Memory effect with model materials: YPO_4: Ce, Nd

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SUCCESS workshop 11-12 December 2012

Memory effect in scintillators…

Definition:

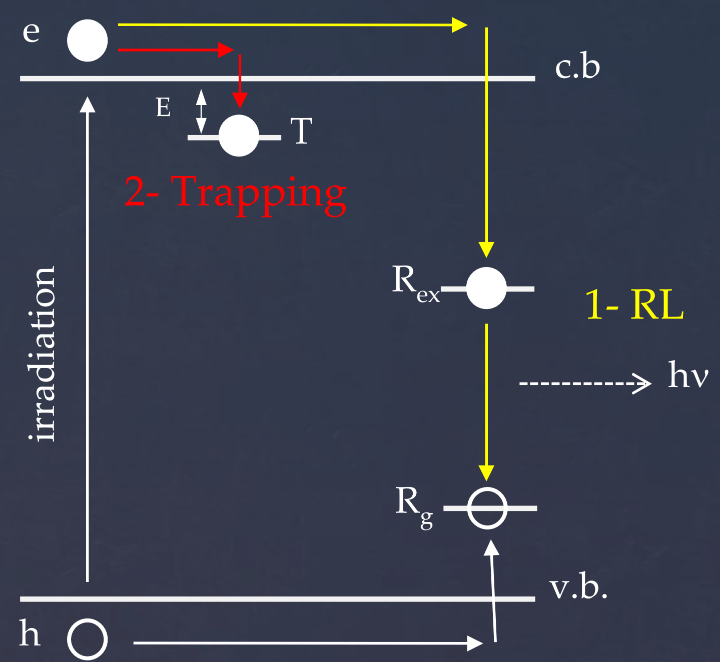
Radio-luminescence (RL) intensity increase with the accumulated dose
Also called: "bright burn", RL sensitization

Cause:

Progressive filling of traps present in the scintillator during irradiation

↓

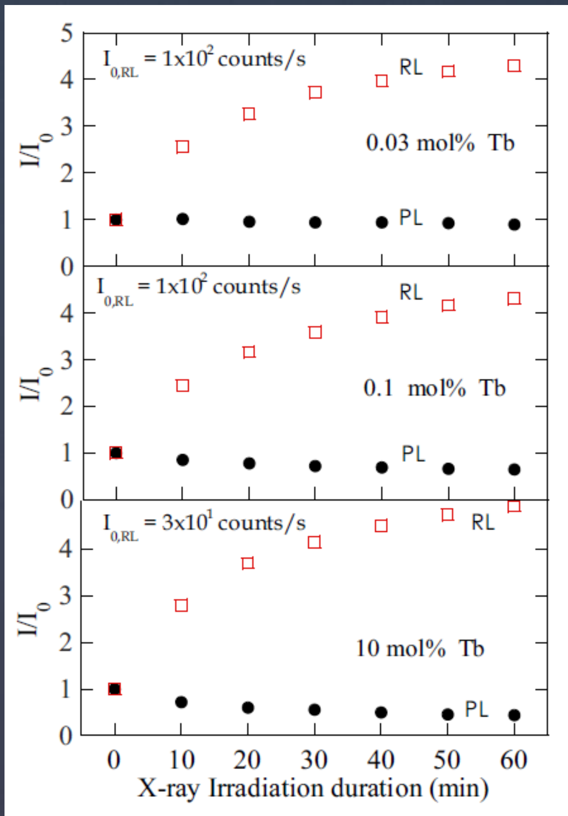
Increase of the radiative recombination probability of free carriers due to reduced competition between emission centres (1) and traps in carrier capture (2)



Memory effect may represent a problem in those applications which rely on consistent RL intensity as a function of the dose rate (e.g. CT, digital radiography, real time RL dosimetry …)

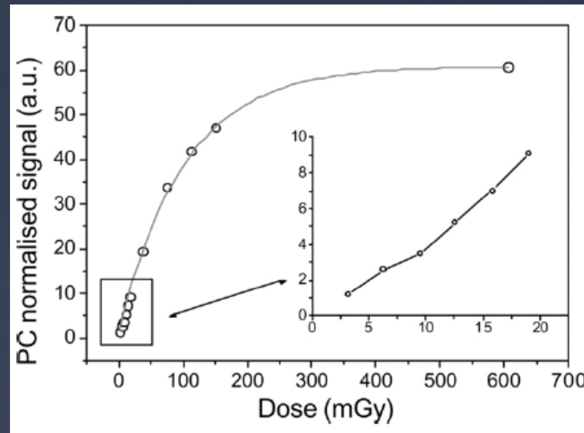
... and a few examples

Sol-gel SiO_2:Tb



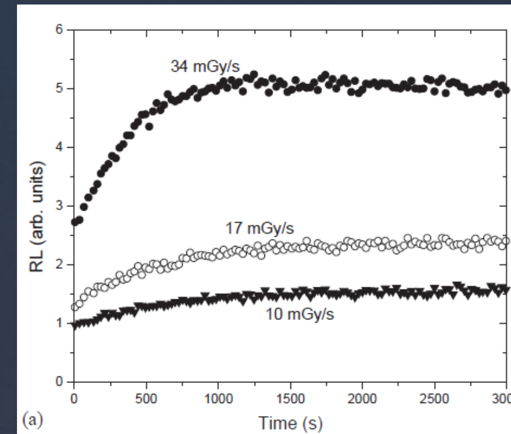
Fasoli *et al.* PSSc, 4 (2007) 1056

Diamond (photo-conductivity)



