Aliovalent co-doping and annealing effect on photoluminescence and scintillation properties of $Lu_3Al_5O_{12}$: Ce^{3+} epitaxial films

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Outline

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- Results & Discussion
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  - PL Excitation & Emission
  - PL Decay
  - Radioluminescence
  - Alpha Decay
- Conclusions

Motivation

- Scintillation materials have thundering applications in the field of high-energy physics, medical imaging, geological exploration, homeland security *etc*.

- Ce^{3+} doped garnets are good scintillators due to their less point defects and traps, large band gap, high chemical and thermal stability, high density, broad transmission range.

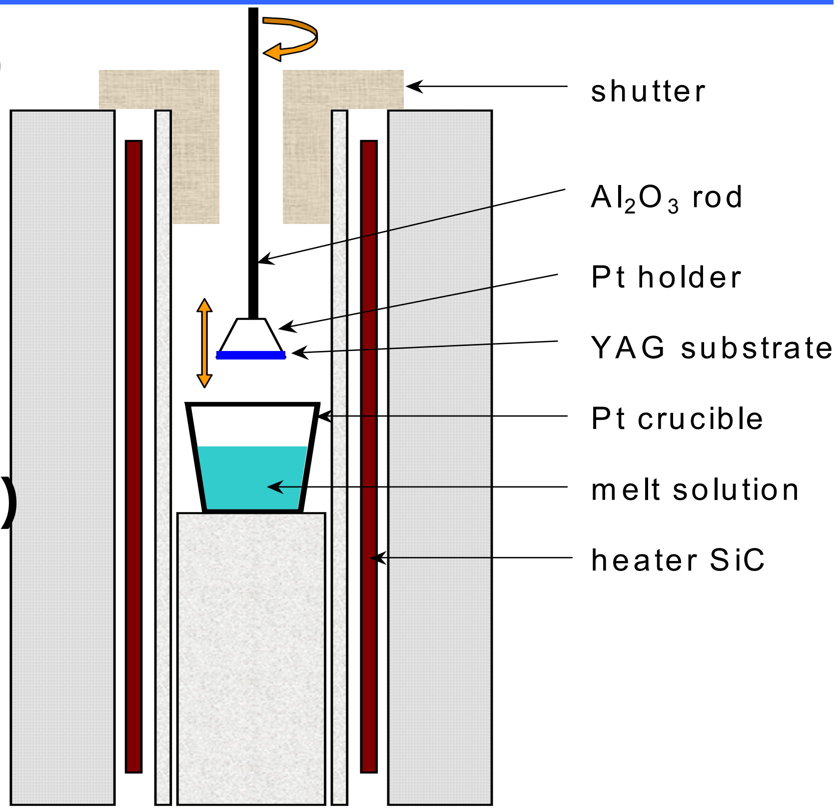
- The aliovalent co-dopants (Mg^{2+}, Ca^{2+} *etc.*) could alter the point defect structure, reduces the rise and decay times, and suppresses the charge carrier trapping on defect sites. The Si^{4+} co-doping would give interesting results.

- Annealing the Mg^{2+}/Ca^{2+} rich Ce^{3+} doped garnets in reducing atmosphere at higher temperatures would give exciting results.

- The liquid phase epitaxy is a unique technique for the growth of high quality single crystalline films with minimal concentration of the vacancy- and antisite-related defects.

Experimental work

- Growth technique: **Liquid phase epitaxy (LPE)**
- Used fluxes: **$BaO – B_2O_3 – BaF_2$**
- Growth temperature: **~1030 °C**
- Growth rate: **~0.12 μm/min**
- Composition» **$Ce_{0.02}Lu_{2.98}Al_5O_{12}$:X**
  [X=Mg(0-7000 ppm), Ca (0-6000 ppm), Ca+Si)
- Thickness of grown films: **30-12 μm**



Absorption: Specord 250, range: 190-1100 nm

Excitation & Emission: Horiba JY Fluoromax 3

Radioluminescence: Custom made spectrofluorometer 5000M, Horiba Jobin Yvon using an X-ray tube (10 kV, 50 mA)

PL Decay: Spectrofluorometer 5000M , nanoLED, Hamamatsu R7207-01 and Tektronix TDS3052C digital Phosphor Oscilloscope

