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1. Introduction

Owing to the growing importance of energy storage and environmental friendliness, much attention has been paid to solid-state lighting (SSL) as the alternative to existing lighting technologies.¹⁻³ White lighting emitting diodes (WLEDs) have many advantages, such as a high energy efficiency, a long life, a good reliability and an excellent performance at low temperature.^{4,5} Generally, WLEDs are produced by mixing red, green and blue (RGB) LEDs, combining a blue LED with a yellow phosphor $(Y,Gd)_3(Al,Ga)_5O_{12}:Ce^{3+}$ or using ultraviolet (UV) LED-stimulated RGB phosphors.⁶⁻⁹ By means of the three methods, WLEDs with high efficiencies, high chromatic stabilities and tunable color temperatures, can be fabricated by UV-LED chips coated with RGB phosphors. However, the lack of efficient red phosphors drastically hinders the development of UV WLEDs. As an important branch of WLEDs, nanophosphors have gained much research interest due to their unique optical properties.¹⁰⁻¹³ Compared to ceramic materials, a reduction in particle size can dramatically modify the optical and electronic properties, *i.e.*, reducing the scattering losses

Hydrothermal synthesis and photoluminescence properties of rare-earth niobate and tantalate nanophosphors†

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Rare-earth niobate and tantalate materials are of considerable interest for use as phosphors, photocatalysts and ionic conductors. We successfully synthesized Ln_3MO_7 (Ln = Y, Ce, Er, Ho, Tm, Yb and Lu, M = Ta, Nb) nanophosphors by a hydrothermal method using the water-soluble Lindqvist ion polyoxometalates $[HNb_6O_{19}]^{7-}$ and $[Ta_6O_{19}]^{8-}$ as Nb and Ta sources. The Pawley refinements of these nanophosphors revealed that the Lu₃TaO₇ and Lu₃NbO₇ nanophosphors could be indexed in the cubic system with the space group $Fm\bar{3}m$, and that Y₃TaO₇ crystallizes in orthorhombic symmetry with the space group of C2221. These three solid compounds are nanoparticles with average particle sizes of 7.5, 5.9 and 4.0 nm for Lu₃TaO₇, Lu₃NbO₇ and Y₃TaO₇, respectively. The photoluminescence properties of Eu³⁺ doped Lu₃TaO₇, Lu₃NbO₇ and Y₃TaO₇ were studied, and Eu³⁺/Sr²⁺ co-doped Ln₃MO₇ (M = Ta, Nb) has an enhanced emission intensity compared to that of Ln₃MO₇:Eu³⁺ (M = Ta, Nb).

and receiving a high quantum yield. Among numerous nanophosphors, lanthanum-doped phosphors generate much interest for the narrow and independent intra 4f emission spectra of the rare-earth ions.^{14–16} For instance, Eu³⁺ doped phosphors can be used as the red component of WLEDs.^{17,18} There are many methods used to synthesize rare-earth nanophosphors, such as sol–gel,¹⁹ hydrothermal,²⁰ solvothermal,²¹ coprecipitation,²² sputtering and so on.²³ Phosphors prepared by the hydrothermal method crystallize well with a high purity. In addition, they were made at relatively low temperatures, and the agglomeration of nanoparticles was avoided.

Over the past decades, niobates and tantalates have attracted wide attention in the fields of photoluminescence and photocatalysis. Such nanocrystals are particularly attractive for bioimaging, as they do not degrade during analysis (photobleaching) compared to organic dyes, and are nontoxic compared to semiconducting nanoparticles (e.g., CdSe, PbS).²⁴ To our knowledge, there are few reports on the hydrothermal synthesis of lanthanum niobates and tantalates due to the difficulties caused by the low solubilities for both the rareearth species and the niobium and tantalum oxides. The common raw materials of niobium/tantalum, Nb₂O₅/Ta₂O₅ react under alkaline conditions while the solubilities of the rare-earth ions under alkaline conditions is rather low. It is remarkable that the May Nyman group have made a great contribution to the aqueous synthesis of polyionic clusters and have proposed the aqueous synthesis of rare-earth niobate and tantalate nanophosphors, e.g., LaTaO₄:Eu³⁺,²⁵ (La, K, \Box)₂- $Nb_2O_{7-x}(OH)_2$ and $Ln_2Ta_2O_7(OH)_2$ (\Box = vacancy; Ln = La-