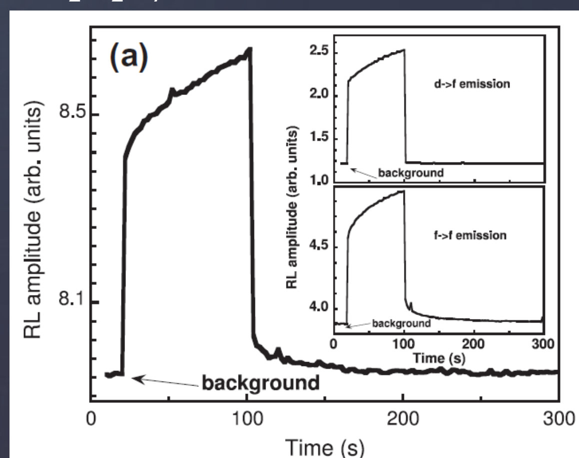
Manfredotti *et al.* Diam&Rel Mat, 13 (2004) 914

Al_2O_3:C



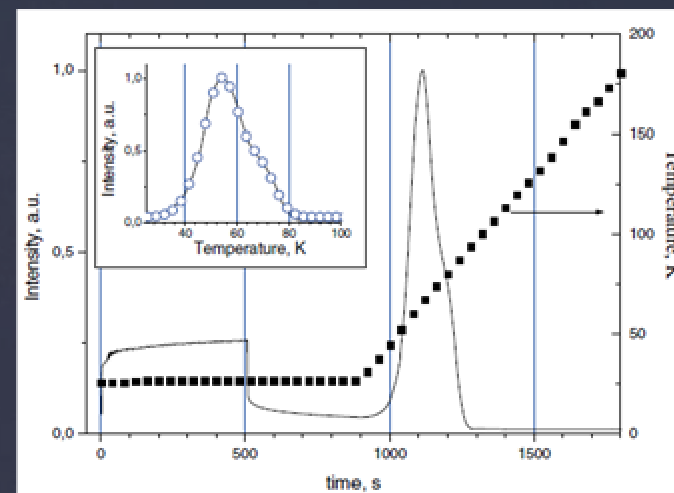
Polf, *et al*. Rad Meas, 38 (2004) 227

$Lu_2Si_2O_7$:Pr



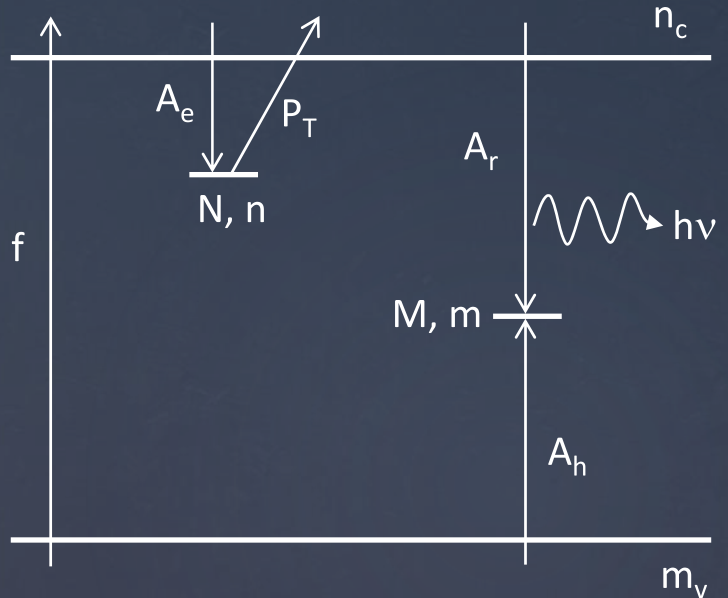
Mihokova *et al.* Opt Mater, 34 (2012) 872

$ZnMoO_4$



Spassky *et al.* PSSa, 206 (2009) 1579

Memory effect: model



$$\frac{dn_c}{dt} = f - n_c(N-n)A_e + nP_T - n_cA_rm$$

$$\frac{dn}{dt} = n_c(N-n)A_e - nP_T$$

$$\frac{dm_v}{dt} = f - m_v(M-m)A_h$$

$$\frac{dm}{dt} = m_v(M-m)A_h - n_cA_rm$$

$$n_c + n = m_v + m$$

$$I_{RL} \propto n_cA_rm \qquad P_T = s\,exp\left(-\frac{E}{kT}\right)$$

Where:

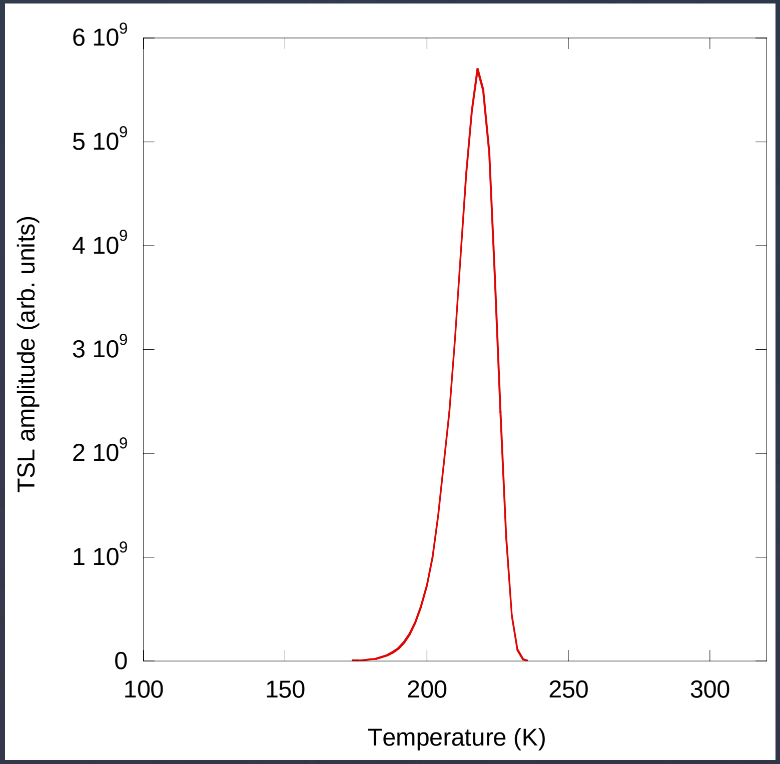
n, n_c : electron concentration (cm^{-3}) on traps and in the conduction band
m, m_v : hole concentration (cm^{-3}) on traps and in the valence band
M, N : hole and electron traps concentration (cm^{-3})
f : electron/hole pair creation rate (cm^{-3} s^{-1})
A_e, A_r and A_h : transition coefficients (cm^3 s^{-1})

Setting the parameters

Some parameter values are of difficult guessing.
Best strategy: change one parameter at a time while keeping the other fixed, thus allowing to obtain general trends on the parameters role in the obtained results

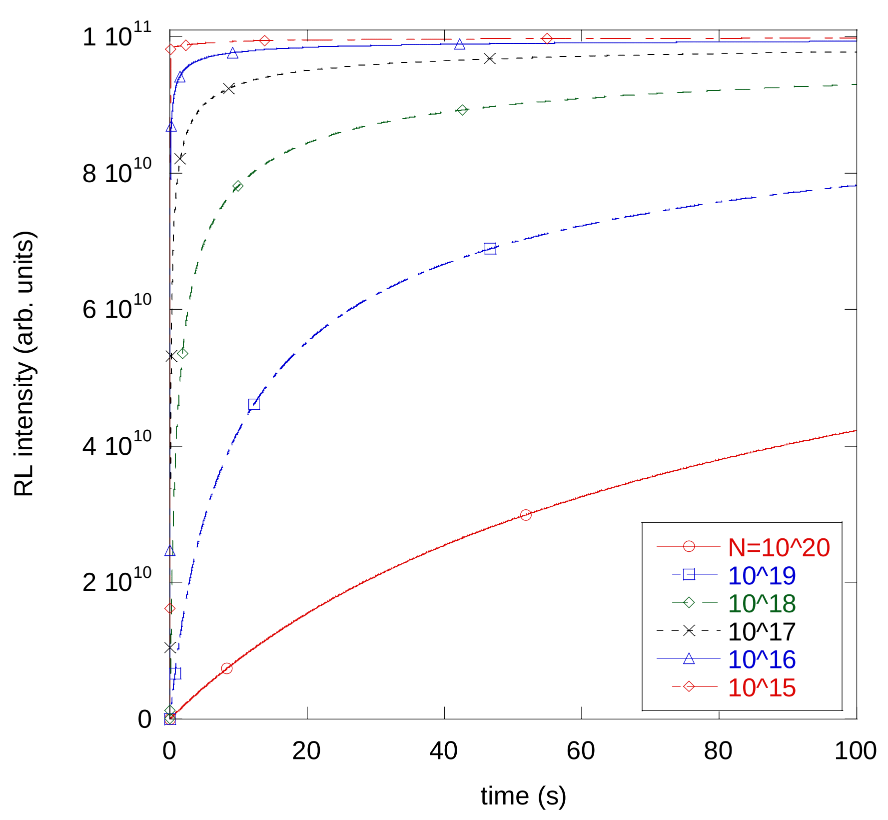
Parameter	Initial value	interval
f ($cm^{-3}s^{-1}$)	10^{11}	10^{8} - 10^{12}
M (cm^{-3})	10^{19}	-
N (cm^{-3})	10^{16}	10^{14} - 10^{20}
A_e (cm^3s^{-1})	10^{-15}	10^{-9} - 10^{-17}
A_r (cm^3s^{-1})	10^{-8}	-
A_h (cm^3s^{-1})	10^{-7}	-
E (eV)	0.6	-
s (s^{-1})	10^{12}	-
T (K)	10	10 - 300

Simulated glow curve obtained with the trap parameter listed in the table.
Heating rate 0.1 K/s

