

The Luminescence Properties of Sm^{3+} or Tb^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ Phosphors for X-ray Imaging

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Sm^{3+} or Tb^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ phosphors were synthesized by using a solid-state reaction method with Lutetium oxide, gallium oxide and aluminum oxide as host materials, samarium oxide and terbium oxide as doping materials. The mixed powders were sintered at 1500 °C for 5 hours by using a horizontal furnace. In this study, the structures of Sm^{3+} or Tb^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ phosphors were observed using an XRD (X-ray diffraction). The emission spectrum of Sm^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ phosphor shows 550~650 nm in wavelength due to $^4\text{G}_5$ to $^6\text{H}_j$. The measurement of emission spectrum of Tb^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ phosphor was confirmed to 450 to 650 nm wavelength due to $^5\text{D}_j$ to $^7\text{F}_j$. These phosphors have a sharp emission spectrum due to their physical properties of f-f transition. The $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ has a high effective Z-number ($Z_{\text{eff}} = 57$) which is sensitive to X-ray or γ -ray. Therefore Sm^{3+} or Tb^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ phosphors can be applied for diagnostic X-ray imaging in the medical field.

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I. INTRODUCTION

The phosphors have been attracted a lot of researchers because they can be applied to various fields such as medical imaging field, lamp, display, laser material, and nondestructive industry, *etc.* [1–6]. The purpose of this study is to develop new fluorescent material for diagnostic X-ray imaging such as digital radiography (DR) in the medical field. There are several required parameters; the most ideal phosphor generally requires strong X-ray absorption and high light emission [1,5].

To develop the phosphor materials for X-ray the properties of LuGAG:Tb, LuGAG:Sm phosphors were studied. The rare earth doped phosphors have received much interest from researchers in medical imaging, and various studies are underway. Because their high effective Z-number ($Z_{\text{eff}} = 57$) results a strong X-ray absorption. It can reduce the radiation dose of patients providing the high quality radiation imaging in a shorter time [1,

5]. $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ (7.0 g/cm³, $Z_{\text{eff}} = 57.36$) is compared with the most representative $\text{Gd}_2\text{O}_2\text{S}$ (7.32 g/cm³, $Z_{\text{eff}} = 59.37$). The effective Z-number and density is similar with reference phosphor. The host material, Lu_2O_3 can be applied to various kinds of activators because of a wide band gap between the conduction band and valence band. It has an excellent absorption characteristic for X-ray or γ -ray due to the high density and effective Z-number [1–6].

The rare earth phosphors have the *f-f* and *f-d* transitions. The *f-f* transitions are identified as several sharp emission lines in the spectrum [3,7]. Also, the rare earth phosphors with *f-f* transition have a long decay time, about several microseconds to milliseconds due to parity and spin forbidden such as Tb^{3+} , Gd^{3+} or Eu^{3+} [3,7]. A few rare earth phosphors also have an *f-d* transition results the broadband emission such as Ce^{3+} , Eu^{2+} or Pr^{3+} because of the *5d-4f* transition [3,7]. These transitions have a very fast decay time about a few nanoseconds due to the parity and spin allowed [3,7].

In this study, Sm^{3+} or Tb^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ phosphors were fabricated respectively for the first time

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as we know. These phosphors appear the physical properties of $f-f$ transition [3, 7]. The $f-f$ transition is defined the transfer of electrons from low energy of f -orbital to higher energy of f -orbital. The Sm^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ phosphor shows the high luminescence efficiency due to $^4\text{G}_j$ to $^6\text{H}_j$ transition. The Tb^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ phosphor also seems to be a effective luminescence material due to transition from $^5\text{D}_j$ to $^7\text{F}_j$. In order to confirm the above, excitation and emission wavelengths were analyzed by using an X-ray and UV light sources for analyzing the emission spectrum.