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E-mail: shfeng@mail.jlu.edu.cn; Fax: +86-431-85168624; Tel: +86-431-85168661 †Electronic supplementary information (ESI) available: XRD patterns of Lu₃TaO₇ nanophosphors synthesized under different conditions (Fig. S1), and EDS spectra of Lu₃TaO₇, Lu₃NbO₇ and Y₃TaO₇ nanophosphors (Fig. S2). See DOI: 10.1039/c3dt50141h

Sm),²⁶ KLnTa₂O₇ and Eu³⁺-doped KLnTa₂O₇ (Ln = Lu, Y, Gd and Y).²⁷

In the present study, we have chosen Ln_3MO_7 (Ln = Y, Ce, Er, Ho, Tm, Yb and Lu, M = Ta, Nb) systems to investigate the effect of crystal structure on the photoluminescence properties. It has been reported that the crystal structure of Ln₃MO₇ is more distorted for the larger ionic radius of the Ln³⁺ ion, causing a change from the fluorite-type cubic structure to the pyrochlore-type cubic structure, and then finally to the weberite-type orthorhombic structure.^{28,29} Here, we report the synthesis of two families of Ln_3MO_7 (Ln = Y, Ce, Er, Ho, Tm, Yb and Lu, M = Ta, Nb) and detailed studies on the Lu_3TaO_7 , Lu₃NbO₇ and Y₃TaO₇ nanophosphors due to the fact that these three members can be readily used as host materials for doping rare-earth elements. In addition, the rare-earth ions may be coordinated with citric acid and dissolved in a weak alkaline solution, which is a base for further rare-earth element doping. Furthermore, compared to those of Lu₃MO₇: Eu^{3+} (M = Ta, Nb) nanophosphors, the emission intensities for $Lu_3MO_7:Eu^{3+}/Sr^{2+}$ (M = Ta, Nb) are enhanced.

2. Experimental

2.1 Chemicals

The raw materials $Lu(NO_3)_3$ (99.9%), $Eu(NO_3)_3$ (99.9%), $Y(NO_3)_3$ (99.9%), $Ce(NO_3)_3$ (99.9%), $Er(NO_3)_3$ (99.9%), $Ho(NO_3)_3$ (99.9%), $Tm(NO_3)_3$ (99.9%), $Yb(NO_3)_3$ (99.9%), Nb_2O_5 (99.99%), Ta_2O_5 (99.99%), $Sr(NO_3)_2$ (99.99%), KOH (AR grade) and citric acid (AR grade) were purchased from Sinopharm Chemical Reagent Co., Ltd. All chemicals were used without further purification.