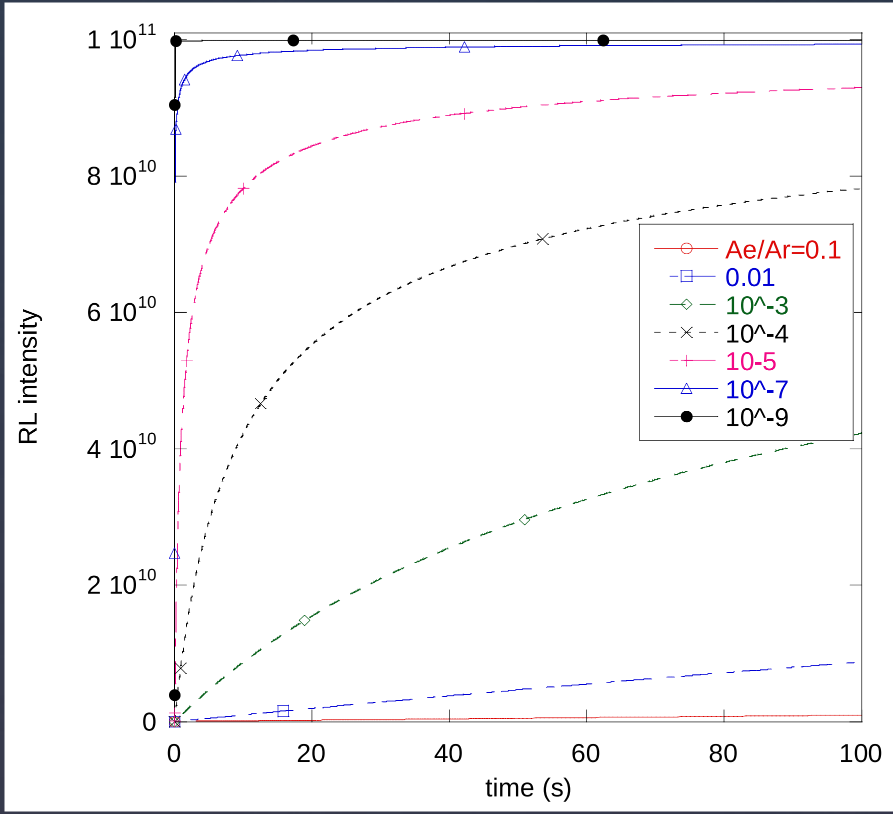


Model results (1)

RL intensity Vs number of electron traps ($M=10^{19}$, T = 10 K)

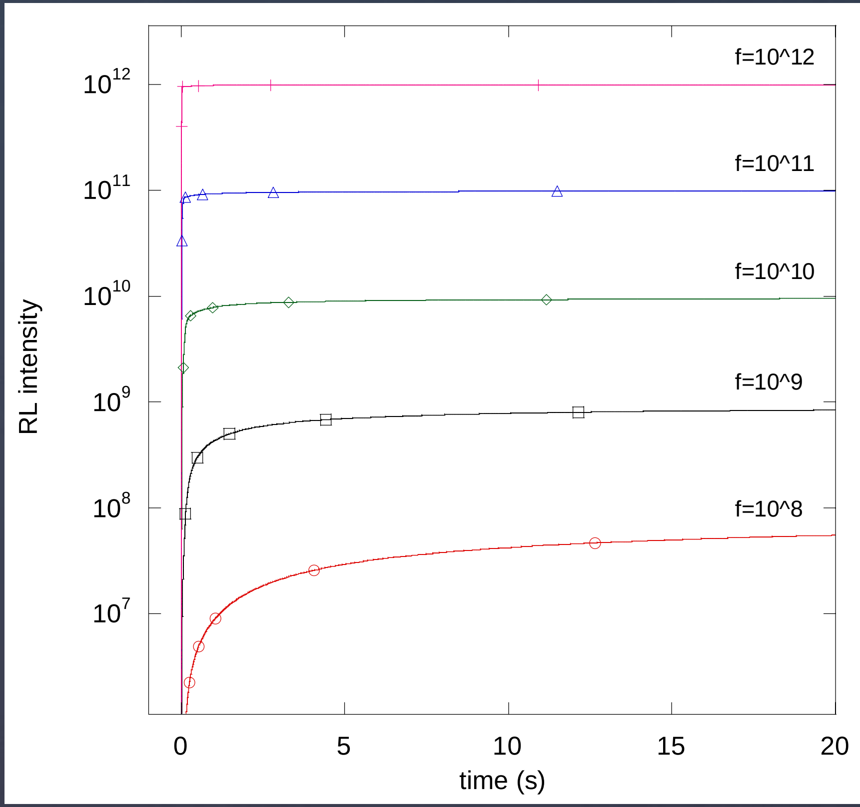


RL intensity Vs trapping probability (A_e) ($A_r=10^{-8}$, T = 10 K)