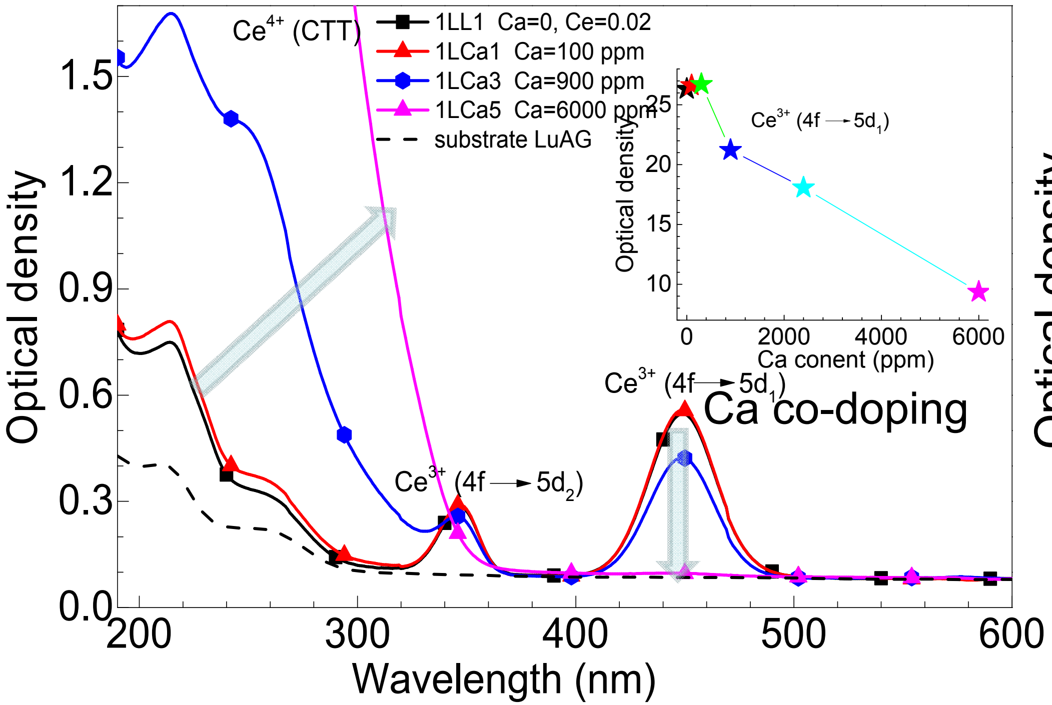
Alpha decay: Source- ^{241}Am, 5 mV

Annealing: 1100 °C, 10 hrs, reducing atmosphere Ar: 5%H_2

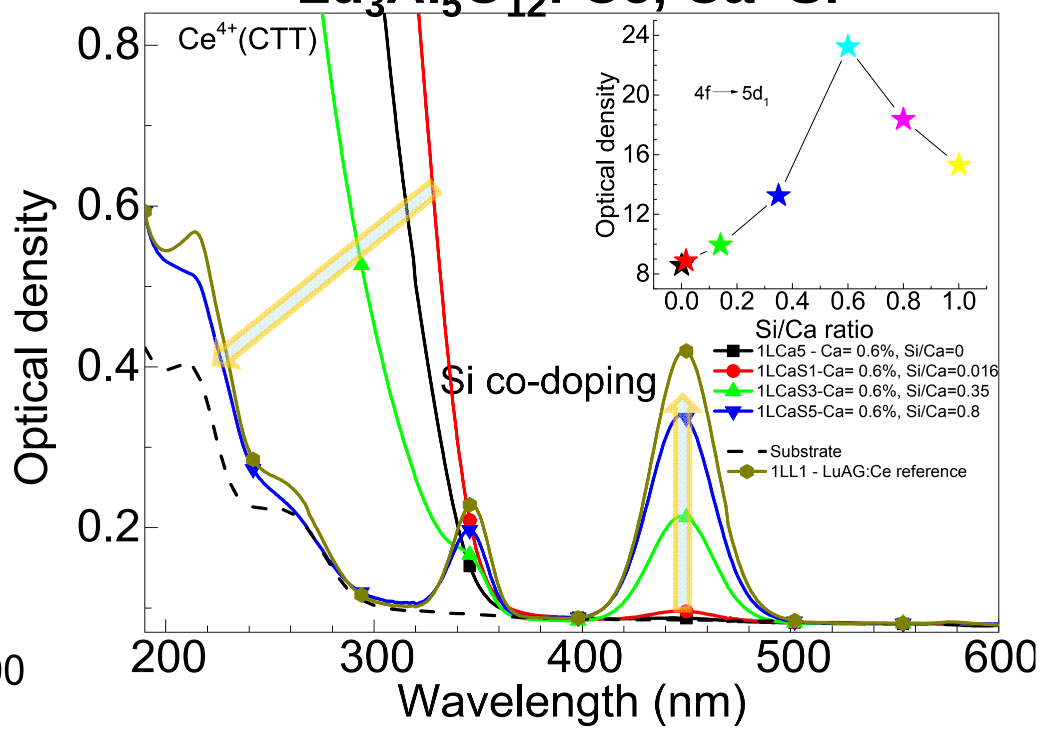
Results & Discussion

Absorption

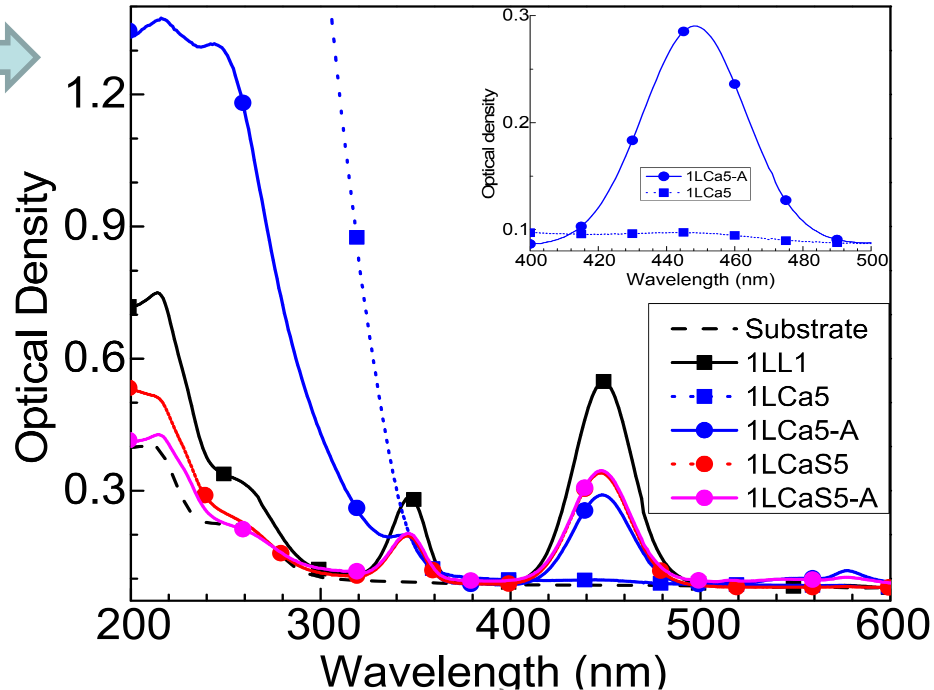
$Lu_3Al_5O_{12}$: Ce, Ca



$Lu_3Al_5O_{12}$: Ce, Ca+Si