2.2 The synthesis of the Lu₃TaO₇, Lu₃NbO₇ and Y₃TaO₇ nanophosphors

Lu₃TaO₇, Lu₃NbO₇ and Y₃TaO₇ nanophosphors were synthesized by the hydrothermal method. The water-soluble polyoxometalates $\left[HNb_6O_{19}\right]^{7-}$ and $\left[Ta_6O_{19}\right]^{8-}$ were obtained as previously reported.^{30,31} In a typical synthetic procedure for the Lu₃TaO₇ nanophosphor, 2 mL 0.25 M citric acid (0.50 mmol) was added to 2 mL 0.25 M Lu(NO₃)₃ solution (0.50 mmol) with stirring to form a suspension, to which 1 mL 4 M KOH solution (4 mmol) was added dropwise and the suspension became transparent. Finally, 1.11 mL 0.25 M [Ta₆O₁₉]⁸⁻ solution (0.28 mmol) was added to form the reaction mixture. After magnetically stirring for 30 min, the reaction mixture was transferred into a volume 25 mL and Teflon-lined stainless steel autoclave until it was 80% full. The crystallization was carried out under autogenous pressure at 240 °C for 3 days. The autoclave was cooled to room temperature naturally, and the solid product nanoparticles were separated by centrifugation at 3800 rpm. The nanoparticles were washed with deionized water several times until NO3⁻ could no longer be detected, and dried at 70 °C for 12 h. For the synthesis of Lu₃NbO₇, 0.83 mL 0.25 M [HNb₆O₁₉]⁷⁻ solution (0.21 mmol) was added instead. For the Eu³⁺-doped and Eu³⁺/Sr²⁺ co-doped

phosphors, the synthesis procedure was the same except for the fact that a certain proportion of Lu^{3+} was substituted by Eu^{3+} or Eu^{3+}/Sr^{2+} ions.

Although we have synthesized and X-ray characterized all the family members of Ln_3MO_7 (Ln = Y, Ce, Er, Ho, Tm, and Yb, M = Ta, Nb) nanophosphors by this method, here we present detailed studies on only the Lu_3TaO_7 , Lu_3NbO_7 and Y_3TaO_7 nanophosphors.

2.3 Characterization

Powder X-ray diffraction (XRD) data were collected using a Rigaku D/Max 2550 diffractometer with a graphite monochromator using CuK α radiation (λ = 1.5418 Å) operating at 50 kV and 200 mA at room temperature by step scanning in the angle range of $20^{\circ} \le 2\theta \le 80^{\circ}$ with increments of 0.02°. EDS was performed using the FIB-SEM instrument (Helios Nanolab 600i from FEI Company). Transmission electron microscopy (TEM) and high-resolution transmission electron microscopy (HRTEM) images were obtained using a Tecnai G2 FEI Company electron microscope. The emission and excitation spectra of the phosphors were recorded using an Edinburgh Instruments FLS 920 spectrofluorimeter equipped with a continuous 450 W xenon lamp and using a R928 photomultiplier tube detector. Low-temperature luminescence spectra were measured using an ARS (Advanced Research Systems Inc., USA) Model 4HW compressor and the temperature was controlled over the range of 6-280 K by a Lakeshore Model 331 temperature controller. Lifetime measurements were performed using the same spectrophotometer and detectors using a 100 W pulsed xenon lamp µF920H (200-900 nm, 10-100 Hz), Edinburgh. The optical absorption spectra of the phosphors were measured on a HITACHI U-4100 spectrometer. A Perkin-Elmer TGA 7 thermogravimetric analyzer was used to obtain the TG curve under an N2 atmosphere with a heating rate of 10 °C min⁻¹. The absolute quantum efficiency measurements were recorded using an integration sphere coupled with a photonic multichannel analyzer C10027 (HAMAMATSU).

3. Results and discussion

To coordinate the reaction conditions of the lanthanum niobates and tantalates, the water-soluble polyoxometalates $[HNb_6O_{19}]^{7-}$ and $[Ta_6O_{19}]^{8-}$, rather than Nb_2O_5 and Ta_2O_5 , were used as reagents. Moreover, the rare-earth ions were complexed with citric acid so that the compounds could be dissolved in weak alkaline conditions. As a result, the lanthanum niobates and tantalates can be obtained under weak alkaline conditions by the hydrothermal method.