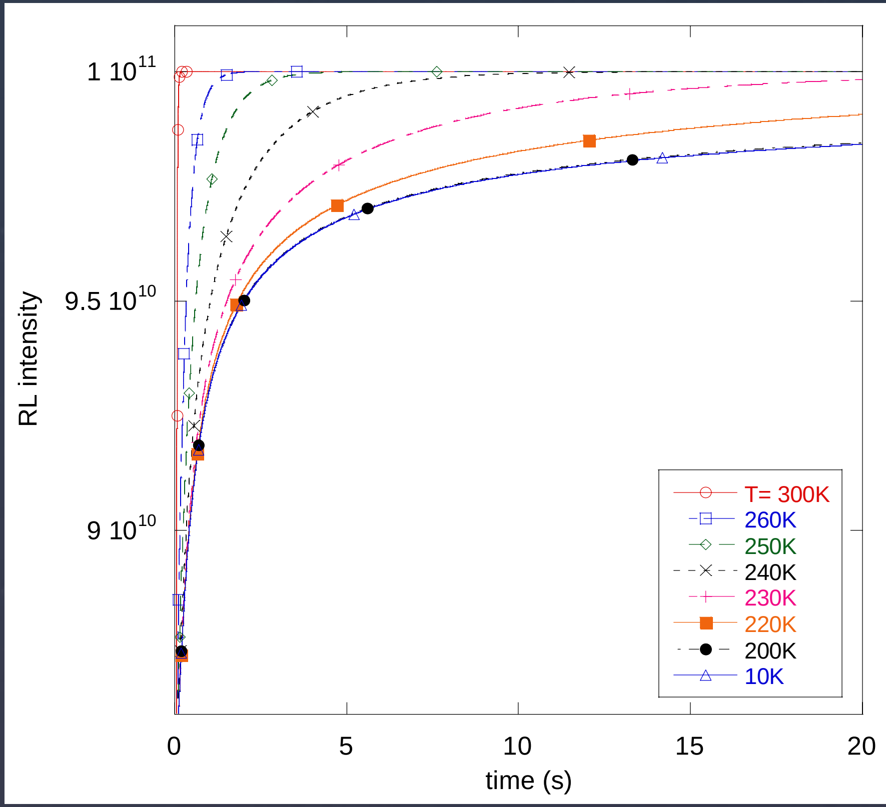


Model results (2)

RL intensity Vs e/h pair creation rate
(T = 10 K)



RL intensity Vs irradiation temperature



The model clearly predict an increase in the RL luminescence. Electron trapping probability and trap concentration seem to play the most relevant role.

The memory effect can be seen also for relatively high temperatures with respect to the trap thermal stability, and it is more evident for low dose rates.

Model testing: looking for the right samples

The ideal sample: 1 trap with known concentration.

Usual scintillators (YAG, LSO, …) have too many traps, and their concentration is essentially unknown.

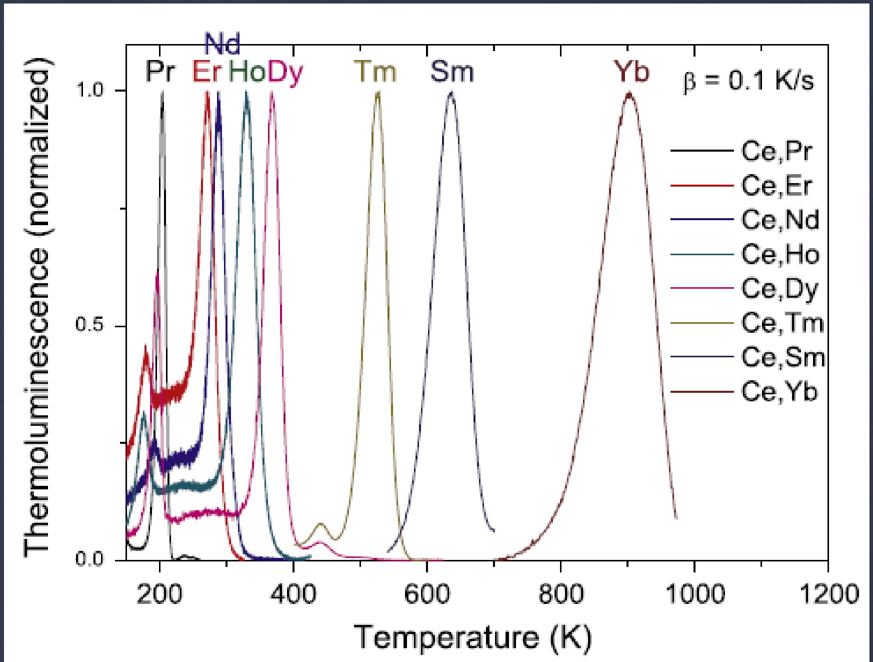
YPO_4:Ce, RE

Ce acts as recombination centre
RE behave as a electron trap

Ce and RE concentration can be chosen during synthesis, and can be checked.

Bos’s results suggest that RE-related glow peak is dominant.