II. EXPERIMENTAL

To fabricate the Sm^{3+} or Tb^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ phosphors, the host materials Lu_2O_3 , Ga_2O_3 and Al_2O_3 and the activators Sm_2O_3 and Tb_4O_7 were synthesized by using a solid-state reaction method [3]. The mixed powders were sintered at 1500 °C for 5 hours. The sintered material was slowly cooled at room temperature. In order to confirm the properties of sintered phosphors, the crystalline of the powder was measured using X-ray diffraction (XRD-Panalytical X'Pert Pro). The X-ray tube used a Cu $\text{K}\alpha$ X-ray, which was scanned in the range of 10-80° (2θ) with a voltage of 40 kV and a current of 30 mA. The scanning speed and step size were 0.02° / sec and 0.025° [3]. The XRD measurement results were compared to the JCPDS card PDF #46-0448 ($\text{Gd}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$). To confirm the photoluminescence spectra and decay time, spectro fluorometer (Fluorolog-3) with a 450 W Xenon light source was used. To measure the X-ray induced luminescence spectra, an X-ray generator (65 kV, 1 mA) and a spectrophotometer (Ocean optics QE 65000 spectrometer) were used at room temperature.

III. RESULT AND DISCUSSION

Figure 1 shows the X-ray diffraction (XRD) patterns of $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}:\text{Sm}^{3+}$ and $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}:\text{Tb}^{3+}$ phosphors. The results of XRD were compared to JCPDS card number of #046-0448 ($\text{Gd}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$), and it was confirmed that the Miller index (h, k, l) coincided with the JCPDS value. There are no unnecessary peaks due to impurities, and no additional peaks due to the active material. The XRD analysis confirmed lattice constants and crystal structure.

Figure 2 shows the photoluminescence spectrum of Tb^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ phosphor. The emission spectrum of $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}:\text{Tb}^{3+}$ is caused by energy transitions from $^5\text{D}_j$ to $^7\text{F}_j$ [8–10]. A strong emission peak was observed at 546 nm wavelength due to the $^5\text{D}_4$ to $^7\text{F}_5$ energy transition by $\lambda_{\text{ex}} = 272\text{nm}$. Another main emission peaks were observed at $^5\text{D}_3 \rightarrow ^7\text{F}_5$ (417 nm),

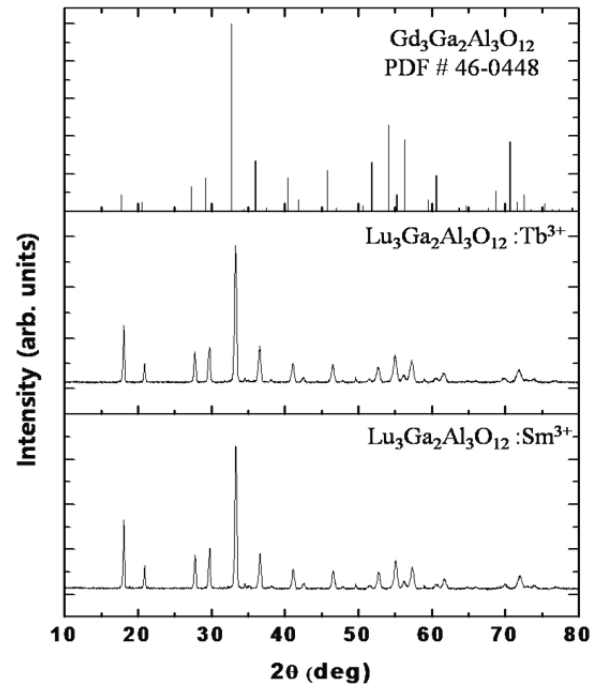


Fig. 1. The XRD patterns of Tb^{3+} or Sm^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ phosphors and standard PDF card of #46-0448 ($\text{Gd}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$).

