## Achieving Dynamic Quintuple-mode Luminescence in Ca<sub>3</sub>Ti<sub>2</sub>O<sub>7</sub>: Pr<sup>3+</sup>,

## Er<sup>3+</sup> Phosphor for Anti-counterfeiting Applications

Jian Zhang<sup>a, †</sup>, Xin You<sup>a, †</sup>, Ting Wang<sup>b,\*</sup>, Yiyu Cai<sup>a</sup>, Chao Wang<sup>a</sup>, Xin Li<sup>a</sup>, Zhichao Liu<sup>a</sup>, Heng Dai<sup>a</sup>, Alexey Nikolaevich Yakovlev<sup>c</sup>, Xuhui Xu<sup>a</sup>, Jie Yu<sup>a,\*</sup>.

<sup>a</sup> Faculty of Materials Science and Engineering, Kunming University of Science and Technology, Kunming 650093, China.

<sup>b</sup> College of Materials and Chemistry & Chemical Engineering, Chengdu University of Technology, Chengdu 610059, China.

<sup>c</sup> T.F. Gorbachev Kuzbass State Technical University, Kemerovo 650000, Russia.

## **Corresponding Authors**

E-mail addresses: yujie@kust.edu.cn (J. Yu), wangtkm@foxmail.com (T. Wang).



**Figure S1.** The XRD patterns of (a)  $Ca_3Ti_2O_7$ :  $xPr^{3+}$  (x = 0.1%, 0.2%, 0.25%, 0.5%, 0.75% and 1%) and (b)  $Ca_3Ti_2O_7$ : 1%Er<sup>3+</sup> phosphors.



Figure S2. The EDS spectrum of representative Ca<sub>3</sub>Ti<sub>2</sub>O<sub>7</sub>: 0.25%Pr<sup>3+</sup>, 1%Er<sup>3+</sup>.



Figure S3. The PL spectra of Ca<sub>3</sub>Ti<sub>2</sub>O<sub>7</sub>:  $xPr^{3+}$  (x = 0.1%, 0.2%, 0.25%, 0.5%, 0.75% and 1%).



Figure S4. (a) The LPL decay curves and (b) TL curves of Ca<sub>3</sub>Ti<sub>2</sub>O<sub>7</sub>:  $xPr^{3+}$  (x = 0.1%, 0.2%, 0.25%, 0.5%, 0.75% and 1%).



Figure S5. (a) The LPL decay curves and (b) TL curves of  $Ca_3Ti_2O_7$ : 0.25%  $Pr^{3+}$  and  $Ca_3Ti_2O_7$ : 0.25%  $Pr^{3+}$ , 1%  $Er^{3+}$ .



**Figure S6.** The UCL spectra of Ca<sub>3</sub>Ti<sub>2</sub>O<sub>7</sub>: 0.25%Pr<sup>3+</sup>, 1%Er<sup>3+</sup> under 980 nm (1.5 W) laser with the irradiation time prolonged to 21 s.



**Figure S7.** The TL curves of  $Ca_3Ti_2O_7$ : 0.25%  $Pr^{3+}$ , 1%  $Er^{3+}$  treated with different pump power of 980 nm laser. The phosphors are charged by UV irradiation for 5 mins.



Figure S8. The schematic diagram of MS-T3001 multifunctional material surface property tester cooperated with an optical fiber.



**Figure S9.** The TL curves of Ca<sub>3</sub>Ti<sub>2</sub>O<sub>7</sub>: 0.25%Pr<sup>3+</sup>, 1%Er<sup>3+</sup> with UV irradiation for 5 mins or heating treatment at 400 K for 5 mins.

The ML composited films were fabricated by uniformly mixing the sifted powders (600 mesh size) with the PDMS at the weight ratio of 1:3, followed by transferring the mixture into cylindrical mold. Finally, the ML films were successfully shaped in the drying oven at 340 K for 3 h.



Figure S10. (a) The PL, LPL, PSL, ML spectra of  $Ca_3Ti_2O_7$ : 0.25%  $Pr^{3+}$ , (b) The PL and UCL spectra of  $Ca_3Ti_2O_7$ : 1%  $Er^{3+}$ .