3.1 Phase structures

The XRD patterns of the Ln_3MO_7 (Ln = Y, Ce, Er, Ho, Tm, Yb and Lu, M = Ta, Nb) nanophosphors and the standard XRD pattern of Lu_3TaO_7 at room temperature are shown in Fig. 1. It can be concluded from Fig. 1 that all of the nanophosphors can be indexed in the pure Ln_3MO_7 phase without any



Fig. 1 XRD patterns of the Ln_3MO_7 (Ln = Y, Ce, Er, Ho, Tm, Yb and Lu, M = Ta, Nb) nanophosphors. The red vertical lines at the bottom indicate the standard XRD pattern of Lu_3TaO_7 .

impurities. In Fig. 1, the XRD peaks of the Ce₃NbO₇ and Ce₃TaO₇ nanoparticles are shifted to lower 2-theta values and they are broad in comparison to that of other samples. This is because the ionic radius of Ce³⁺ is 0.114 nm, which is the biggest among the rare-earth ions used in this study. In addition, since the rare-earth niobates and tantalates are stabilized by small rare-earth ions, Ce₃NbO₇ and Ce₃TaO₇ show a relatively low crystallinity as seen from their broad XRD peaks.

The room temperature powder XRD patterns of the Lu₃TaO₇, Lu₃NbO₇ and Y₃TaO₇ nanophosphors and the Pawley refinements are shown in Fig. 2. No additional peaks of other phases have been detected, indicating the high purity and good crystallinity of the Lu₃TaO₇, Lu₃NbO₇ and Y₃TaO₇ products. All of the XRD peaks of Lu₃TaO₇ and Lu₃NbO₇ could be indexed in the cubic system with the space group $Fm\bar{3}m$, whereas Y₃TaO₇ crystallized with orthorhombic symmetry in the space group *C*2221. According to the EDS calculation, all nanophosphors contain the corresponding elements of Ln and M in a molar ratio of approximately 3 : 1. The obtained experimental powder diffraction patterns in the range of 20–80° are very similar with the simulated powder diffraction patterns of the previously reported single crystal structure. The lattice parameters refined by the Pawley method are listed in Table 1.

All sample nanophosphors can be hydrothermally synthesized from reaction mixtures with an initial alkaline content of 0–4 mL 4 M KOH and different amounts of citric acid (0–5 mL) at temperatures of 180–240 °C. The XRD patterns of the Lu₃TaO₇ nanophosphor synthesized under various conditions are given in Fig. S1.[†]

3.2 TG curves

Thermogravimetric analysis (TGA) is shown in Fig. 3 and indicates that the three compounds are thermally stable up to 1000 °C, having weight losses of 2.73%, 4.55% and 10.4% for Lu₃TaO₇, Lu₃NbO₇ and Y₃TaO₇, respectively, which can be attributed to surface water. Hence, these samples can be used as phosphors at high temperatures as they do not undergo significant structural changes compared to organic phosphors.



Fig. 2 The room temperature powder XRD patterns and the Pawley refinements of (a) Lu_3TaO_7 , (b) Lu_3NbO_7 , and (c) Y_3TaO_7 nanophosphors.

Table 1 The unit cell parameters of the $Lu_3TaO_7,\ Lu_3NbO_7$ and Y_3TaO_7 nanophosphors

	Lu ₃ TaO ₇	Lu ₃ NbO ₇	Y ₃ TaO ₇
a (Å)	5.1687(1)	5.1985(1)	10.4266(4)
b (Å)	5.1687(1)	5.1985(1)	7.3644(1)
c (Å)	5.1687(1)	5.1985(1)	7.4634(1)
$V(Å^3)$	138.08	140.49	573.08
Space group	$Fm\bar{3}m$	$Fm\bar{3}m$	C2221
$R_{\rm wp}$ (%)	7.09	4.26	3.41
$R_{\rm p}(\%)$	5.27	3.25	2.69



Fig. 3 Thermogravimetric analysis of the Lu_3TaO_7 , Lu_3NbO_7 and Y_3TaO_7 nanophosphors in the temperature range 20–1000 °C.