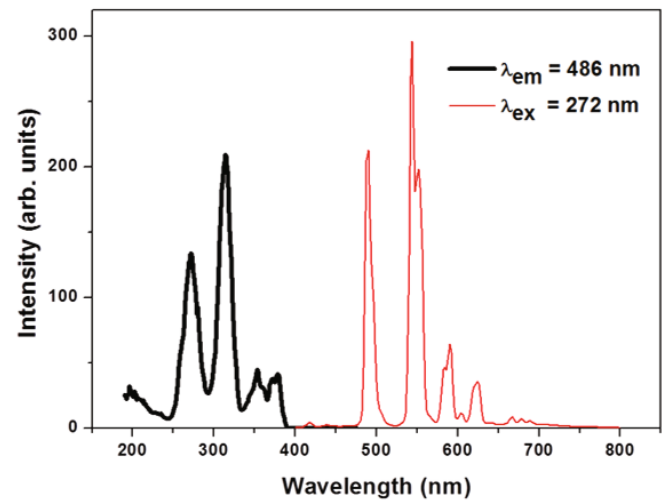


Fig. 2. (Color online) The excitation and emission spectra of Tb^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ phosphor by UV.

$^5\text{D}_3 \rightarrow ^7\text{F}_4$ (438 nm), $^5\text{D}_4 \rightarrow ^7\text{F}_6$ (486 nm), $^5\text{D}_4 \rightarrow ^7\text{F}_5$ (546 nm), $^5\text{D}_4 \rightarrow ^7\text{F}_4$ (587 nm), $^5\text{D}_4 \rightarrow ^7\text{F}_3$ (623 nm) and $^5\text{D}_4 \rightarrow ^7\text{F}_{2,1,0}$ (650 ~ 700 nm) transitions [8–10]. The excitation spectrum of $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}:\text{Tb}^{3+}$ phosphor was observed due to the excitation of $^7\text{F}_j$ to $^5\text{D}_j$ by $\lambda_{\text{em}} = 486\text{nm}$. The excitation wavelengths were appeared at 273 nm ($4f^8 \rightarrow 4f^75d^1$), 315 nm ($^7\text{F}_6 \rightarrow ^5\text{D}_0$), 354 nm ($^7\text{F}_6 \rightarrow ^5\text{D}_2$) and 380 nm ($^7\text{F}_6 \rightarrow ^5\text{G}_6$), respectively [8–10].

Figure 3 shows the emission spectrum of

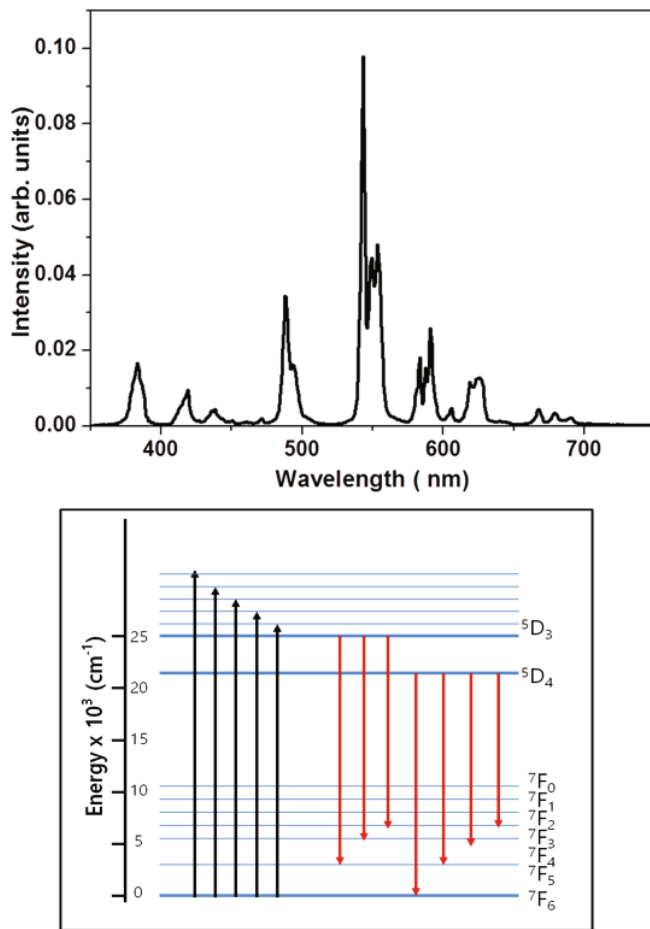


Fig. 3. (Color online) The emission spectrum of Tb^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ phosphor by X-ray and the energy level diagram of Tb^{3+} .

