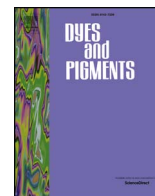




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Emission enhancement of $\text{LiLaMo}_2\text{O}_8:\text{Eu}^{3+}$ phosphor by co-doping with Bi^{3+} and Sm^{3+} ions

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ABSTRACT

A series of $\text{LiLaMo}_2\text{O}_8:\text{Eu}^{3+}$, R (R = Bi^{3+} and Sm^{3+}) phosphors are synthesized by a high temperature solid-state reaction method in air. The crystal structures, fluorescence lifetimes, and luminescence properties are investigated in detail. $\text{LiLaMo}_2\text{O}_8:\text{Eu}^{3+}$ phosphor with excitation 395 nm shows red light in the range of 525–725 nm and its photoluminescence intensity may be enhanced ~2.35 times due to energy transfer (ET) from a sensitizing species (Bi^{3+} or Sm^{3+}) to Eu^{3+} ion after Bi^{3+} or Sm^{3+} ion is co-doped. The optimal Eu^{3+} , Bi^{3+} , and Sm^{3+} ion concentrations are 4%. Luminous mechanism of $\text{LiLaMo}_2\text{O}_8:\text{Eu}^{3+}$, R (R = Bi^{3+} and Sm^{3+}) phosphors and ET processes between Eu^{3+} and $\text{Bi}^{3+}/\text{Sm}^{3+}$ ions are analyzed and explained by their excitation and emission spectra and energy level diagrams of Eu^{3+} , Bi^{3+} , and Sm^{3+} ions. The experimental results indicate that $\text{LiLaMo}_2\text{O}_8:\text{Eu}^{3+}$, R (R = Bi^{3+} and Sm^{3+}) phosphors may be used as potential red emitting materials for white light emitting diodes (LEDs) based on either a near ultraviolet (~395 nm) or blue (~465 nm) chip.

1. Introduction

The double molybdates with formula $\text{MRe}(\text{MoO}_4)_2$ (M = alkali metal and Re = trivalent rare earth ions) belong to the family of double molybdate compounds and have attracted a tremendous amount of attention because of the excellent thermal and chemical stabilities and the good absorption and emission cross-sections of rare earth ions in their lattices [1–3]. $\text{MRe}(\text{MoO}_4)_2$ is considered to be efficient luminescent host and rare earth ion doped. $\text{MRe}(\text{MoO}_4)_2$ phosphors have been reported widely due to their potential applications in solid-state lighting and lasers, such as $\text{Ca}_{0.5}\text{Y}_{1-x}(\text{MoO}_4)_2:\text{xRE}^{3+}$ (RE = Pr, Sm, Eu, Tb, Dy), $\text{NaGd}(\text{MoO}_4)_2:\text{Eu}^{3+}$, Tb^{3+} , $\text{Li}_x\text{Ag}_{1-x}\text{Yb}_{0.99}(\text{MoO}_4)_2:0.01\text{Er}^{3+}$, $\text{NaLa}(\text{MoO}_4)_2:\text{RE}^{3+}$ (RE³⁺ = Eu^{3+} , Sm^{3+} , $\text{Er}^{3+}/\text{Yb}^{3+}$), $\text{AgLa}(\text{MoO}_4)_2:\text{Yb}^{3+}$, Er^{3+} , $\text{KLa}(\text{MoO}_4)_2:\text{Dy}^{3+}$, Eu^{3+} , $\text{CaGd}_2(\text{MoO}_4)_4:\text{Eu}^{3+}$, $\text{NaLu}(\text{MoO}_4)_2:\text{Nd}^{3+}$, $\text{KGd}(\text{MoO}_4)_2:\text{Sm}^{3+}$, and $\text{AgTb}(\text{WO}_4)_2:\text{Ce}^{3+}$ [4–13]. $\text{LiLa}(\text{MoO}_4)_2$ as one of the double molybdates with structure similar to the scheelite CaWO_4 has a number of attractive peculiar features due to the low symmetry of the crystal lattice [14]. In $\text{LiLa}(\text{MoO}_4)_2$ crystal lattice, Mo^{6+} ion is coordinated by four O^{2-} ions in a tetrahedral symmetry, Li^+ and La^{3+} cations are randomly distributed over the same sites, which are coordinated by eight O^{2-} ions from near four MoO_4^{2-} [15,16]. The random distribution of La^{3+} is helpful for the inhomogeneous broadening of optical spectra when rare earth ions are doped into the crystal lattice and replace the positions of La^{3+} ions.

