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# Crystal structure and luminescence properties of $(\text{Ca}_{2.94-x}\text{Lu}_x\text{Ce}_{0.06})(\text{Sc}_{2-y}\text{Mg}_y)\text{Si}_3\text{O}_{12}$ phosphors for white LEDs with excellent colour rendering and high luminous efficiency

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## Abstract

Lu-modified  $(\text{Ca}_{2.94-x}\text{Lu}_x\text{Ce}_{0.06})(\text{Sc}_{2-y}\text{Mg}_y)\text{Si}_3\text{O}_{12}$  (CLSMS :  $\text{Ce}^{3+}$ ) yellow emitting phosphors are prepared by a solid-state reaction. Controllable luminescent intensity and emitting colour are studied as a function of Lu and Mg contents. Fixing the Mg content to be 1, the effect of Lu content on crystal phase formation, luminescence properties and temperature characteristics is studied. It is revealed that the Lu-induced luminescent enhancement is the result of an increase in absorbance of  $\text{Ce}^{3+}$  rather than the internal quantum efficiency. Intense and broadband emission is realized by controlling the Lu content to obtain a pure CLSMS crystal phase. The maximum luminescence intensity is obtained at  $x = 0.54$ , which is as high as 156% of the Lu-free phosphor. The Lu-containing phosphor also exhibits better temperature characteristics for its big activation energy (0.20 eV) than the Lu-free one (0.18 eV). Combining the present phosphor with a blue light-emitting diode (LED) chip, a white LED with an excellent colour rendering index  $R_a$  of 86 and a high luminous efficiency of  $86 \text{ lm W}^{-1}$  is obtained. The results of the present study demonstrate that the CLSMS :  $\text{Ce}^{3+}$  phosphors show a good performance and are attractive candidates for commercial applications when used in white LEDs.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

White light-emitting diodes (LEDs) are a potential replacement for conventional light sources such as incandescent or fluorescent lamps as a consequence of their high efficiency,

long lifetime, low environment impact and lack of requirement for pollutants such as Hg [1, 2]. The most common and simple method to fabricate white LEDs is to mix the emission from a blue LED with a longer wavelength light downconverted from the LED emission using phosphors [3–6]. Until now, the most widely used commercial white LEDs consist of combinations of an InGaN-based blue LED

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and a  $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Ce}^{3+}$  (YAG: $\text{Ce}^{3+}$ ) yellow-emitting garnet phosphor [7, 8]. The YAG: $\text{Ce}^{3+}$  phosphor emits a yellow band peaking at 530 nm with a bandwidth of about 100 nm due to  $5d \rightarrow {}^2F_{7/2}, {}^2F_{5/2}$  transition [9]. The band shows a deficient red component [10, 11], consequently leading to a low colour rendering index ( $R_a < 80$ ) of white LEDs, which continues to be a major concern. To improve colour balance for true colour rendition, studies on the enhancement of the red component in luminescence of YAG: $\text{Ce}^{3+}$  have been done by co-doping other lanthanide ions [12, 13], such as partially replacing  $\text{Y}^{3+}$  by  $\text{Gd}^{3+}$  to shift the emission band to longer wavelengths. Coding  $\text{Gd}^{3+}$ , however, leads to a limited shift ( $< 10$  nm) and reduction in luminescence intensity.

In addition to aluminium garnets (YAG: $\text{Ce}^{3+}$ ),  $\text{Ce}^{3+}$  activated silicate garnets,  $\text{Lu}_2\text{CaMg}_2\text{Si}_3\text{O}_{12}$  red emitting phosphor ( $\lambda_{\text{max}} \sim 605$  nm) and  $\text{Ca}_3\text{Sc}_2\text{Si}_3\text{O}_{12}$  (CSS) green emitting phosphor ( $\lambda_{\text{max}} \sim 505$  nm), were studied in 2006 and 2007 by Setlur *et al* [14] and Shimomura *et al* [15], respectively. In particular, the luminescent efficiency of CSS: $\text{Ce}^{3+}$  can compete with commercial YAG: $\text{Ce}^{3+}$ . It is, therefore, significant to find an approach to modify the green emission CSS phosphor to be a yellow one suitable for white LEDs. For the garnet structure, it can be represented by the general formula  $\text{C}_3\text{A}_2\text{D}_3\text{O}_{12}$ , where C, A and D cations are eight-, six- and four-coordinated in dodecahedron, octahedron and tetrahedron, respectively [16]. The position of the  $\text{Ce}^{3+}$  5d levels is strongly influenced by the host lattice selection [17–19], and hence, the  $\text{Ce}^{3+}$  emission can be controlled through careful substitutions in the dodecahedral, octahedral and tetrahedral sites. For the silicate garnets CSS and  $\text{Lu}_2\text{CaMg}_2\text{Si}_3\text{O}_{12}$ , in 2010, Setlur and co-workers [20, 21] described the solid solution formation and demonstrated that  $\text{Sc}^{3+}$  has a strong preference towards the A (octahedral) site, and the emission maximum for  $\text{Ce}^{3+}$  5d  $\rightarrow$  4f emission can be shifted continuously from 505 to 605 nm, corresponding to shifts in the lowest energy  $\text{Ce}^{3+}$  4f  $\rightarrow$  5d excitation band for a large crystal splitting. However, the luminescence intensities and temperature characteristics decrease following the  $\text{Ce}^{3+}$  emission red shift in the solid solution. Earlier in 2008, Shimomura *et al* [22] performed a remarkable red shift of the emitting peak of CSS: $\text{Ce}^{3+}$  from 505 to 550 nm by partially replacing Sc by Mg to obtain a yellow emitting phosphor  $\text{Ca}_3(\text{Sc}_{2-b}\text{Mg}_b)\text{Si}_3\text{O}_{12}:\text{Ce}^{3+}$  ( $0 \leq b \leq 1$ ) (CSMS: $\text{Ce}^{3+}$ ), which holds a more broad emitting band than YAG: $\text{Ce}^{3+}$ , demonstrating the advantage of fabricating white LEDs with excellent colour rendering. It was, however, also observed that a decrease in luminescence intensity occurs for Mg content  $b$  higher than 0.06.