3.3 TEM images

Representative transmission electron microscopy (TEM) images of the Lu₃TaO₇, Lu₃NbO₇ and Y₃TaO₇ nanophosphors are shown in Fig. 4. Their average sizes determined from the longest dimensions among the Lu₃TaO₇, Lu₃NbO₇ and Y₃TaO₇ nanoparticles by the software Nano Measurer 1.2, are 7.5, 5.9 and 4.0 nm, respectively. The mean size of the products calculated from the Scherrer equation is 11.2 nm, 10.1 nm and 7.2 nm, which is very close to the size obtained from the TEM images. The variations within the range of reaction conditions



Fig. 4 Representative TEM images of the Lu₃TaO₇, Lu₃NbO₇ and Y₃TaO₇ nanophosphors. The average diameters of the nanocrystals are 7.5 nm, 5.9 nm and 4.0 nm, for the Lu₃TaO₇ (a), Lu₃NbO₇ (b) and Y₃TaO₇ (c) nanophosphors, respectively. Insets are HRTEM images of individual nanocrystals of (a), (b) and (c).



Fig. 5 A typical HRTEM image of a Lu₃TaO₇ nanophosphor.



Fig. 6 The UV/vis absorption spectra for the $Lu_3TaO_7,\ Lu_3NbO_7$ and Y_3TaO_7 nanophosphors.

seem to have no effect on the topography of the nanophosphors. From Fig. 4, it can be clearly observed that the nanophosphors are irregular in shape.

High-magnification TEM and high-resolution TEM (HRTEM) images provide a further insight into the morphology and structural information of the nanophosphors. The HRTEM image of an individual 7.5 nm nanoparticle displays high crystallinity in the Lu₃TaO₇ nanocrystal (Fig. 5). The distance between the adjacent atomic lattice fringes is about 0.32 nm, which is consistent with that of the (111) planes of tetragonal Lu₃TaO₇ (0.30 nm). In the cases of Lu₃NbO₇ and Y_3 TaO₇, the interplanar spacing distances are 0.34 and 0.37 nm, respectively.

3.4 UV/vis absorption spectra

The UV/vis absorption spectra for the Lu₃TaO₇ and Lu₃NbO₇ nanophosphors have onsets at around 350 nm as shown in Fig. 6. For the Y₃TaO₇ nanophosphor, there are two absorption bands centered at *ca.* 220 nm and 400 nm, corresponding to the charge-transfer transitions within the tantalate group TaO₆.³² This indicates that the Y₃TaO₇ nanophosphor has a



Fig. 7 Excitation and emission spectra for the Eu³⁺-doped Lu₃TaO₇, Lu₃NbO₇ and Y₃TaO₇ nanophosphors at room temperature, indicated by black, red, blue, dark cyan, magenta and navy lines, respectively.

high absorption not only in the UV region but also in the near-UV region (365–410 nm).

3.5 Photoluminescence properties

Fig. 7 clearly shows that the emission spectra of the Eu^{3+} doped Lu₃TaO₇, Lu₃NbO₇ and Y₃TaO₇ nanophosphors have a similar shape, while the excitation spectra are different. In the excitation spectra, values for the maximum λ_{ex} of 276, 296 and 274 nm monitored at 610 nm for the Eu³⁺-doped Lu₃TaO₇, Lu₃NbO₇ and Y₃TaO₇ nanophosphors were observed. The excitation band in the range of 250-350 nm is attributed to the charge-transfer (CT) transitions in the metal-oxygen polyhedra, *i.e.*, the electronic transitions between the filled O^{2-} 2p orbital and the partially filled 4f shell of Eu³⁺.³³ The intense excitation bands centered at 394 nm, 460 nm and 496 nm are due to the ${}^7F_0 \rightarrow {}^5L_6$, ${}^7F_0 \rightarrow {}^5D_2$ and ${}^7F_2 \rightarrow {}^5D_2$ transitions of Eu³⁺. In the emission spectrum of the Lu₃TaO₇:Eu³⁺ nanophosphor, the strongest emission at 610 nm corresponds to the electronic dipole ${}^{5}D_{0} \rightarrow {}^{7}F_{2}$ transition of Eu³⁺, suggesting the presence of Eu³⁺ asymmetric sites. The emission bands at 590, 654 and 710 nm correspond to the transitions of ${}^{5}D_{0} \rightarrow {}^{7}F_{1}$, $^5D_0 \rightarrow \,^7F_3$ and $\,^5D_0 \rightarrow \,^7F_4$ for Eu $^{3+}$ ions. It is worth noting that a peak at ca. 630 nm appears in the emission spectra for the Eu³⁺-doped Lu₃TaO₇ and Lu₃NbO₇ nanophosphors which is shown in Fig. 7, and is probably created by the energy level splitting at the ${}^{5}D_{0} \rightarrow {}^{7}F_{2}$ transition of the Eu³⁺ ions, resulting in more emission peaks.³⁴ There is no emission band around 580 nm (due to the ${}^5D_0 \rightarrow {}^7F_0$ transition), and therefore it could be inferred that the Eu³⁺ ions occupy lattice sites with inversion centres. In addition, the intensity of the line at around 590 nm is rather lower than that at 610 nm in the emission spectrum, indicating a lowering in the local point symmetry of Eu³⁺.

