped rare earth ion, basing on different rare earth ions emitting different color [9–13]. For example, Eu^{3+} is used to provide red emission [11]. A number of researchers tune the color of up-conversion emission by varying the ratio of co-doped rare earth ions [14–17]. Some researchers utilize the energy transfer between the rare earth ion and metal to change the color of up-conversion emission [18–20]. However, these methods of color modulation are realized through changing samples, which limits the applications of Ln^{3+} doped materials. To overcome this

drawback, various strategies have been utilized, such as changing the excitation wavelength [21] and excitation power [22]. Apart from that, Liu's group realized dynamic Commission International de l'Eclairage (CIE) coordinates modulation in $NaYF_4$ -based core-shell nanocrystals through adjusting the pulse width of 980 pulse laser [23]. Here, we offer pulse-width and pulse-frequency-based approaches to control the emission color of $Gd_2(MoO_4)_3:Yb^{3+}$, Ho^{3+} , Tm^{3+} phosphors.

$Gd_2(MoO_4)_3$ is one of the most popular hosts in Ln^{3+} doped materials because of its excellent characteristics, such as low toxicity, thermal stability and high photo-chemical stability [24–26]. Rare earth ions doped $Gd_2(MoO_4)_3$ materials have been extensively investigated in solar cell [27], color emitting device [28] and optical temperature measurement [29]. According to above background, $Gd_2(MoO_4)_3$ is chosen as the host in this work. Tm^{3+} and Ho^{3+} , which provide blue, green and red emission, are chosen as the activators [30]. Yb^{3+} acts as sensitizer in the phosphors, which has a large absorption cross section in the range of 910–980 nm [31].

In this work, we provide two efficient methods to modify the up-conversion luminescence of $Gd_2(MoO_4)_3:Yb^{3+}$, Ho^{3+} , Tm^{3+} phosphors: tuning the power of continuous lasers and changing the pulse width and frequency of pulse laser. These investigations promote the color tunability of the samples, which have great potential in dynamic color display.

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2. Experimental details

2.1. Preparation of $Gd_2(MoO_4)_3:Yb^{3+}, Ho^{3+}, Tm^{3+}$

All chemicals used in the experiments were analytical grade reagents and used without further purification. In this experiment, mole ratio of $Gd^{3+}, Yb^{3+}, Ho^{3+}, Tm^{3+}$ was 83/15/1/1 mol.%. The preparation of the precursor of $Gd_2(MoO_4)_3:Yb^{3+}, Ho^{3+}, Tm^{3+}$ was as follows. Firstly, calculated amounts of $Gd(NO_3)_3 \cdot 6H_2O, Yb(NO_3)_3 \cdot 6H_2O, Ho(NO_3)_3 \cdot 6H_2O, Tm(NO_3)_3 \cdot 6H_2O$ were dissolved into 10 mL deionized water, and then stirred for 10 min at room temperature to form a transparency solution. Simultaneously, calculated amounts of $(NH_4)_6Mo_7O_{24}$ and citric acid were dissolved into two beakers contained 10 mL deionized water respectively and then stirred for 10 min at room temperature. Then, the above solution of nitrate and $(NH_4)_6Mo_7O_{24}$ were mixed into one beaker and stirred for 30 min to form a white suspension. Secondly, the solution of citric acid was poured into the white suspension dropwise under vigorous stirring. After the suspension changing into a transparent solution, the precursor of $Gd_2(MoO_4)_3:Yb^{3+}, Ho^{3+}, Tm^{3+}$ was prepared. After that, the precursor was put into the drying oven at 130 °C for 20 h and muffle furnace at 800 °C for 2 h. Finally, the phosphors were collected after cooled down to room temperature.

2.2. Instruments

X-ray diffraction (XRD) measurements were performed using a X-ray diffractometer (Empyrean, Panalytical, Netherlands), which use Cu $K\alpha$ radiation ($\lambda = 0.15406$ nm). The morphology was examined using a scanning electron microscope (JEM-2100F, JEOL, Japan). In our work, we measured fluorescence of the sample by using a iHR550 grating spectrograph. The dynamic processes of the sample were measured using a DSO5032A Digital Storage Oscilloscope. All the experiments were performed at the room temperature, and the diameter of laser spot was fixed at 0.5 mm.