$\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$: Tb^{3+} phosphor by X-ray. The X-ray excited emission spectrum of $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$: Tb^{3+} phosphor is generated due to an energy transition of $^5\text{D}_j$ to $^7\text{F}_j$ like photoluminescence spectrum in Figure 2. Another emission peaks by X-ray were observed at $^5\text{D}_3 \rightarrow ^7\text{F}_6$ (382 nm), $^5\text{D}_3 \rightarrow ^7\text{F}_5$ (417 nm), $^5\text{D}_3 \rightarrow ^7\text{F}_4$ (438 nm), $^5\text{D}_4 \rightarrow ^7\text{F}_6$ (486 nm), $^5\text{D}_4 \rightarrow ^7\text{F}_5$ (546 nm), $^5\text{D}_4 \rightarrow ^7\text{F}_4$ (587 nm), $^5\text{D}_4 \rightarrow ^7\text{F}_3$ (623 nm) and $^5\text{D}_4 \rightarrow ^7\text{F}_{2,1,0}$ (650~700 nm) transitions [8–10]. The emission spectrum of $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$: Tb^{3+} phosphor by UV was well matched with X-ray excited emission spectrum of $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$: Tb^{3+} phosphor.

Figure 4 shows the decay time of $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$: Tb^{3+} phosphor for 272 nm excitation by UV. The measured data was fitted by an exponential function. The function can be written as $y = A_1^* \exp(-x/t_1) + y_0$ [11]. Where y and y_0 is luminescence intensity; A_1 is constant; x is time; t_1 is decay time [11]. The decay times of $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$: Tb^{3+} phosphor was measured 2.87 ms.

Figure 5 shows the emission an excitation spectrum of Sm^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ phosphor by UV. The excitation spectrum of $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$: Sm^{3+} phosphor

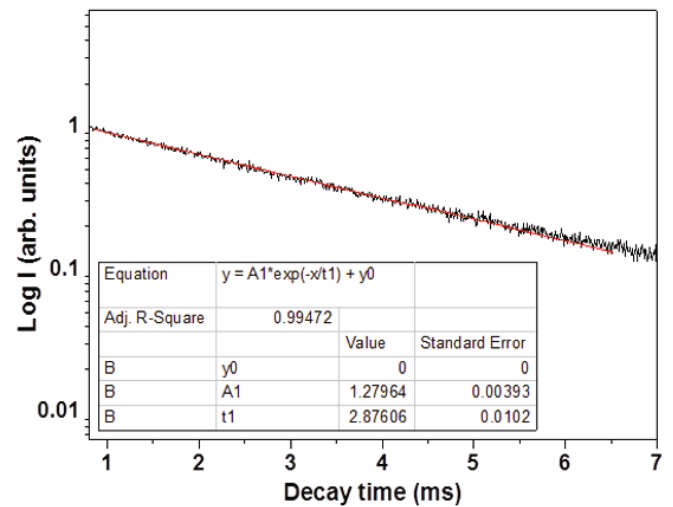


Fig. 4. (Color online) The decay time of Tb^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ phosphor by UV.

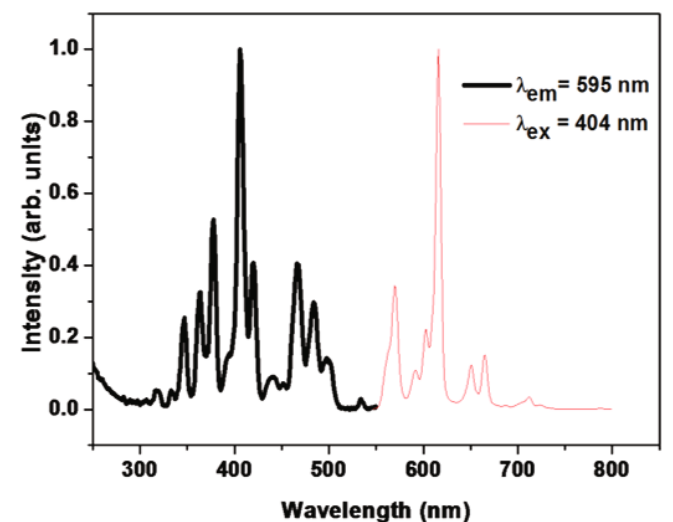


Fig. 5. (Color online) The excitation and emission spectra of Sm^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ phosphor by UV.