YPO4:Ce, RE glow curves



Bos *et al*; Rad Meas 46(2011)1410

Samples and characterization

Single crystals grown by spontaneous nucleation from a $PbO-P_2O_5$ flux

•YPO_4: Ce 0.1%
•YPO_4: Ce 0.1%, Nd 0.01%, 0.1% and 0.5%

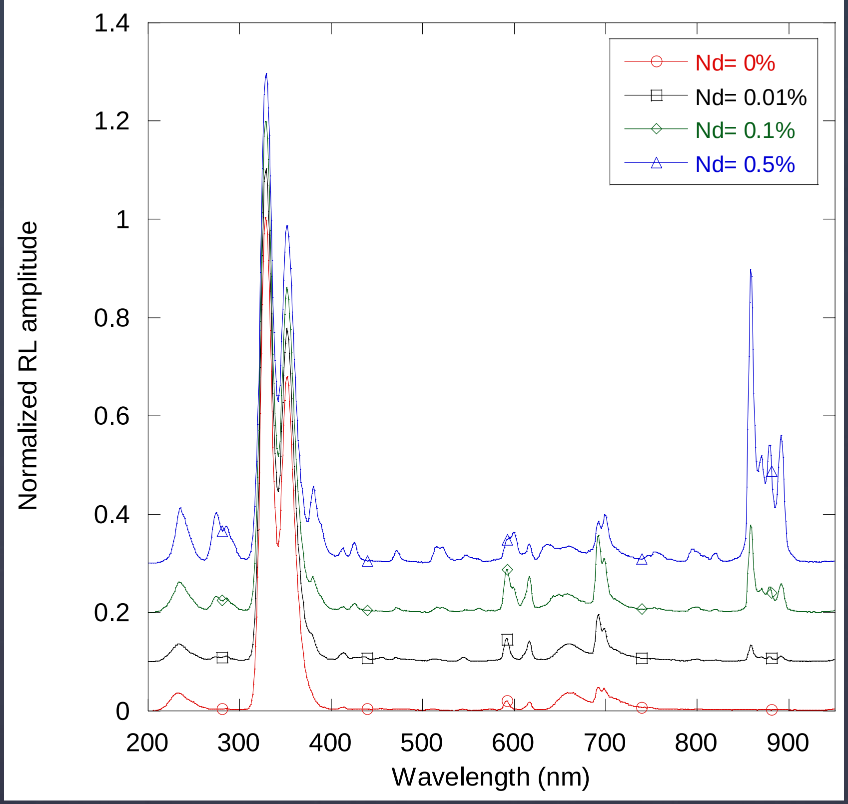
Characterization:

-Radio-luminescence (RT)
-High Temperature (293-473 K) TSL
-Low Temperature (10-320 K) TSL (Ce 0.1%; Ce 0.1%, Nd 0.01%; Ce 0.1%, Nd 0.5%)
-Sensitization (Ce0.1%, Nd 0.5%)

Experimental conditions:
HT-TSL: heating rate 1K/s, irrad. at RT (293K)
LT-TSL: heating rate 0.1K/s irrad. at 10 K or 20 K
Sensitization: irrad 20kV, 5mA (~30mGy/s)

YPO_4:Ce,Nd Radio-luminescence

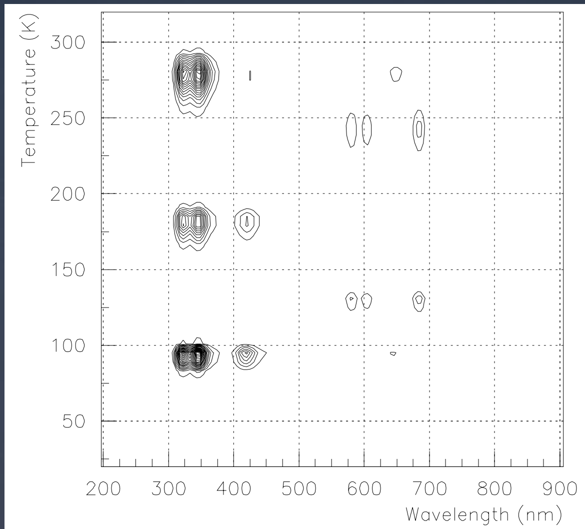
YPO_4:Ce, Nd RL spectra Vs Nd content



- Evident Ce^{3+} 5d-4f radiative transitions (300-400 nm)
- Nd^{3+} 4f-4f (850-920 nm) and 5d-4f (200-300 nm) ones increase with concentration
- Traces of Eu^{3+} (peaks at about 590, 610 and 700 nm)