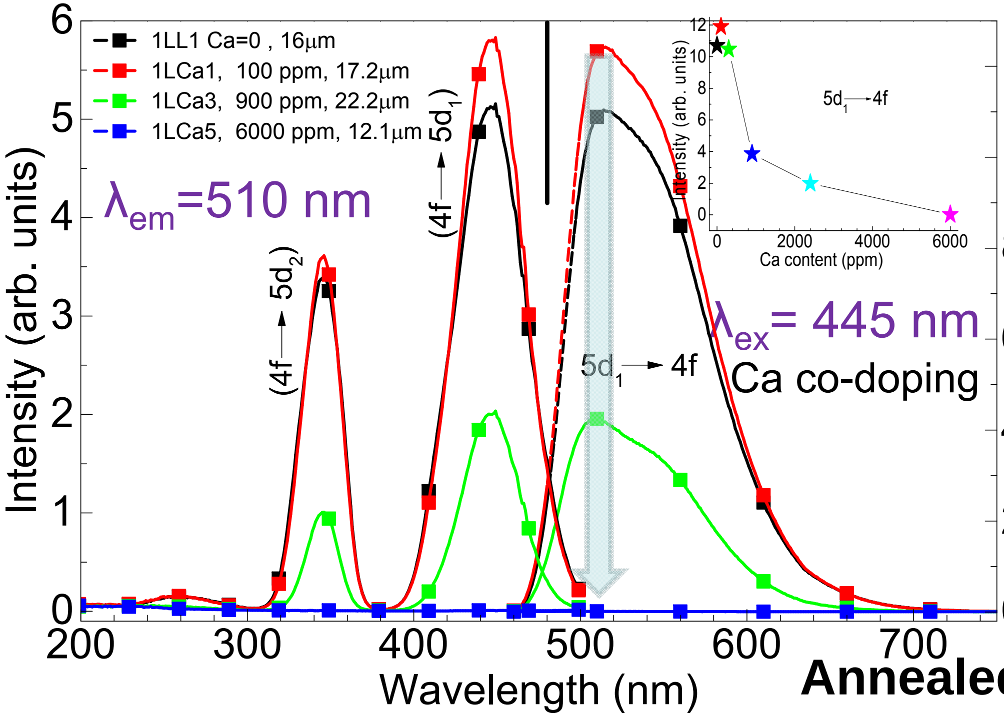
Annealed samples



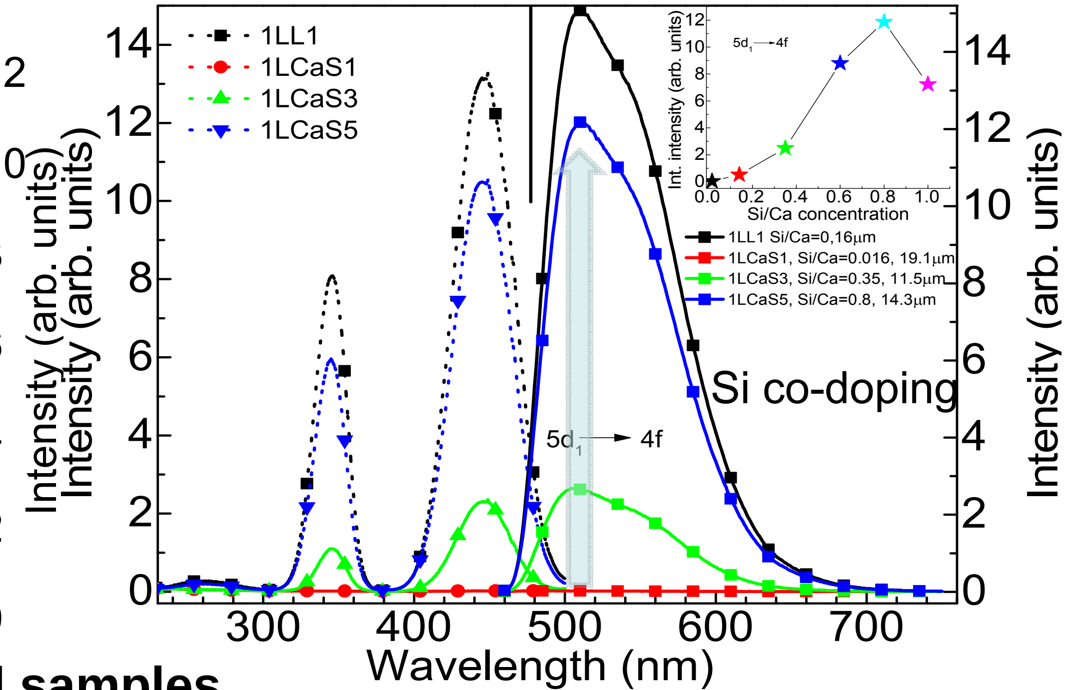
- The charge transfer transition (CTT): O^{2-} (2p) → Ce^{4+} (4f)
- The diminishing in the visible region is due to valence change of Ce^{3+} into Ce^{4+}
- Similar trend was also observed in Mg^{2+} codoped LuAG films