is caused by $^6\text{H}_{5/2} \rightarrow ^4\text{F}_{11/2}$ (318 nm), $^6\text{H}_{5/2} \rightarrow ^4\text{D}_{7/2}$ (332 nm), $^6\text{H}_{5/2} \rightarrow ^3\text{H}_{7/2}$ (346 nm), $^6\text{H}_{5/2} \rightarrow ^4\text{D}_{5/2}$ (363 nm), $^6\text{H}_{5/2} \rightarrow ^6\text{P}_{7/2}$ (377 nm), $^6\text{H}_{5/2} \rightarrow ^4\text{K}_{11/2}$ (405 nm), $^6\text{H}_{5/2} \rightarrow ^6\text{P}_{5/2} + ^4\text{M}_{19/2}$ (419 nm), $^6\text{H}_{5/2} \rightarrow ^4\text{G}_{9/2} + ^4\text{I}_{15/2}$ (441 nm), $^6\text{H}_{5/2} \rightarrow ^4\text{F}_{5/2} + ^4\text{I}_{13/2}$ (464 nm), $^6\text{H}_{5/2} \rightarrow ^4\text{I}_{11/2} + ^4\text{M}_{15/2}$ (484 nm), $^6\text{H}_{5/2} \rightarrow ^4\text{G}_{7/2}$ (499 nm) by $\lambda_{\text{em}} = 595\text{nm}$, respectively [12–15]. These sharp excitation peaks appear due to the f - f transitions of Sm^{3+} ion. The emission spectrum of $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$: Sm^{3+} phosphor is due to $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{5/2}$ (570 nm), $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{7/2}$ (600~620 nm), $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{9/2}$ (650 ~ 670 nm) and $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{11/2}$ (711 nm) energy transition by $\lambda_{\text{ex}} = 404\text{nm}$, respectively [12–15].

Figure 6 shows the emission spectrum of $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$: Sm^{3+} phosphor by X-ray and the

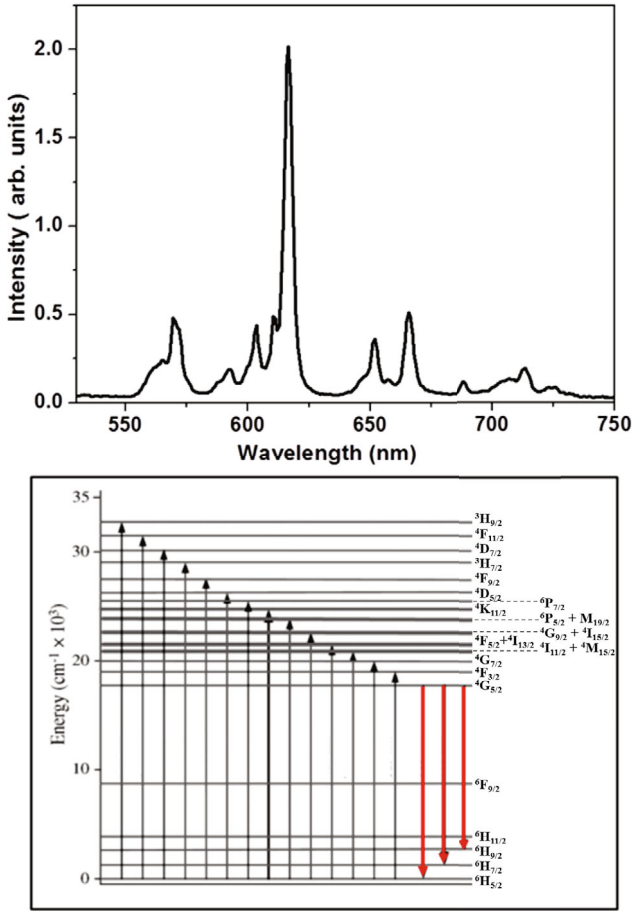


Fig. 6. (Color online) The emission spectrum of Sm^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ phosphor by X-ray and the energy level diagram of Sm^{3+} .