YPO_4:Ce,Nd Low Temperature TSL

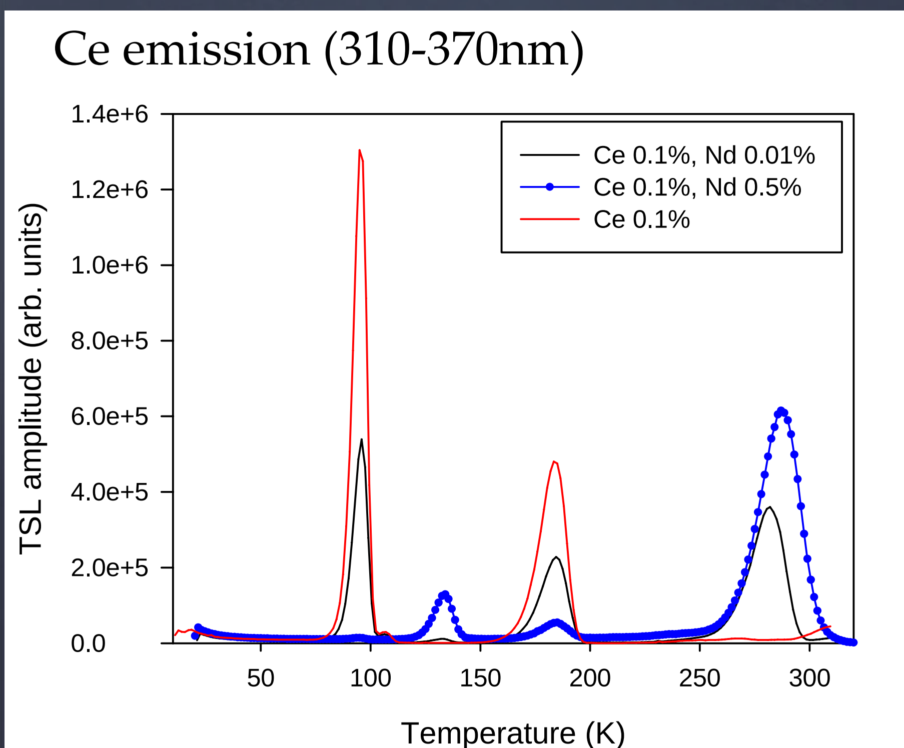
YPO_4:Ce 0.1, Nd 0.01%

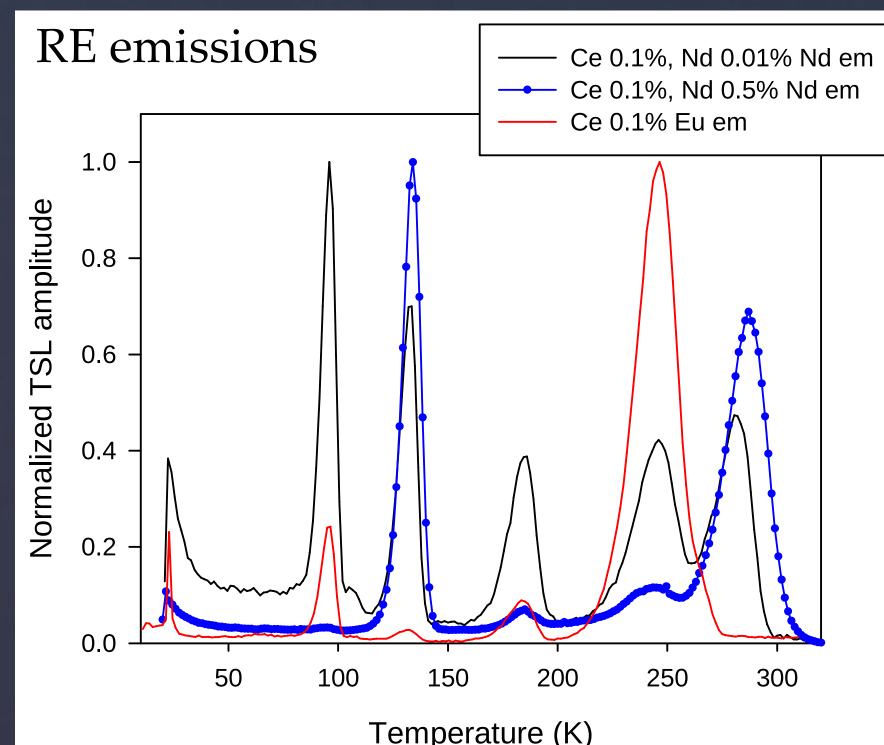


Nd-related glow peaks at 280 K, slightly shifted as the Nd content is increased