Excitation & Emission

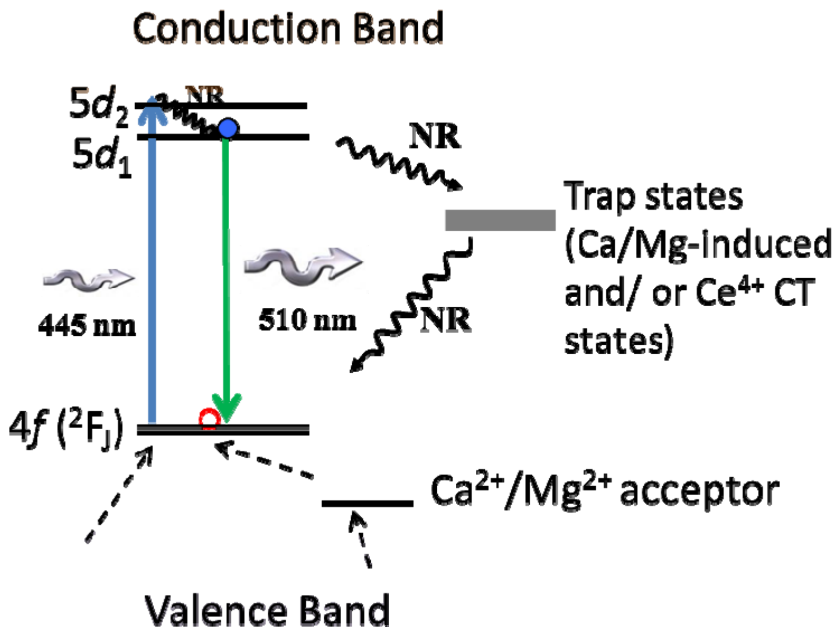
$Lu_3Al_5O_{12}$: Ce, Ca

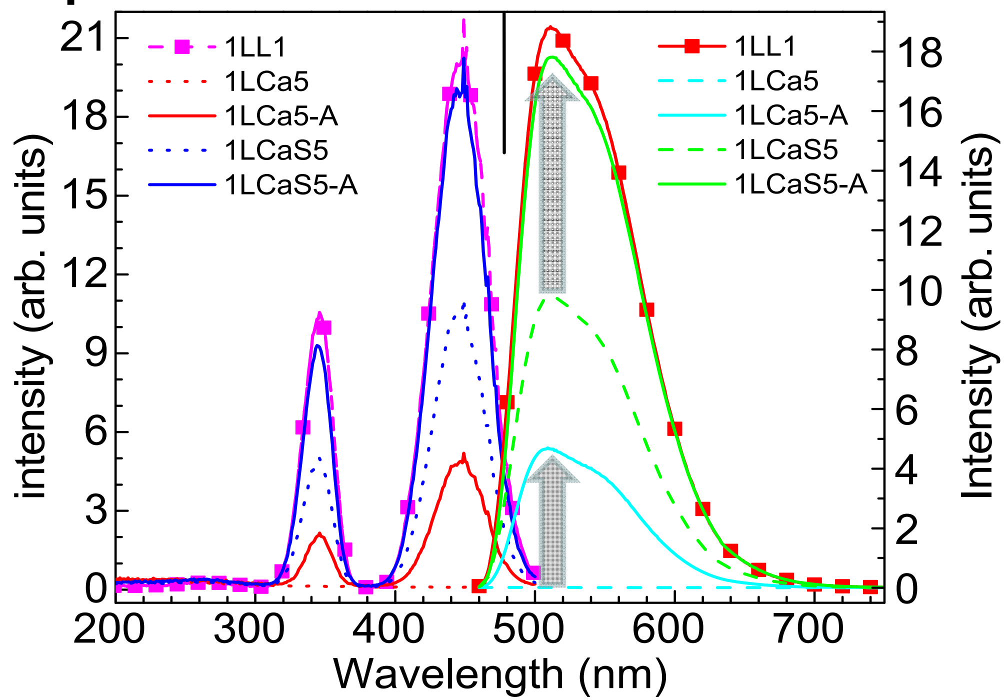


$Lu_3Al_5O_{12}$: Ce, Ca+Si



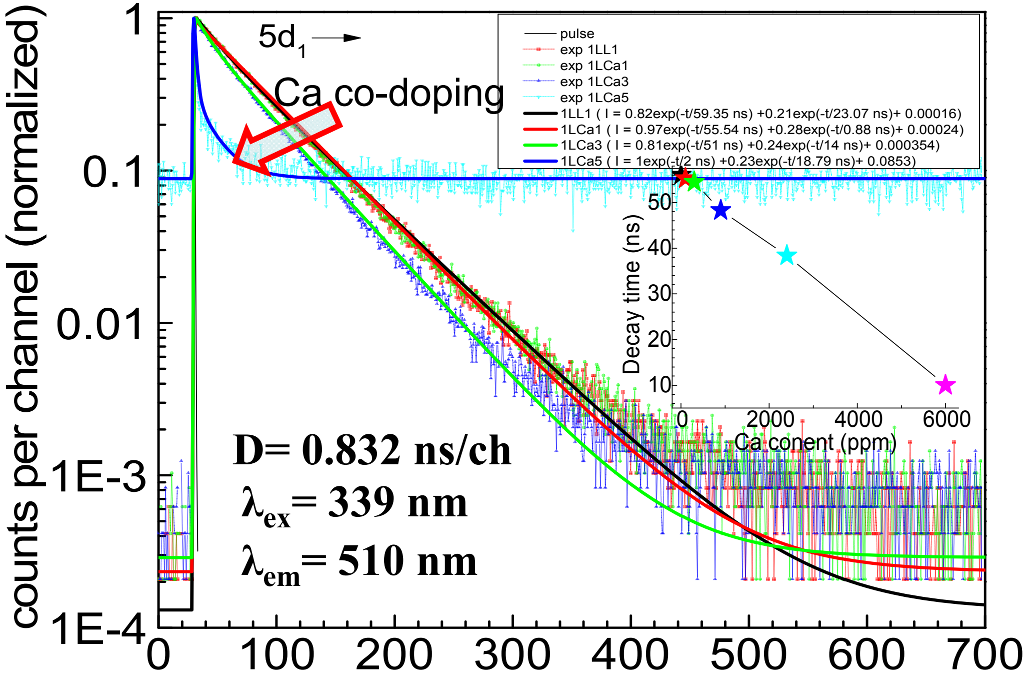
Annealed samples



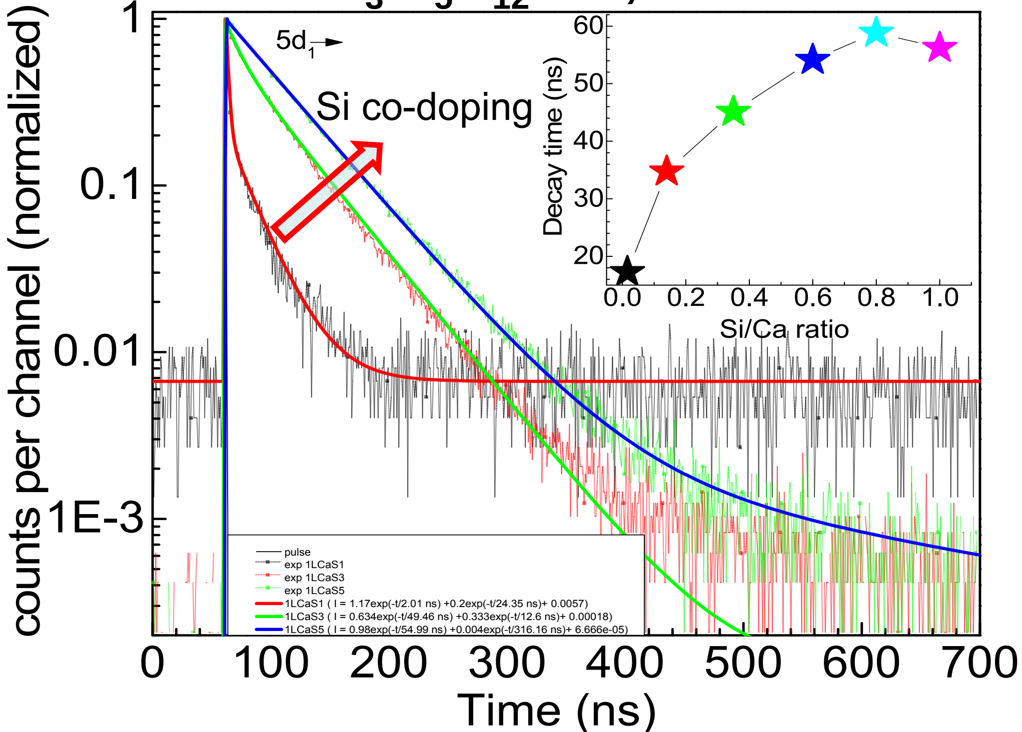


PL Decay

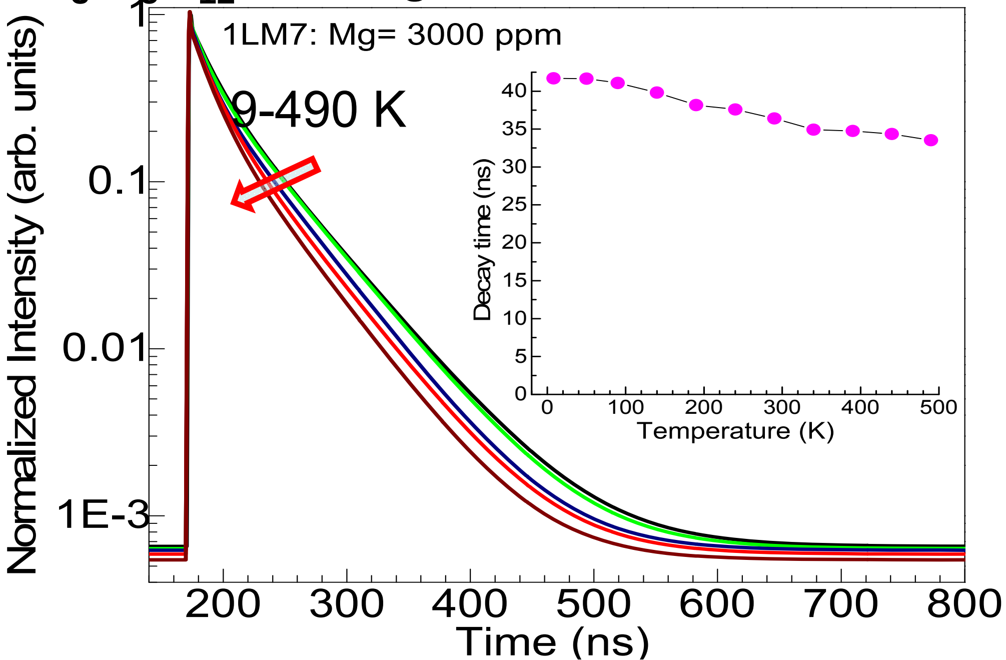
$Lu_3Al_5O_{12}$: Ce, Ca



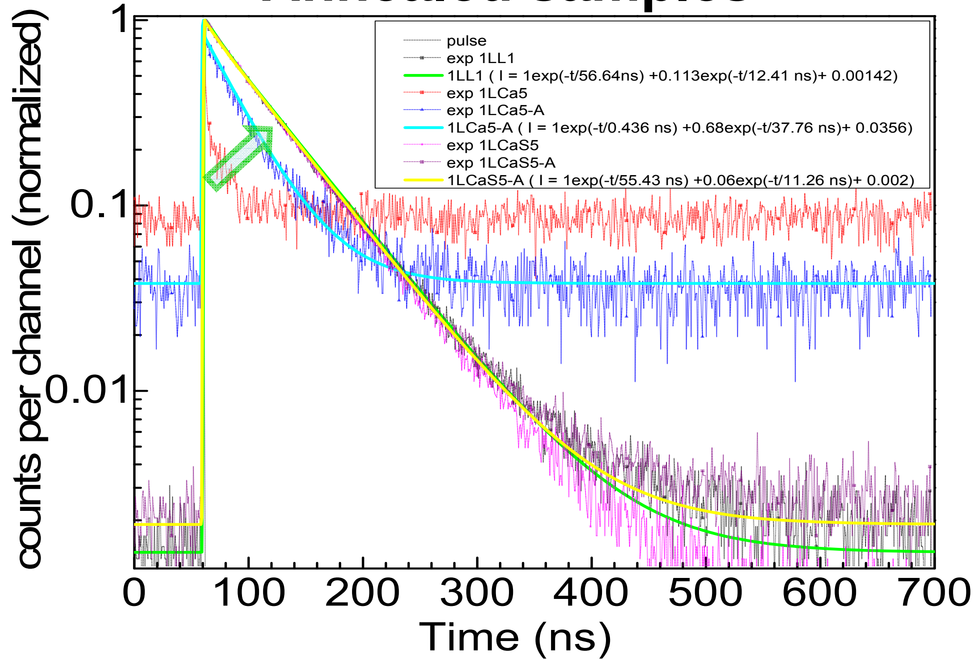
$Lu_3Al_5O_{12}$: Ce, Ca+ Si