In this paper, we aim at enhancing the luminescence intensity of the CSMS: $\text{Ce}^{3+}$  yellow phosphor for developing a promising LED phosphor. Considering the existence of the by-product  $\text{Ca}_2\text{MgSi}_2\text{O}_7$  phase in CSMS: $\text{Ce}^{3+}$  phosphor [22], we consider that the by-product may play an important role in decreasing the luminescence intensity. Based on this consideration, we have successfully enhanced the luminescence in CSMS: $\text{Ce}^{3+}$  by adding Lu to eliminate the by-product  $\text{Ca}_2\text{MgSi}_2\text{O}_7$  phase. The effect of Lu addition on the red shift of the spectral band and the improvement

of luminescence intensities and temperature characteristics are also observed. In addition, initial explanations are given for the enhancements of the emission intensities and temperature properties. Combining the Lu-modified CSMS: $\text{Ce}^{3+}$  phosphor (CLSMS: $\text{Ce}^{3+}$ ) with a blue LED chip, a white LED with an excellent colour rendering index  $R_a$  of 86 and a high luminous efficiency of  $86 \text{ lm W}^{-1}$  is obtained.

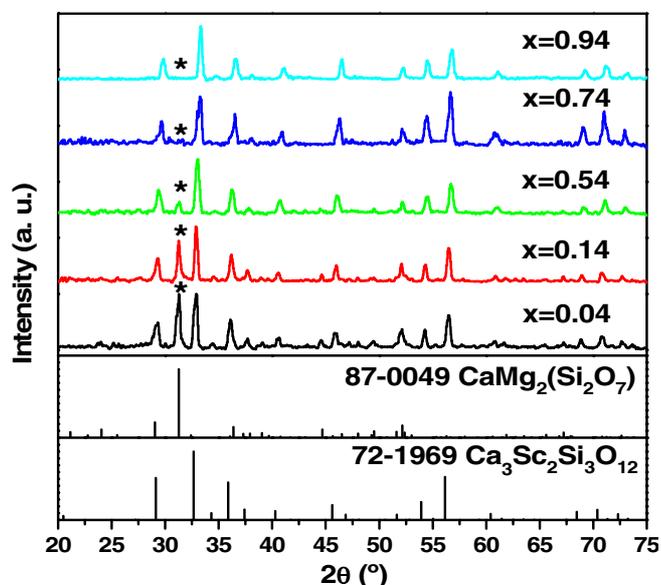
## 2. Experimental

The series of samples investigated in this work with the general formula  $(\text{Ca}_{2.94-x}\text{Lu}_x\text{Ce}_{0.06})(\text{Sc}_{2-y}\text{Mg}_y)\text{Si}_3\text{O}_{12}$  (CLSMS: $\text{Ce}^{3+}$ ) with  $0 \leq x \leq 1$  and  $0 \leq y \leq 1$  were prepared by a solid-state reaction. The raw materials of high-purity  $\text{CaCO}_3$ ,  $\text{Lu}_2\text{O}_3$ ,  $\text{CeO}_2$ ,  $\text{Sc}_2\text{O}_3$ ,  $\text{MgO}$  and  $\text{SiO}_2$  were homogenized by an agate mortar, according to stoichiometric proportions, pestled for 1 h and placed in a crucible with a lid. The crucible was placed in a tubular furnace and fired under reducing conditions of 5%  $\text{H}_2$  + 95%  $\text{N}_2$  at 1250 °C for 4 h. All samples were doped with 6 mol%  $\text{Ce}^{3+}$ , which is the critical concentration for luminescence quenching. The structures of the sintered samples were identified over the scattering range of  $20^\circ \leq 2\theta \leq 75^\circ$  in steps of  $0.02^\circ$  by an x-ray powder diffractometer (XRD) (Rigaku D/M AX-2500V). Photoluminescence (PL), photoluminescence excitation (PLE) and diffuse reflectance (DR) spectra were measured using a Hitachi F4500 fluorescent spectrometer. The decay curve of the yellow emission was measured using an FL920 Fluorescence Lifetime Spectrometer (Edinburgh Instruments Ltd). Prototype LEDs were fabricated by applying an intimate mixture of the phosphor powder and transparent silicone resin on blue InGaN LED chips ( $\lambda_{\text{ex}} = 460$  nm). The chromaticity coordinates, colour rendering indices (CRIs) and the correlated colour temperature ( $T_c$ ) in kelvin of the fabricated white LEDs were measured using an Ocean Optics USB4000 Spectrometer.