3. Results and discussions

3.1. XRD analysis and morphological characterization

The XRD pattern and morphological characterization of our

sample is present in Fig. 1. In Fig. 1(a), the diffractions of $Gd_2(MoO_4)_3:Yb^{3+}, Ho^{3+}, Tm^{3+}$ phosphors are in good agreement with standard JCPDS file number 71-0915 (standard XRD pattern of $Gd_2(MoO_4)_3$) without any secondary phases. Moreover, comparing with the standard pattern (PDF#71-0915), an obvious shift towards higher diffraction angles is observed in the enlarged main peak (222) of the $Gd_2(MoO_4)_3:Yb^{3+}, Ho^{3+}, Tm^{3+}$ XRD pattern, as presented in Fig. 1(b). Thus, it is reasonable to deduce that the Gd^{3+} (93.8 p.m.) is substituted with smaller Yb^{3+} (85.8 p.m.), Ho^{3+} (89.4 p.m.) and Tm^{3+} (86.9 p.m.) in the crystal lattice. In Fig. 1(c), the TEM photo indicates that the sample exhibit a ellipse-like morphology with a long diameter of ~200 nm and short diameter of ~100 nm.

3.2. The up-conversion luminescence and CIE coordinates under different power of 980 and 915 nm continuous laser excitation

The up-conversion processes of the sample are plotted in Fig. 2. It is obviously that the Ho^{3+}, Tm^{3+} and Yb^{3+} co-doped materials

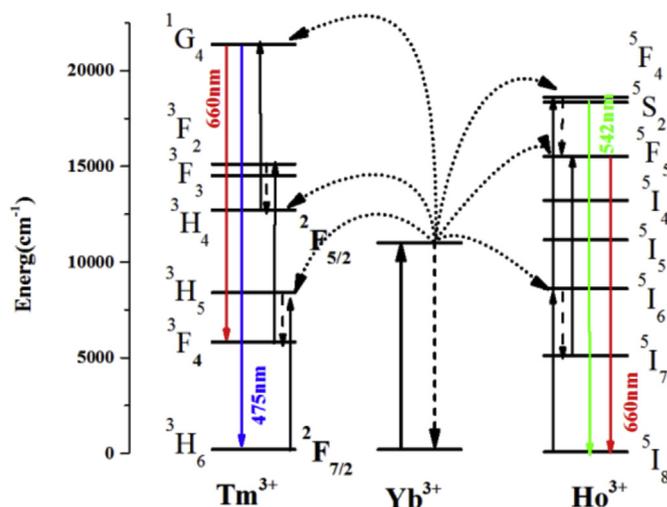


Fig. 2. Schematic energy levels and up-conversion processes among Tm^{3+}, Ho^{3+} and Yb^{3+} .

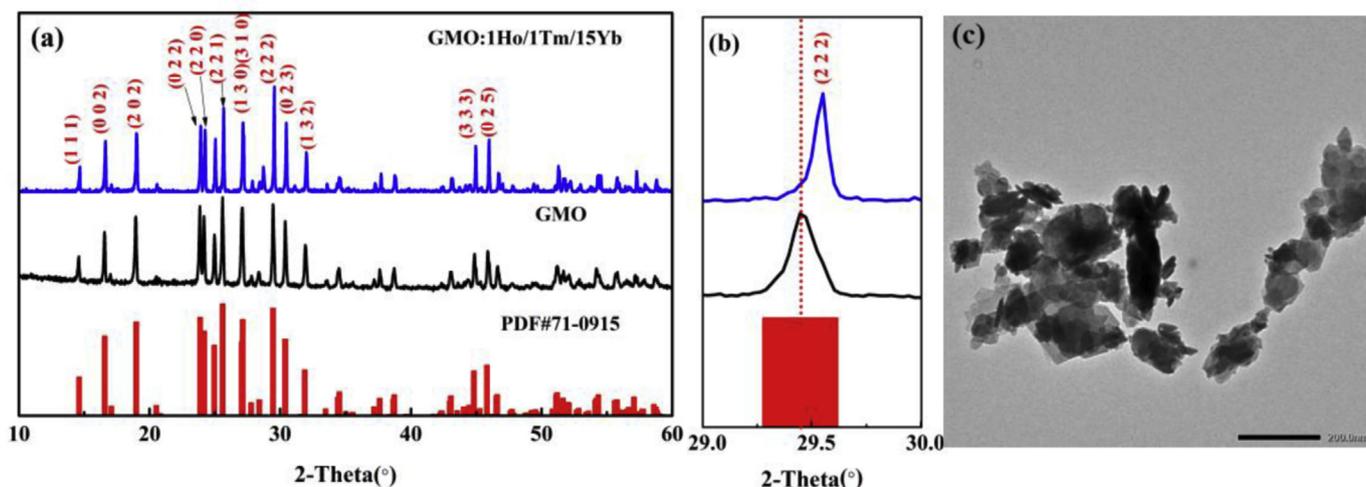


Fig. 1. (a) XRD patterns of $Gd_2(MoO_4)_3:Yb^{3+}, Ho^{3+}, Tm^{3+}$ and $Gd_2(MoO_4)_3$ phosphors; (b) enlarged (222) diffraction peaks in the XRD patterns; (c) TEM image of the $Gd_2(MoO_4)_3:Yb^{3+}, Ho^{3+}, Tm^{3+}$ phosphors (Scale bar is 200 nm).

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