$Lu_3Al_5O_{12}$: Ce, Mg

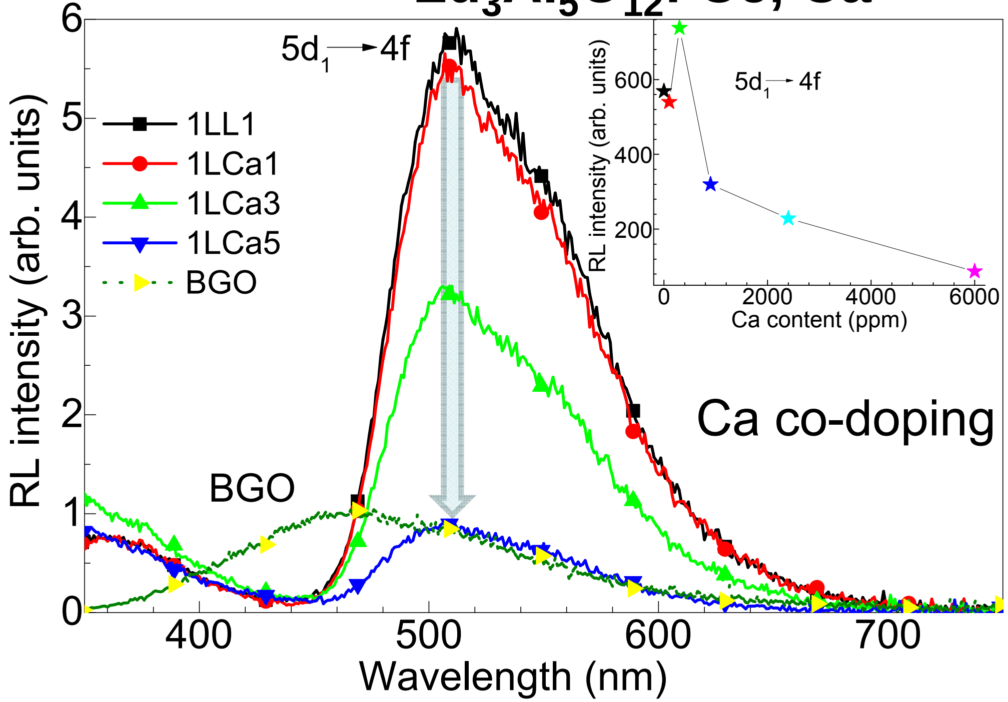


Annealed samples

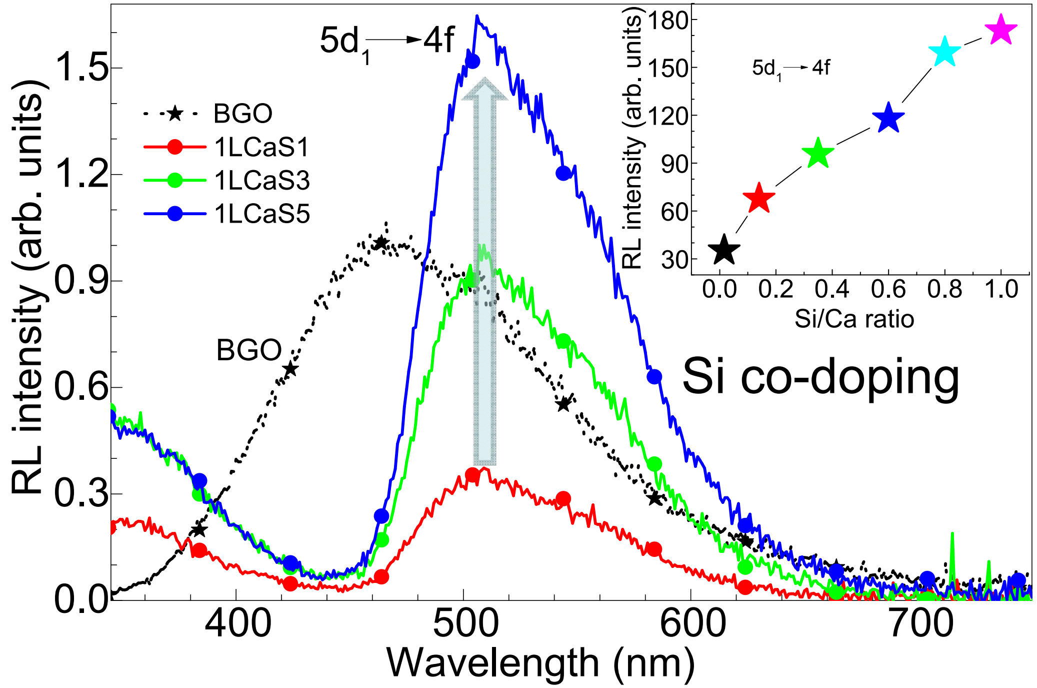


Radioluminescence

$Lu_3Al_5O_{12}$: Ce, Ca

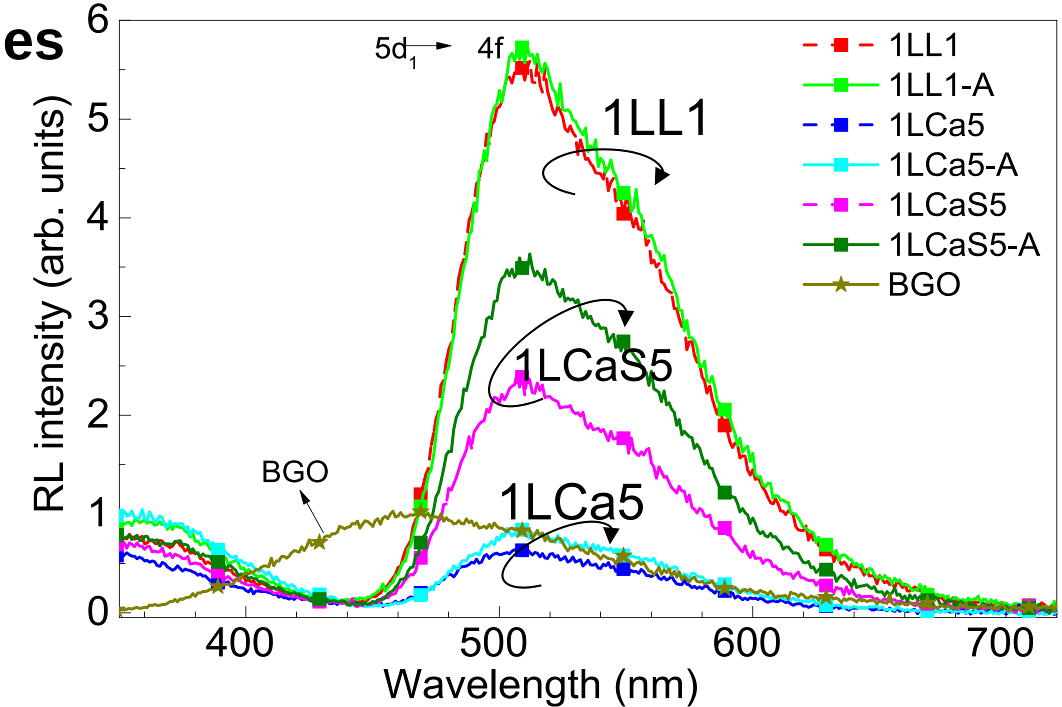


$Lu_3Al_5O_{12}$: Ce, Ca+Si



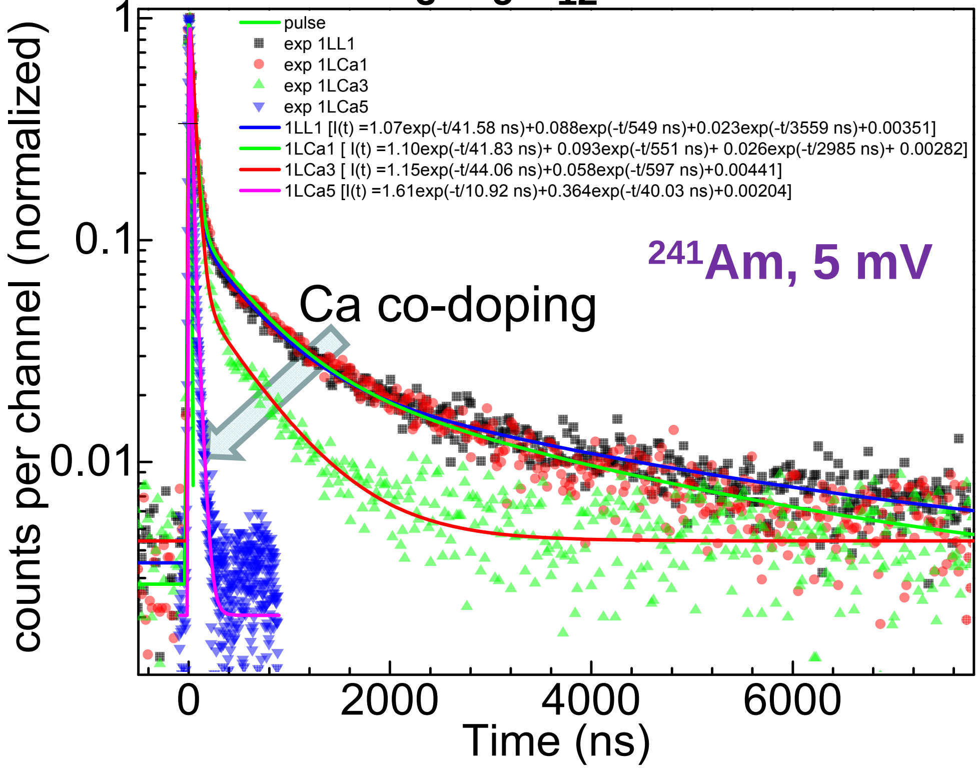
Annealed samples

Power = 10 kV, 50 mA

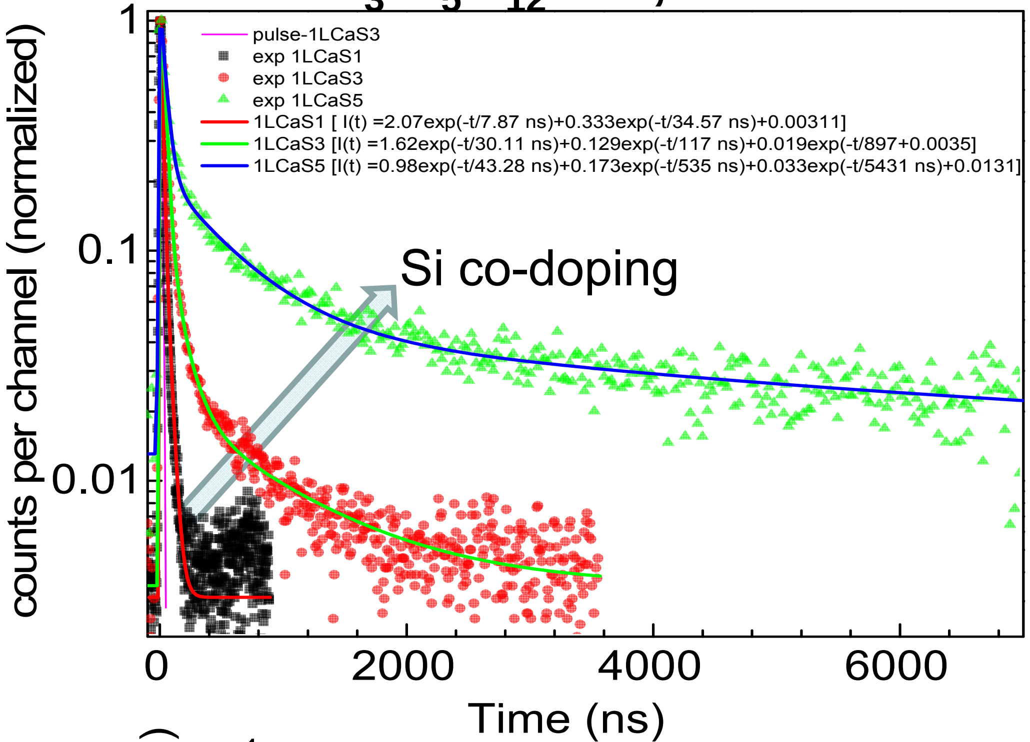


Alpha decay