## 3. Results and discussion

Figure 1 shows the XRD patterns of the Lu-modified CSMS: $\text{Ce}^{3+}$  phosphors (CLSMS: $\text{Ce}^{3+}$ ) with nominal compositions of  $(\text{Ca}_{2.94-x}\text{Lu}_x\text{Ce}_{0.06})(\text{ScMg})\text{Si}_3\text{O}_{12}$  ( $x = 0.04, 0.14, 0.54, 0.74$  and  $0.94$ ). The phosphors do not exhibit a single garnet phase of CSS (JCPDS No 72-1969) but include a by-product, which is the akermanite phase of  $\text{Ca}_2\text{MgSi}_2\text{O}_7$  (JCPDS No 87-0049) as observed in  $\text{Ca}_3(\text{ScMg})\text{Si}_3\text{O}_{12}:\text{Ce}^{3+}$  [22]. With the increase in Lu content  $x$ , the XRD peaks of the by-product (asterisks) become weak and finally disappear at  $x = 0.94$ , indicating that more and more Mg is incorporated in the CSS phase rather than the by-product phase.

It is generally considered that Ce and Mg replace the Ca and Sc sites, respectively, in CSMS: $\text{Ce}^{3+}$ . Accordingly, we consider that Lu most likely occupies the Ca site in the present phosphors CLSMS: $\text{Ce}^{3+}$ . It can be carefully found in figure 1 that the XRD peaks of the CSS phase shift to higher angles with increasing  $x$ . This is an indication of a  $\text{Lu}^{3+}$  (with the ionic radius of  $0.86 \text{ \AA}$ ) substitution for a  $\text{Ca}^{2+}$  with the ionic radius of  $1.00 \text{ \AA}$  larger than  $\text{Lu}^{3+}$  so as to shrink the

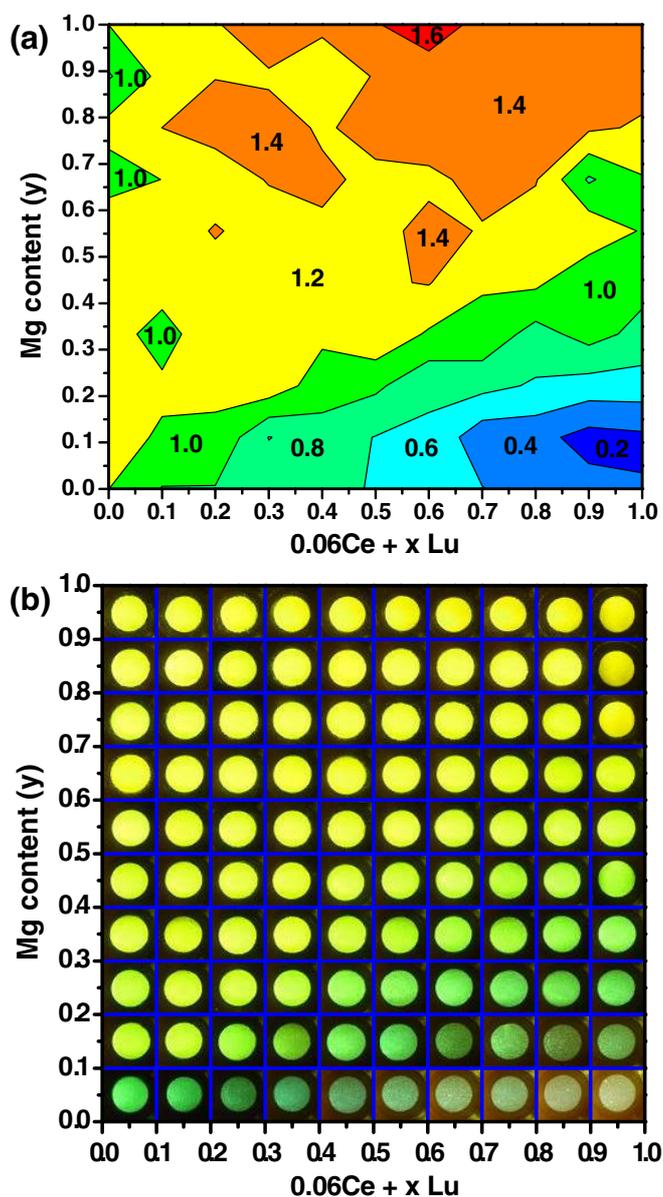


**Figure 1.** XRD patterns for nominal compositions of  $(\text{Ca}_{2.94-x}\text{Lu}_x\text{Ce}_{0.06})(\text{ScMg})\text{Si}_3\text{O}_{12}$  ( $x = 0.04, 0.14, 0.54, 0.74$  and  $0.94$ ). The asterisks indicate the by-products of  $\text{Ca}_2\text{MgSi}_2\text{O}_7$ .

