

## Supporting Information

### **Eu<sub>0.56</sub>Ta<sub>2</sub>O<sub>7</sub>: A New Nanosheet Phosphor with the High Intrananosheet-Site Photoactivator Concentration**

**Tadashi C. Ozawa,<sup>\*†</sup> Katsutoshi Fukuda,<sup>†</sup> Kosho Akatsuka,<sup>†</sup> Yasuo  
Ebina,<sup>†</sup> Takayoshi Sasaki,<sup>†‡</sup> Keiji Kurashima,<sup>§</sup> and Kosuke Kosuda<sup>§</sup>**

<sup>†</sup> *Nanoscale Materials Center, National Institute for Materials Science, 1-1  
Namiki, Tsukuba, Ibaraki, 305-0044, Japan*

<sup>‡</sup> *International Center for Materials Nanoarchitectonics, National Institute  
for Materials Science, 1-1 Namiki, Tsukuba, Ibaraki, 305-0044, Japan*

<sup>§</sup> *Materials Analysis Station, National Institute for Materials Science, 1-1  
Namiki, Tsukuba, Ibaraki, 305-0044, Japan*

#### ***Experimental Procedures***

**Materials.** The first precursor Li<sub>2</sub>Eu<sub>0.56</sub>Ta<sub>2</sub>O<sub>7</sub> was prepared by the solid state reaction of 2:0.667:2 Li<sub>2</sub>CO<sub>3</sub>:Eu<sub>2</sub>O<sub>3</sub>:Ta<sub>2</sub>O<sub>5</sub> with a 10% excess of Li<sub>2</sub>CO<sub>3</sub>, which compensate for its loss by evaporation during the heating reaction. This ratio of starting materials was used because the reaction was originally intended to synthesize Li<sub>2</sub>Eu<sub>2/3</sub>Ta<sub>2</sub>O<sub>7</sub> which is the exact Eu analog of Li<sub>2</sub>La<sub>2/3</sub>Ta<sub>2</sub>O<sub>7</sub> without any extra vacancies in the Ln site.<sup>1</sup> In addition, this ratio of the starting materials yielded the least amount of impurity phases. These materials are mixed thoroughly in an agate mortar, placed in a capped Pt crucible which is enclosed in a capped alumina crucible, heated up to 1600 °C at 30 °C/min, dwelled for 1 min, and furnace-cooled to room temperature.

Second,  $\text{Li}_2\text{Eu}_{0.56}\text{Ta}_2\text{O}_7$  was protonated by a method similar to that for  $\text{H}_2\text{La}_{2/3}\text{Ta}_2\text{O}_7$  and  $\text{H}_2\text{SrTa}_2\text{O}_7$  with some modifications.<sup>1-5</sup> 1.9 g of  $\text{Li}_2\text{Eu}_{0.56}\text{Ta}_2\text{O}_7$  was ground and reacted with 190 mL of 2 M  $\text{HNO}_3$  for 3 days at room temperature under vigorous shaking in order to exchange  $\text{Li}^+$  of with  $\text{H}^+$ .

Finally, 1.5 g of this protonated precursor was reacted with 375 mL of an approximately 3-fold excess TBAOH aqueous solution. After 1 week of vigorous shaking, a translucent white colloidal nanosheet suspension was obtained. This nanosheet suspension was centrifuged at 2500 rpm for 5 min in order to separate the unexfoliated residue from the nanosheet suspension for the characterizations.

**Characterizations.** The elemental compositions of the bulk precursors were analyzed by EPMA on JEOL JXA-8500F using the acceleration voltage of 15 kV. The powder XRD (X-ray diffraction) patterns of the bulk precursors were obtained using  $\text{Cu } K\alpha$  radiation on a Rigaku RINT2200V/PC diffractometer. The diffraction peaks were indexed, and the lattice parameters were refined using APPLEMAN software.<sup>6</sup> The structural data of the previously reported  $\text{Li}_2\text{La}_{2/3}\text{Ta}_2\text{O}_7$  was used as starting parameters for these processes.<sup>1</sup> TEM and SAED were performed on a JEOL JEM-1010 transmission electron microscope at an acceleration voltage of 100 kV. The specimen for this characterization was prepared by dropping and drying diluted nanosheet suspension on a carbon microgrid. The in-plane XRD pattern of  $\text{Eu}_{0.56}\text{Ta}_2\text{O}_7$  nanosheets, deposited on a Si substrate by the Langmuir-Blodgett (LB) method,<sup>7</sup> was obtained using the synchrotron radiation ( $\lambda = 0.11973(9)$  nm) of Photon Factory BL-3A at High Energy Accelerator Research Organization (KEK). The morphology analysis was performed by AFM using a Seiko Instruments SPA-400 AFM system with a Si tip cantilever ( $20 \text{ N m}^{-1}$ ) in the DFM mode. Finally, photoluminescence excitation and emission properties of  $\text{Eu}_{0.56}\text{Ta}_2\text{O}_7$  nanosheets were characterized on a HITACHI F-4500 fluorescence

spectrometer at room temperature. The excitation spectrum was corrected for the spectral distribution of the lamp intensity by the Rhodamine B method, and the emission spectrum was corrected for the spectral response of the instrument using a substandard light source.

## References and Notes

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