The photoluminescence intensity of the Lu_3TaO_7 nanophosphor doped with Eu^{3+} at various temperatures is shown in Fig. 8(a). The photoluminescence intensities decrease as the temperature increases, especially over the range of 6–20 K. This can be easily explained by the enhancement in the nonradiative transition at higher temperatures, *e.g.*, thermal



Fig. 8 (a) The temperature dependent photoluminescence intensity of Lu_3TaO_7 : Eu^{3+} . (b) The emission spectra of Lu_3TaO_7 : Eu^{3+} at 6 K and 280 K.

quenching. The photoluminescence intensity is almost constant over the temperature range 20–280 K. It may be predicted that there will not be much change in the photoluminescence intensity above room temperature. As the temperature increases, the emission shape and peak positions have no change but the intensity decreases, as illustrated in Fig. 8(b).

The luminescence decay curves of the Eu^{3+} -doped Lu_3TaO_7 , Lu_3NbO_7 and Y_3TaO_7 nanophosphors, obtained from timeresolved luminescence experiments are shown in Fig. 9. All plots are fitted by the single-exponential function

$$y = A\exp(-t/\tau) + y_0$$

where *A* is a pre-exponential factor obtained from the curve fitting and τ is the life time. The fluorescence life times of the Lu₃TaO₇:5% Eu³⁺, Lu₃NbO₇:10% Eu³⁺ and Y₃TaO₇:5% Eu³⁺ nanophosphors are 1.25 ms, 1.10 ms and 0.93 ms, respectively. The single decay time indicates that the chemical environment of the Eu³⁺ ions is uniform.

The photoluminescence intensities of the Eu^{3+} -doped Lu_3TaO_7 , Lu_3NbO_7 and Y_3TaO_7 nanophosphors quench with the increasing concentration of the doped Eu^{3+} as illustrated in Fig. 10. The intensity reaches a maximum when the mole ratio of Eu^{3+} is 5%, 10% and 5% for the Eu^{3+} -doped Lu_3TaO_7 , Lu_3NbO_7 and Y_3TaO_7 nanophosphors, respectively. The quantum yields of the optimized Eu^{3+} -doped Lu_3TaO_7 , Lu_3NbO_7 and Y_3TaO_7 nanophosphors are 4.3, 2.5 and 1.4%,



 $\label{eq:Fig.9} \mbox{Fig. 9} \mbox{ Decay curves of (a) Lu_3TaO_7:Eu^{3+}, (b) Lu_3NbO_7:Eu^{3+}$ and (c) Y_3TaO_7:Eu^{3+}$.}$

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 $\label{eq:Fig.10} \begin{array}{l} \mbox{Photoluminescence intensity as a function of the Eu^{3+} concentration for the Eu^{3+}-doped Lu_3TaO_7, Lu_3NbO_7 and Y_3TaO_7 nanophosphors. \end{array}$



respectively. The quantum yields of these nanoparticles are relatively low due to the nonradiative decay from defects on the surface of the nanocrystals. We thus carried out co-doping in the systems to enhance the luminescence density.