lattice parameters. Due to the difference in valency between  $\text{Mg}^{2+}$  and  $\text{Sc}^{3+}$ , and between  $\text{Lu}^{3+}$  and  $\text{Ca}^{2+}$ , a  $\text{Lu}^{3+}$  at a  $\text{Ca}^{2+}$  site  $[\text{Lu}_{\text{Ca}}]^+$  may compensate for the charge of a  $\text{Mg}^{2+}$  at a  $\text{Sc}^{3+}$  site  $[\text{Mg}_{\text{Sc}}]^-$  in  $\text{CLSMS}:\text{Ce}^{3+}$ . When the amount of Mg is fixed, the negatively charged  $[\text{Mg}_{\text{Sc}}]^-$  may gradually be compensated with increasing content of Lu, suggesting that charge compensation by the addition of Lu favours the formation of the CSS phase and consequently suppresses the formation of the by-product.

The dependence of PL intensities on both Lu and Mg contents is examined in  $\text{CLSMS}:\text{Ce}^{3+}$  phosphors with the composition of  $(\text{Ca}_{2.94-x}\text{Lu}_x\text{Ce}_{0.06})(\text{Sc}_{2-y}\text{Mg}_y)\text{Si}_3\text{O}_{12}$  ( $x = 0-0.94$ ,  $y = 0-1$ ), as shown in figure 2(a), in which the PL intensity of the green phosphor  $\text{CSS}:\text{Ce}^{3+}$  ( $x = 0$ ,  $y = 0$ ) is normalized. All samples are excited at 450 nm. The photographs of the emission phosphors are taken through a 500 nm cut-off filter, as illustrated in figure 2(b), which demonstrates controllable emitting colours as a function of Mg and Lu contents. In figure 2(a), one finds that the intense PL occurs in the content region of  $y > 0.5$ . In this work, we focus our study on the samples with a fixed  $y = 1$  and different Lu contents.

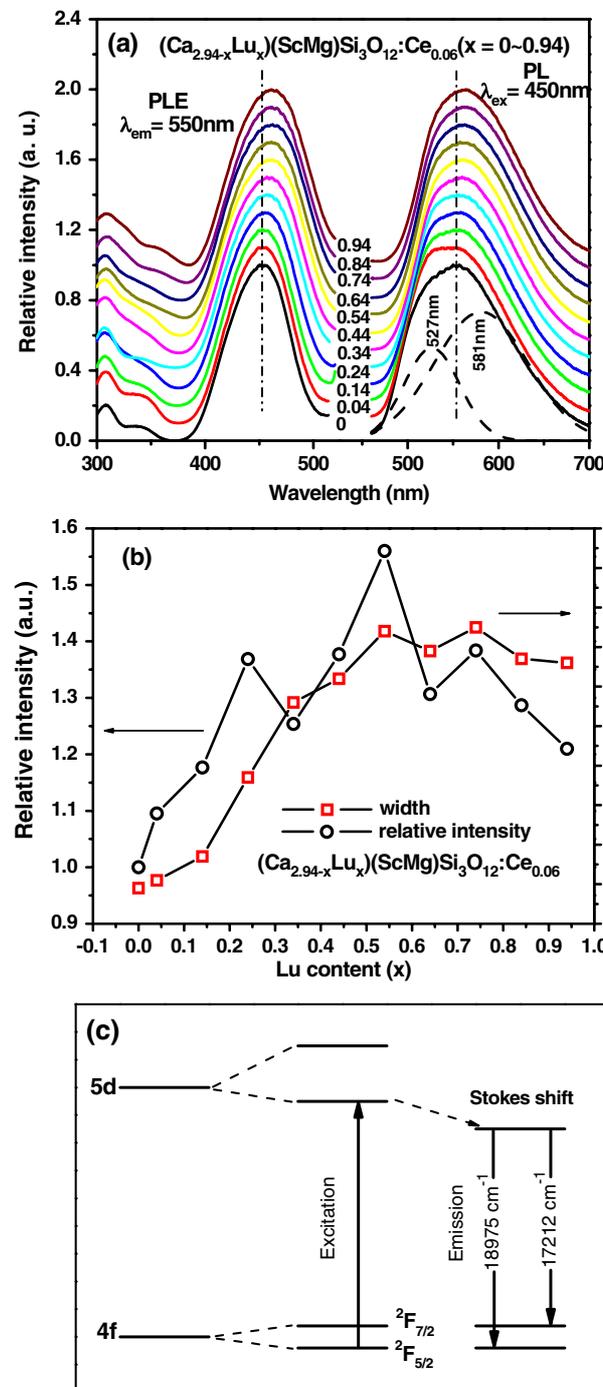
Figure 3(a) shows the normalized PL and PLE spectra of  $\text{CLSMS}:\text{Ce}^{3+}$  phosphors  $(\text{Ca}_{2.94-x}\text{Lu}_x\text{Ce}_{0.06})(\text{ScMg})\text{Si}_3\text{O}_{12}$  with  $x = 0-0.94$  and  $y = 1.0$ . The PL bands are centred within 545 and 560 nm depending on  $x$ , responsible for the transition of  $\text{Ce}^{3+}$  from the 5d excited state to the  $^2\text{F}_{5/2}$  and  $^2\text{F}_{7/2}$  ground states. The band can be decomposed into two Gaussian bands (dashed lines) centered at about 527 nm ( $18975\text{ cm}^{-1}$ ) and 581 nm ( $17212\text{ cm}^{-1}$ ) with an energy difference of about  $1763\text{ cm}^{-1}$ , which is very close to the energy separation between  $^2\text{F}_{5/2}$  and  $^2\text{F}_{7/2}$  levels. The PL band shifts to longer wavelengths by addition of Lu with the bandwidth in the range 120–130 nm, somewhat wider than the width of  $\sim 100$  nm in  $\text{YAG}:\text{Ce}^{3+}$ . This implies that  $\text{CLSMS}:\text{Ce}^{3+}$  phosphors may