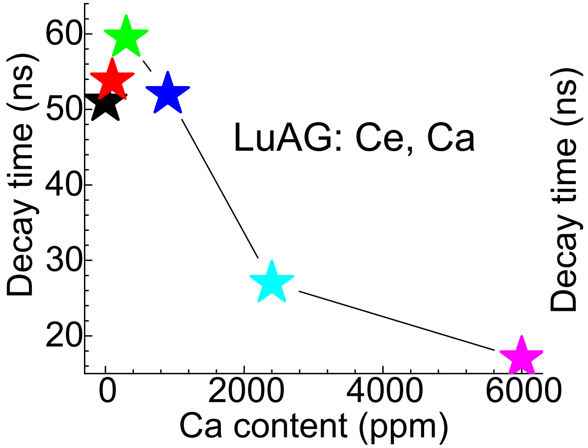
$Lu_3Al_5O_{12}$: Ce, Ca

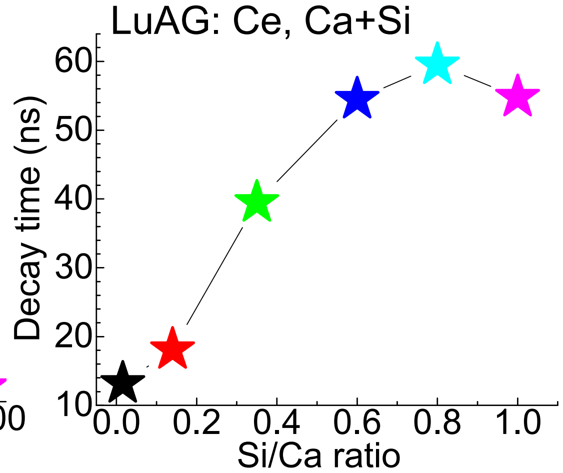


$Lu_3Al_5O_{12}$: Ce, Ca+Si

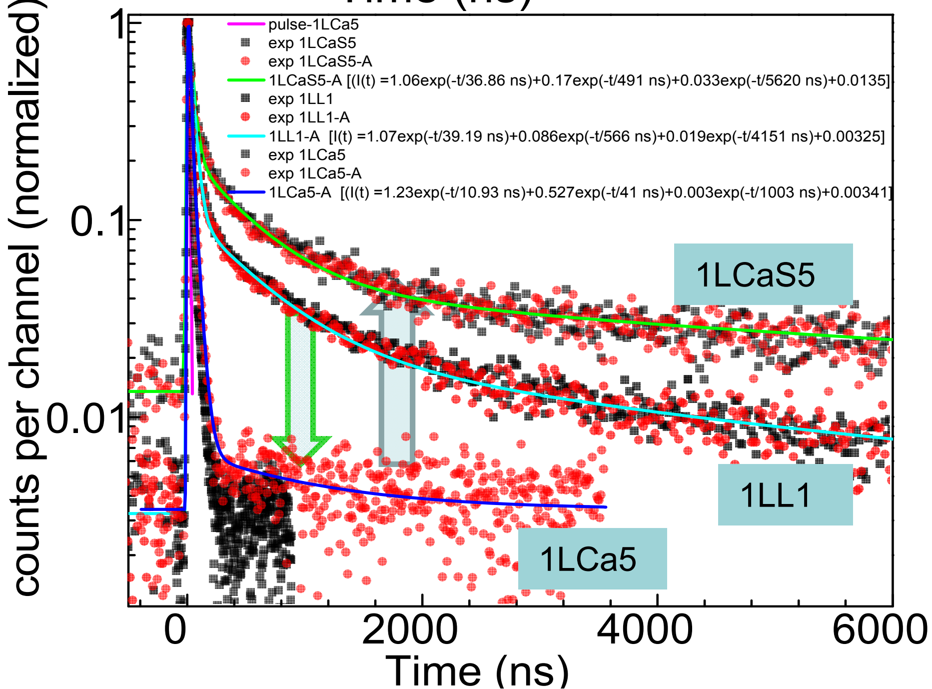


Mean decay time





Annealed samples



Conclusions

- Aliovalent (Mg^{2+}, Ca^{2+}, Si^{4+}) co-doped $Lu_3Al_5O_{12}$: Ce^{3+} epitaxial films have been prepared by liquid phase epitaxy technique.

- The absorption results indicate that at high Mg^{2+} or Ca^{2+} co-doping, all the Ce^{3+} ions tend to convert into Ce^{4+} ions due to change of valence. Upon adding Si^{4+} ions into the highly Ca^{2+} co-doped $Lu_3Al_5O_{12}$: Ce^{3+} films, the Ce^{3+} ions have been re-established due to charge compensation.

- Intense visible emission is observed from low Mg^{2+} or Ca^{2+} co-doped $Lu_3Al_5O_{12}$: Ce^{3+} films, excited at 445 nm. At high Ca^{2+} co-doping, the emission gets quenched, but Si^{4+} addition could regenerate the visible emission.

- At low Mg^{2+} or Ca^{2+} content, the decay curves are found slightly non-exponential with mean decay time around 55 ns.

- After annealing the highly Ca^{2+} co-doped $Lu_3Al_5O_{12}$: Ce^{3+} films at 1100 ^{0}C in reducing atmosphere, part of the Ce^{4+} ions were converted into Ce^{3+} ions.

Thank you