At present, rare earth ion doped $\text{LiLa}(\text{MoO}_4)_2$ phosphors have also been reported widely, such as $\text{LiLa}(\text{MoO}_4)_2:\text{Yb}^{3+}$, $\text{LiLa}(\text{MoO}_4)_2:\text{Tm}^{3+}$, $\text{LiLa}(\text{MoO}_4)_2:\text{Er}^{3+}$, $\text{LiLa}(\text{MoO}_4)_2:\text{Tm}^{3+}$, Ho^{3+} , $\text{LiLa}(\text{MoO}_4)_2:\text{Eu}^{3+}$, $\text{LiLa}(\text{MoO}_4)_2:\text{Dy}^{3+}$, and $\text{LiLa}(\text{MoO}_4)_2:\text{Nd}^{3+}$ [17–23]. However, their luminescence properties need to be further improved. It is well known that the energy transfer (ET) is one of important methods for the improvement of the luminescence properties of phosphors. In order to gain the practical application of rare earth ions doped $\text{LiLa}(\text{MoO}_4)_2$ phosphors, we study their luminescence properties improvements by ET process between different ions.

Bi^{3+} ion with the $[\text{Xe}]4f^{14}5d^{10}6s^2$ electronic configuration can show emission in the region from near ultraviolet (UV) to yellow light [24]. The improvement of rare earth ion luminescent properties has been investigated and reported extensively by ET between Bi^{3+} and rare earth ions, such as $\text{SrSb}_2\text{O}_6:\text{Eu}^{3+}$, Bi^{3+} , $\text{CaZrO}_3:\text{Sm}^{3+}$, Bi^{3+} , and $\text{Ca}_2\text{MgWO}_6:\text{Sm}^{3+}$, Bi^{3+} [25–27]. Besides, ET between rare earth ions can also improve the luminescence properties of phosphors and has been studied by many researchers, such as $\text{GdNbTiO}_6:\text{Eu}^{3+}/\text{Dy}^{3+}$, $\text{SrMoO}_4:\text{Sm}^{3+}$, Tb^{3+} , Na^+ , $\text{LaAl}_{11}\text{O}_{18}:\text{Eu}$, Tb , $\text{CaGd}_2(\text{WO}_4)_4:\text{Eu}^{3+}$, Sm^{3+} , $\text{Ca}_{19}\text{Ce}(\text{PO}_4)_{14}:\text{A}$ (A = $\text{Eu}^{3+}/\text{Tb}^{3+}/\text{Mn}^{2+}$), $\text{SrZn}_2(\text{PO}_4)_2:\text{Eu}^{3+}$, Tb^{3+} , Li^+ , $\text{NaLa}(\text{MoO}_4)_2:\text{Sm}^{3+}/\text{Dy}^{3+}$, and $\text{Ba}_3\text{La}(\text{PO}_4)_3:\text{Tb}^{3+}$, Sm^{3+} [28–35].