**Figure 2.** (a) Integrated relative luminescence intensity of phosphors  $(\text{Ca}_{2.94-x}\text{Lu}_x\text{Ce}_{0.06})(\text{Sc}_{2-y}\text{Mg}_y)\text{Si}_3\text{O}_{12}$  with  $x = 0-0.94$  and  $y = 0-1$ . The relative intensity is signed in the figure; (b) corresponding photographs of emitting colours of phosphors in the  $(\text{Ca}_{2.94-x}\text{Lu}_x\text{Ce}_{0.06})(\text{Sc}_{2-y}\text{Mg}_y)\text{Si}_3\text{O}_{12}$  system. All the samples are excited at 450 nm.

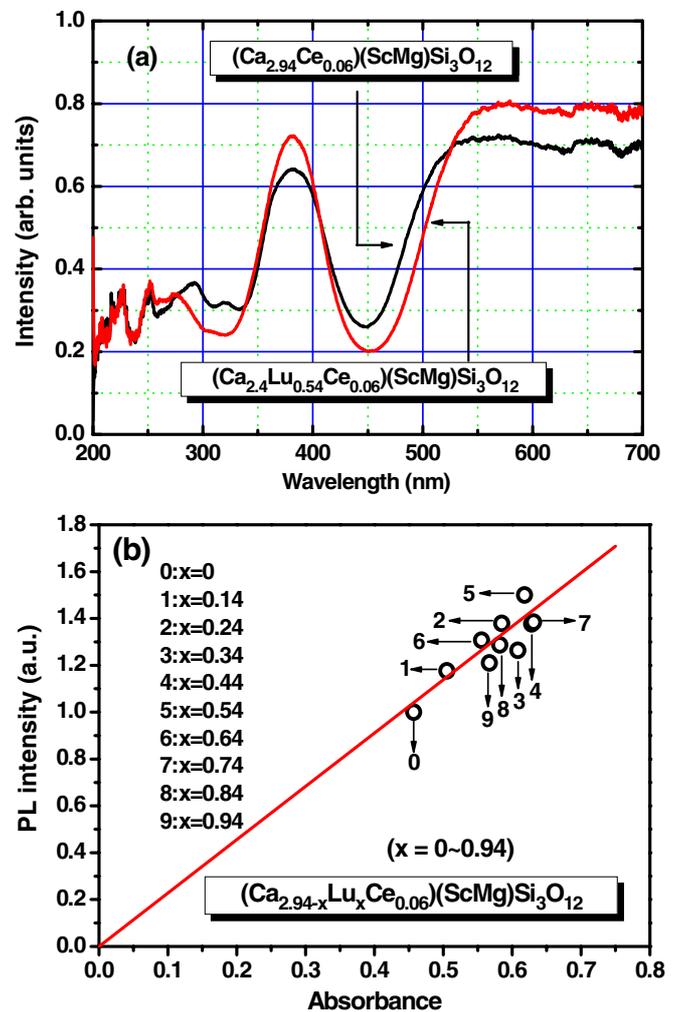
provide a high colour rendering  $R_a$  when incorporated into white LED lamps. Meanwhile, the PLE band also exhibits a red shift and broadens from 68 to 81 nm with increasing  $x$  up to 0.94, as shown in figure 3(b), indicating that the Lu-containing phosphors can take a better advantage of the blue light than the Lu-free phosphors. From the PLE spectra in figure 3(a), some excitation bands in the ultraviolet region are also observed, which are related to the high energy 5d levels of  $\text{Ce}^{3+}$ . On the basis of the excitation and emission spectra analysed above, an energy band diagram of  $\text{Ce}^{3+}$  is proposed as illustrated in figure 3(c).