energy level diagram of Sm^{3+} . The X-ray excited emission spectrum of $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}:\text{Sm}^{3+}$ phosphor is due to an energy transition of $^4\text{G}_{5/2}$ to $^6\text{H}_j$, like photoluminescence spectrum in Fig. 5. The detailed emission peaks by X-ray were observed at $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{5/2}$ (570 nm), $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{7/2}$ (600 ~ 620 nm), $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{9/2}$ (650 ~ 670 nm) and $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{11/2}$ (711 nm) energy transitions [12–15]. The emission spectrum of $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}:\text{Sm}^{3+}$ phosphor by UV was well matched to X-ray excited emission spectrum of $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}:\text{Sm}^{3+}$ phosphor.

Figure 7 shows the decay times of $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}:\text{Sm}^{3+}$ phosphor for 404 nm excitation by UV. The measured data was fitted by an exponential function. The function can be written as $y = A_1^* \exp(-x/t_1) + y_0$ [11]. The decay times of $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}:\text{Sm}^{3+}$ phosphor was measured 2.11 ms.

IV. CONCLUSIONS

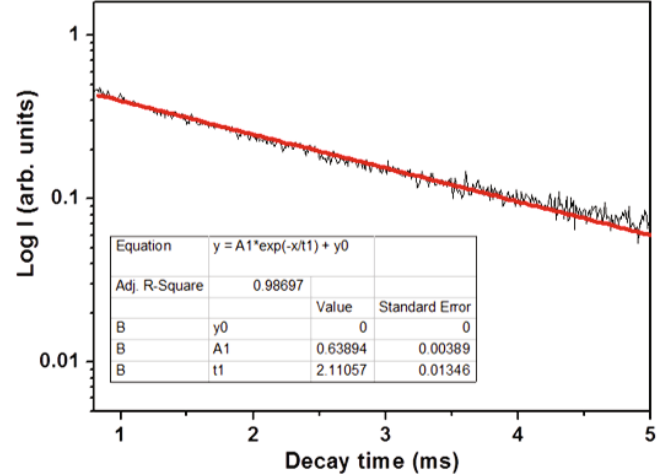


Fig. 7. (Color online) The decay times of Sm^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ phosphor by UV.

In this study, Sm^{3+} or Tb^{3+} -doped $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ phosphors were fabricated by solid-state reaction and the XRD of the $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}:\text{Sm}^{3+}$ and $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}:\text{Tb}^{3+}$ phosphors were analyzed. The results of XRD of $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$ were compared to JCPDS card number of #046-0448 ($\text{Gd}_3\text{Ga}_2\text{Al}_3\text{O}_{12}$), and it was coincided with the JCPDS value. The emission spectra by X-ray and UV of the $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}:\text{Sm}^{3+}$ and $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}:\text{Tb}^{3+}$ phosphors were also analyzed. As a result, it was confirmed that the emission spectra of $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}:\text{Sm}^{3+}$ and $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}:\text{Tb}^{3+}$ phosphors were a well match to those of UV and X-ray, respectively. The decay times of $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}:\text{Sm}^{3+}$ and $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}:\text{Tb}^{3+}$ phosphors were measured about several milliseconds, respectively. The $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}:\text{Tb}^{3+}$ and $\text{Lu}_3\text{Ga}_2\text{Al}_3\text{O}_{12}:\text{Sm}^{3+}$ phosphors have a high atomic number due to the rare earth element. It has an excellent absorption coefficient for X-ray or γ -ray. Therefore it can be applied for the diagnostic X-ray imaging in the medical field because of their high detection efficiency for X-ray and γ -ray [3].

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