According to the above investigations and motivated by the attempt to develop efficient red-emitting phosphors for the application of white

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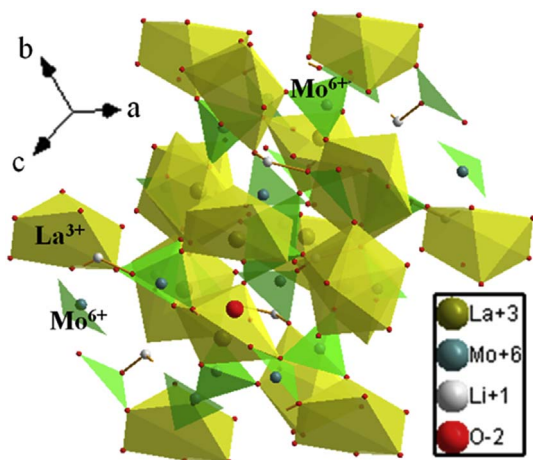


Fig. 1. Unit cell representation of the LiLaMo₂O₈ crystal structure drawn based on ICSD #4185.

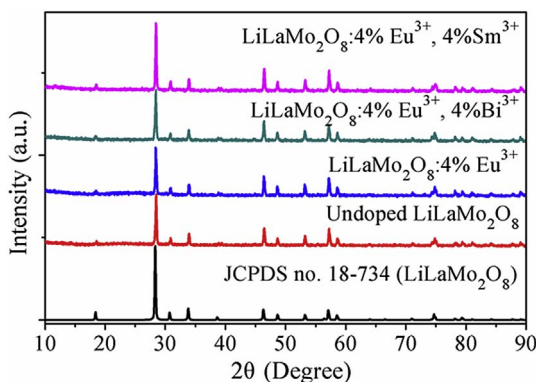


Fig. 2. XRD patterns of JCPDS [No. 18–734 (LiLaMo₂O₈)], undoped LiLaMo₂O₈, LiLaMo₂O₈:4%Eu³⁺, LiLaMo₂O₈:4%Eu³⁺, 4%Bi³⁺, and LiLaMo₂O₈:4%Eu³⁺, 4%Sm³⁺ phosphors at room temperature.

light emitting diodes (LEDs) based on either a near UV or a blue chip, we synthesized a series of LiLaMo₂O₈:Eu³⁺, R (R = Bi³⁺ and Sm³⁺) phosphors by high-temperature solid-state reaction method in air. We investigated their crystal structures and luminescence properties. The influence of Eu³⁺, Bi³⁺, and Sm³⁺ ions on the luminescence properties of phosphors is discussed. Luminous mechanism of LiLaMo₂O₈:Eu³⁺, R (R = Bi³⁺ and Sm³⁺) phosphors and ET processes between Eu³⁺ and Bi³⁺/Sm³⁺ ions are analyzed and explained by their spectra and energy level diagrams of Eu³⁺, Bi³⁺, and Sm³⁺ ions.