The dependence of the integrated PL intensity on Lu content in  $(\text{Ca}_{2.94-x}\text{Lu}_x\text{Ce}_{0.06})(\text{ScMg})\text{Si}_3\text{O}_{12}$  ( $x = 0-0.94$ )



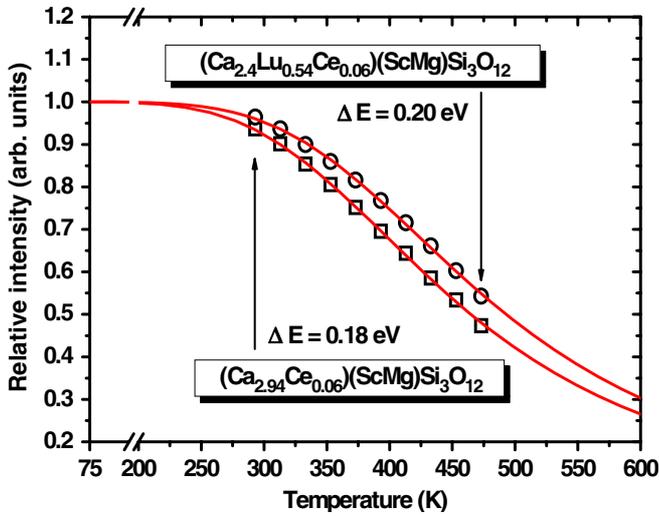
**Figure 3.** (a) Normalized PLE ( $\lambda_{em} = 550$  nm) and PL ( $\lambda_{ex} = 450$  nm) spectrum of  $(Ca_{2.94-x}Lu_x)(ScMg)Si_3O_{12} : Ce_{0.06}^{3+}$  with  $x = 0-0.94$ , and (b) integrated relative luminescence intensity and width of PLE band as a function of the Lu content  $x$  and (c) energy band diagram of  $Ce^{3+}$ .

is displayed in figure 3(b). It can be clearly seen that the PL intensity is significantly enhanced with increasing  $x$  and approaches a maximum at  $x = 0.54$ , which is as high as 156% of the Lu-free phosphor. According to the XRD, the PL enhancement on increasing  $x$  is reasonably attributed to the reduction of the by-product. As  $x$  goes beyond 0.54, a decrease in PL intensity is observed. To understand this phenomenon, DR spectra of the phosphors are measured.



**Figure 4.** (a) DR spectra for phosphors  $(Ca_{2.6}Lu_{0.54}Ce_{0.06})(ScMg)Si_3O_{12}$  and  $(Ca_{2.94}Ce_{0.06})(ScMg)Si_3O_{12}$ , and (b) relationship between PL intensity and absorption capacity of phosphors  $(Ca_{2.94-x}Lu_xCe_{0.06})(ScMg)Si_3O_{12}$  with  $x = 0-0.94$ .

Figure 4(a) shows the DR spectra of Lu-free CSMS :  $Ce^{3+}$  and 0.54 Lu-containing CLSMS :  $Ce^{3+}$  phosphors. There appears a strong absorption band of  $Ce^{3+}$  in the blue spectral region. The relationship between the PL intensity and the absorbance of  $Ce^{3+}$  is shown in figure 4(b). The PL intensities exhibit a linear relationship with absorbance, which means that the internal quantum efficiency of the phosphors is almost unchanged with the addition of Lu. Consequently, the change in PL intensity is the indication of the change in absorbance of  $Ce^{3+}$ . The increase in PL intensity on increasing  $x$  up to 0.54 reflects the increase in absorbance of  $Ce^{3+}$ , which is well understood knowing the reduction of the by-product so as to increase the number of  $Ce^{3+}$  in the CLSMS matrix. For  $x > 0.54$ , the by-product is almost eliminated according to XRD. In this case, it is speculated that more defects are formed due to the addition of Lu. These defects may act as absorbers to compete with  $Ce^{3+}$  so as to reduce the absorbance of  $Ce^{3+}$  and then PL intensity. A wider and red-shifted absorption band of the Lu-modified phosphor is also noted in figure 4(a), which is consistent with the PLE spectra in figure 3(a).