3.6 Enhanced luminescence from Lu_3MO_7 (M = Ta, Nb): Eu^{3+}/Sr^{2+}

To enhance the photoluminescence intensities of the Eu³⁺doped nanophosphors, Sr^{2+} was co-doped in the nanophosphors and the luminescence spectra were investigated. Fig. 11 shows the room temperature excitation and emission spectra of the Lu₃MO₇:Eu³⁺ and Lu₃MO₇:Eu³⁺/Sr²⁺ (M = Ta, Nb) nanophosphors. As shown in Fig. 11(a), the excitation and emission spectra of the Lu₃TaO₇:Eu³⁺ and Lu₃TaO₇:Eu³⁺/Sr²⁺

Fig. 11 (a) Excitation and emission spectra of the Lu₃TaO₇:Eu³⁺ (5%) (black line) and Lu₃TaO₇:Eu³⁺ (5%)/Sr²⁺ (5%) (red line) nanophosphors under 276 nm excitation. (b) Excitation and emission spectra of the Lu₃NbO₇:Eu³⁺ (5%) (black line) and Lu₃NbO₇:Eu³⁺ (5%)/Sr²⁺ (5%) (red line) nanophosphors under 296 nm excitation. All excitation and emission spectra are obtained by measuring the powder samples and the particle concentrations are kept the same.

nanophosphors have similar features, but the luminescence density is enhanced. The peak positions and the shape of the emission spectra are similar because the 4f energy levels of Eu^{3+} are hardly affected by the crystal field. The ${}^5D_0 \rightarrow {}^7F_1$ and ${}^5D_1 \rightarrow {}^7F_1$ transitions are magnetic-dipole-allowed and their

intensities are almost independent of the local environment around the Eu³⁺ ions. Compared to the Lu₃TaO₇:5% Eu³⁺ nanophosphors, the Lu₃TaO₇:5% Eu³⁺/5% Sr²⁺ nanophosphor shows an enhanced excitation and emission intensity, and the quantum yield of 5.4% is increased by about 26%. The enhanced emissions of Lu₃TaO₇:Eu³⁺/Sr²⁺ are either attributed to the increased absorption coefficient of the UV pump light or the enhanced luminescence efficiency caused by changing the composition and lattice parameters through co-doping with Sr²⁺.^{35,36} Compared to the Lu₃NbO₇:Eu³⁺ nanophosphors, the Lu₃NbO₇:Eu³⁺/Sr²⁺ nanophosphors have an enhanced excitation and emission intensity of about 8%, and a quantum yield of 2.7%. Further tuning of the photoluminescence intensities of this family of nanocrystals by co-doping with other ions is expected.

4. Conclusions

In summary, we have hydrothermally synthesized Lu₃TaO₇, Lu₃NbO₇ and Y₃TaO₇ nanophosphors and their doped and codoped compounds. We have understood the synthetic strategies, crystal structures and nanometer scale features for this family of nanophosphors. The photoluminescence properties of the Eu3+-doped nanophosphors show potential for applications but need further improvement to enhance the luminescence intensities. The optimum doping concentration for the Eu³⁺ doped Lu₃TaO₇, Lu₃NbO₇ and Y₃TaO₇ nanophosphors is 5%, 10% and 5%, with quantum yields of 4.3%, 2.5% and 1.4%, respectively. Compared to the $Lu_3MO_7:Eu^{3+}$ (M = Ta, Nb) nanophosphors, the emission intensities of Lu₃MO₇:Eu³⁺/Sr²⁺ (M = Ta, Nb) are enhanced. This work demonstrates the high temperature application of nanoparticle rare-earth niobates and tantalates as red phosphor candidates for WLEDs upon further improvements.

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