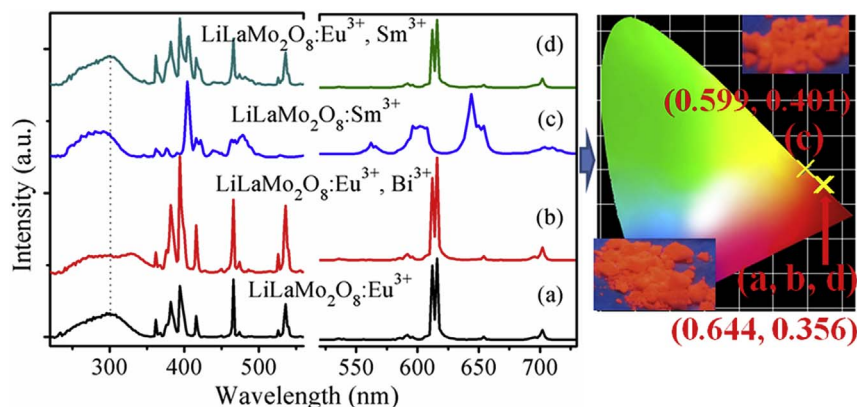


Fig. 3. PLE and PL spectra of (a) LiLaMo₂O₈:4%Eu³⁺ ($\lambda_{ex} = 395$ nm and $\lambda_{em} = 616$ nm), (b) LiLaMo₂O₈:4%Eu³⁺, 4% Bi³⁺ ($\lambda_{ex} = 395$ nm and $\lambda_{em} = 616$ nm), (c) LiLaMo₂O₈:4%Sm³⁺ ($\lambda_{ex} = 404$ nm and $\lambda_{em} = 644$ nm), and (d) LiLaMo₂O₈:4%Eu³⁺, 4%Sm³⁺ ($\lambda_{ex} = 395$ nm and $\lambda_{em} = 616$ nm) phosphors at room temperature and the corresponding CIE chromaticity diagram and chromaticity coordinates. The inset: The pictures of samples under 254 nm UV lamp.

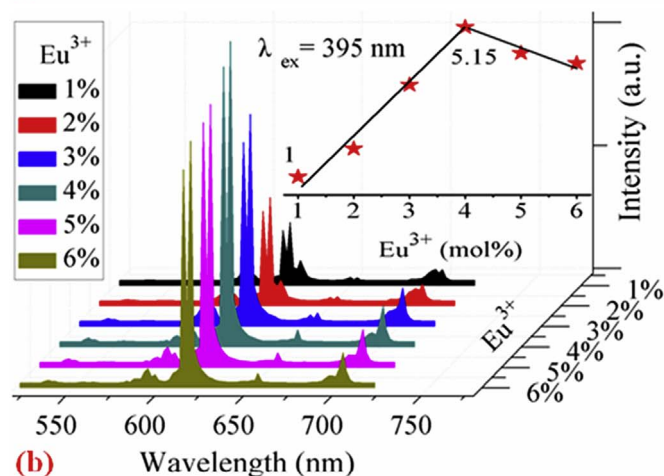
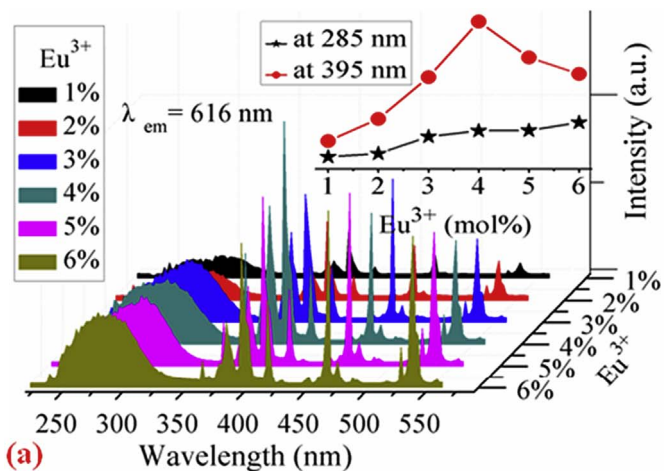


Fig. 4. (a) PLE and (b) PL spectra of LiLa_(1-x)Mo₂O₈:xEu³⁺ (1 ≤ x ≤ 6 mol%) phosphors at room temperature ($\lambda_{ex} = 395$ nm and $\lambda_{em} = 616$ nm). The insets: The influences of Eu³⁺ concentration to PLE and PL intensities.

2. Experimental section

2.1. Raw materials and sample synthesis

Here, pure chemicals Li₂CO₃ (99.9%), MoO₃ (99.9%), Bi₂O₃ (99.95%), Eu₂O₃ (99.99%), Sm₂O₃ (99.99%), and La₂O₃ (99.99%) are directly used as raw materials without further purification, which are purchased from the Aladdin Chemical Reagent Company in Shanghai, China.

In this work, a series of LiLa_(1-x)Mo₂O₈:xEu³⁺ (x = 0, 1, 2, 3, 4, 5, and 6 mol%), LiLa_(0.96-y)Mo₂O₈:4%Eu³⁺, yBi³⁺ (y = 1, 2, 3, 4, 5, and

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