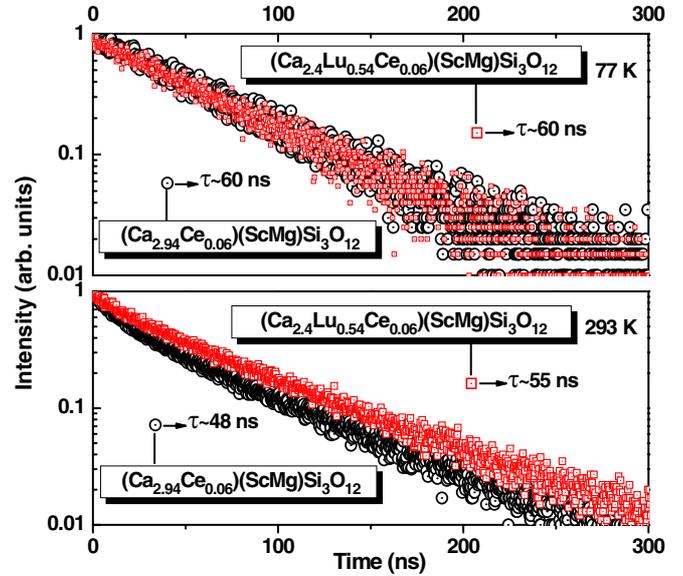
**Figure 5.** Temperature dependence of PL intensity of  $(\text{Ca}_{2.94}\text{Ce}_{0.06})(\text{ScMg})\text{Si}_3\text{O}_{12}$  and  $(\text{Ca}_{2.4}\text{Lu}_{0.54}\text{Ce}_{0.06})(\text{ScMg})\text{Si}_3\text{O}_{12}$  under excitation at 450 nm. The black dots and squares are experimental data and the red lines are fitting functions.

From our study, not only the luminescence properties but also the temperature-quenching characteristics are improved, as described in figure 5. The PL intensity of the Lu-containing phosphor  $(\text{Ca}_{2.4}\text{Lu}_{0.54}\text{Ce}_{0.06})(\text{ScMg})\text{Si}_3\text{O}_{12}$  (CLSMS:  $\text{Ce}^{3+}$ ) decreases slower than that of the Lu-free CSMS:  $\text{Ce}^{3+}$  phosphor as temperature rises. According to the classical theory of thermal quenching, the temperature-dependent PL intensity can be described by the equation

$$I(T) = \frac{I(0)}{1 + A \exp(-\Delta E/k_B T)} \quad (1)$$

with constant  $A$ , activation energy  $\Delta E$  and Boltzmann constant  $k_B$  [23]. The experimental data are well fitted by equation (1), as shown in figure 5. An activation energy of 0.20 eV is obtained for the Lu-modified phosphor, which is bigger than that for the Lu-free phosphor with  $\Delta E = 0.18$  eV. This is why the Lu-modified phosphor exhibits a better temperature characteristic.

Fluorescent decay curves of the Lu-containing CLSMS:  $\text{Ce}^{3+}$  phosphor and the Lu-free CSMS:  $\text{Ce}^{3+}$  phosphor are examined as presented in figure 6. At room temperature (RT) (293 K), the fluorescence in Lu-containing phosphor decays slower than that in the Lu-free sample. The measured fluorescence lifetimes ( $\tau$ ) for the Lu-containing and Lu-free phosphors are 55 ns and 48 ns, respectively. At liquid nitrogen temperature (77 K), the decay curves of the two samples are almost identical, showing an exponential decay pattern with a lifetime of 60 ns. Using equation (1) and the fitting parameters in figure 5, as well as  $\tau = 60$  ns at 77 K, the fluorescence lifetimes for the Lu-containing and Lu-free phosphors at RT can be estimated to be 57 ns and 55 ns, respectively, which are nearly consistent with the experimental values. Based on the results discussed above, the Lu-modified CLSMS:  $\text{Ce}^{3+}$  phosphor can exhibit good performance if employed in white LED lamps.

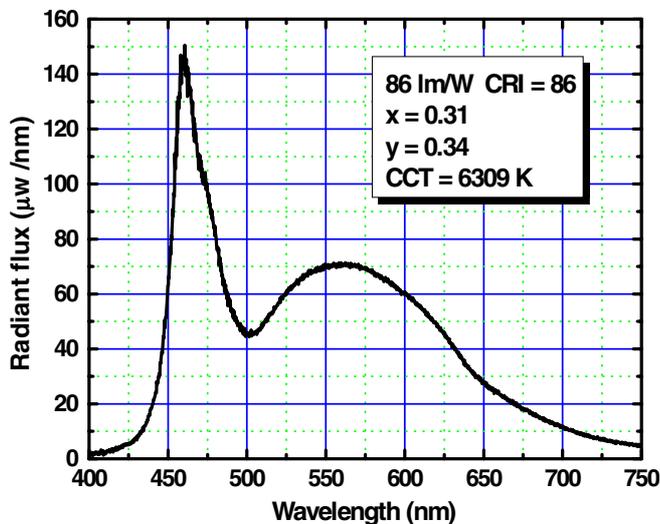


**Figure 6.** Decay curves of phosphors  $(\text{Ca}_{2.94-x}\text{Lu}_x\text{Ce}_{0.06})(\text{ScMg})\text{Si}_3\text{O}_{12}$  ( $x = 0$  and  $0.54$ ) at RT (293 K) and liquid nitrogen temperature (77 K).

Furthermore, a white LED combined with a blue (460 nm) InGaN LED having the Lu-modified yellow phosphor CLSMS:  $\text{Ce}^{3+}$  is fabricated. The emission spectrum of the white LED under a forward-bias current of 20 mA is shown in figure 7. It shows the Commission International de l'Eclairage (CIE) colour coordinates of (0.31, 0.34), a colour temperature ( $T_C$ ) of 6309 K, a high colour rendering index  $R_a$  of 86 and a high luminous efficiency of  $86 \text{ lm W}^{-1}$ . These results confirm that the Lu-modified yellow phosphors CLSMS:  $\text{Ce}^{3+}$  exhibit an excellent performance for white LEDs, which will be an attractive alternative to commercially used YAG:  $\text{Ce}^{3+}$  phosphors.

#### 4. Conclusions

In this report, we have detailed the synthesis and luminescence properties of Lu-modified CLSMS:  $\text{Ce}^{3+}$  phosphors with yellow emission. The experimental results obtained in this study reveal that the control of the content of Lu is important for the phase formation of the CLSMS:  $\text{Ce}^{3+}$  phosphors with excellent PL properties. In addition, we presented initial explanations for the significant enhancements of the luminescence intensities and temperature characteristics of the Lu-modified yellow phosphor CLSMS:  $\text{Ce}^{3+}$  by the improved absorbance and activation energy, respectively. When the Lu-modified yellow CLSMS:  $\text{Ce}^{3+}$  phosphors are coated on a blue LED chip, white LED lamps with an excellent colour rendering property and high luminous efficiency are obtained. The white LED exhibits a high luminous efficiency of  $86 \text{ lm W}^{-1}$ , a  $R_a$  of 86 and a  $T_C$  of 6309 K. The colour rendering is excellent. These results of this study suggest that the CLSMS:  $\text{Ce}^{3+}$  phosphors prepared here are attractive candidates for commercial applications when used in white LEDs.



**Figure 7.** Luminescence spectra of white LEDs consisting of blue InGaN LEDs and the yellow phosphors ( $\text{Ca}_{2.4}\text{Lu}_{0.54}\text{Ce}_{0.06}$ )(ScMg)Si<sub>3</sub>O<sub>12</sub> under the forward-bias currents of 20 mA.

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### References

- [1] Nakamura S, Mukai T and Senoh M 1994 *Appl. Phys. Lett.* **64** 1687–9
- [2] Schubert E F and Kim J K 2005 *Science* **308** 1274–8
- [3] Nakamura S 1997 *Proc. SPIE* **3002** 26–35
- [4] Shur M S and Zukauskas A 2005 *Proc. IEEE* **93** 1691–703
- [5] Park J K, Lim M A, Kim C H, Park H D, Park J T and Choi S Y 2003 *Appl. Phys. Lett.* **82** 683–5
- [6] Kim J S, Jeon P E, Choi J C, Park H L, Mho S I and Kim G C 2004 *Appl. Phys. Lett.* **84** 2931–3
- [7] Narukawa Y, Narita J, Sakamoto T, Deguchi K, Yamada T and Mukai T 2006 *Japan. J. Appl. Phys. Part 2* **45** L1084–6
- [8] Bando K, Sakano K, Noguchi Y and Shimizu Y 1998 *J. Light Visual Environ.* **22** 2–5
- [9] Blasse G and Bril A 1967 *Appl. Phys. Lett.* **11** 53–5
- [10] Chen Y, Gong M, Wang G and Su Q 2007 *Appl. Phys. Lett.* **91** 071117
- [11] Jang H S, Im W B, Lee D C, Jeon D Y and Kim S S 2007 *J. Lumin.* **126** 371–7
- [12] Kong L, Gan S C, Hong G Y and Zhang J L 2007 *J. Rare Earths* **25** 692–6
- [13] Zhang K, Hu W B, Wu Y T and Liu H Z 2008 *Physica B* **403** 1678–81
- [14] Setlur A A, Heward W J, Gao Y, Srivastava A M, Chandran R G and Shankar M V 2006 *Chem. Mater.* **18** 3314–22
- [15] Shimomura Y, Honma T, Shigeiwa M, Akai T, Okamoto K and Kijima N 2007 *J. Electrochem. Soc.* **154** J35–8
- [16] Geller S 1967 *Z. Kristallogr.* **125** 1–47
- [17] Dorenbos P 2001 *Phys. Rev. B* **64** 125117
- [18] Dorenbos P 2002 *Phys. Rev. B* **65** 235110
- [19] Dorenbos P 2003 *J. Lumin.* **105** 117–9
- [20] Kumar N P, Kishore M S, Chandran R G and Setlur A A 2008 *Electrochem. Soc. Trans.* **16** (30) 57–64
- [21] Kishore M S, Kumar N P, Chandran R G and Setlur A A 2010 *Electrochem. Solid-State Lett.* **13** J77–80
- [22] Shimomura Y, Kurushima T, Shigeiwa M and Kijima N 2008 *J. Electrochem. Soc.* **155** J45–9
- [23] Chen Y H, Liu B, Shi C S, Ren G H and Zimmerer G 2005 *Nucl. Instrum. Methods Phys. Res. A* **537** 31–5