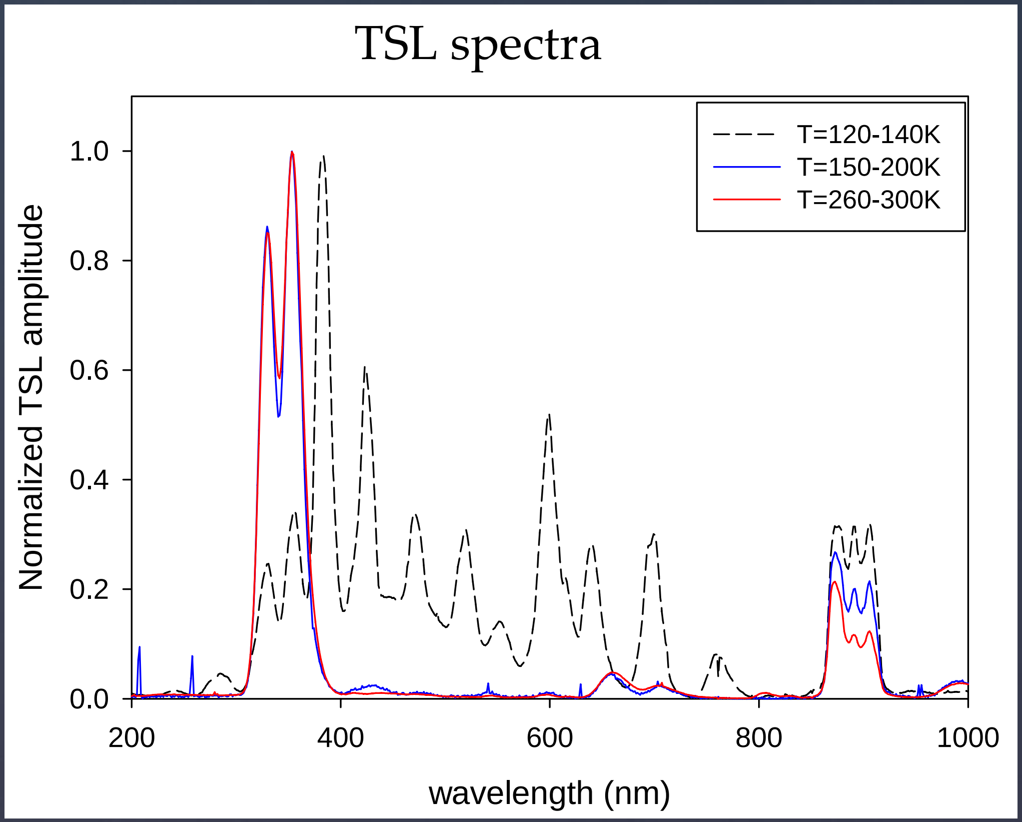
Electron traps at 90 and 183 K

Hole traps at 130 and 250 K

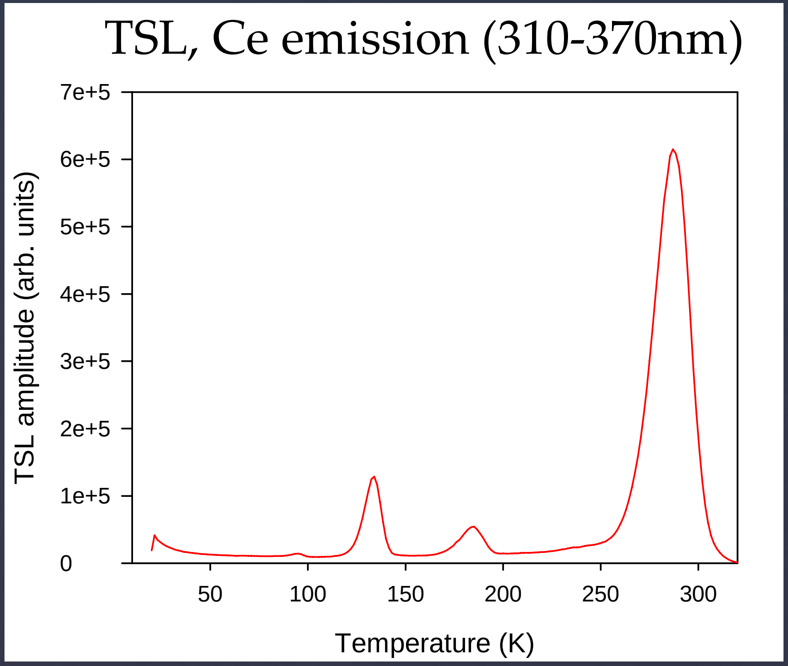




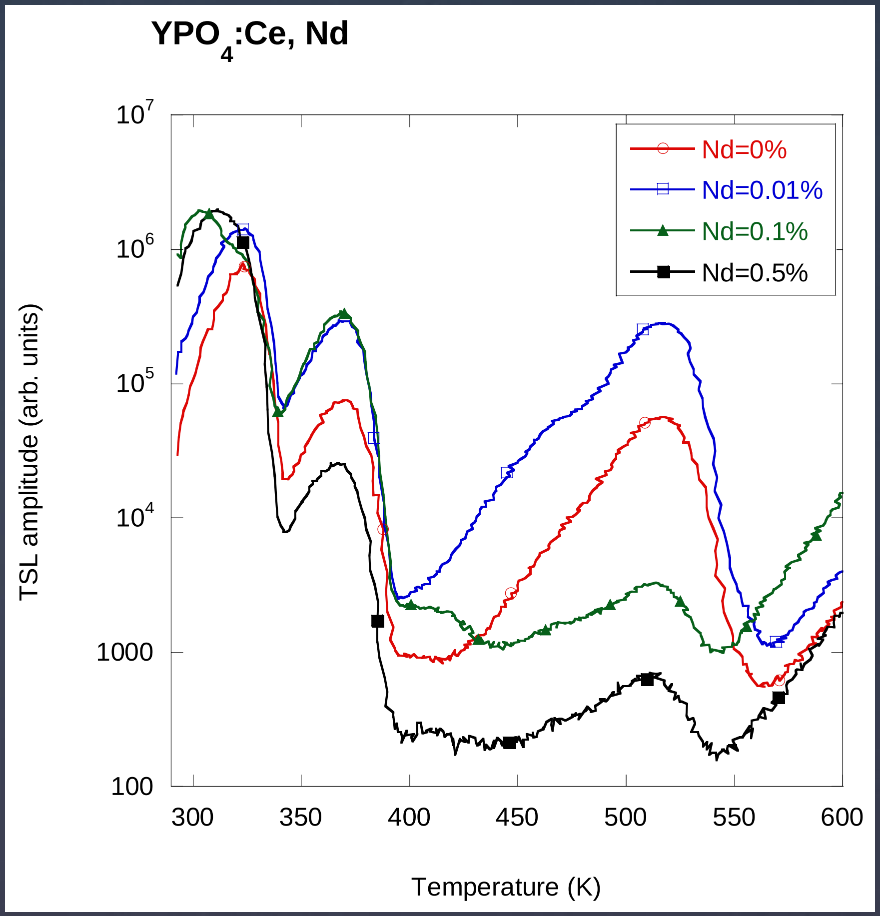
YPO_4:Ce 0.1%, Nd 0.5% Low Temperature TSL spectra



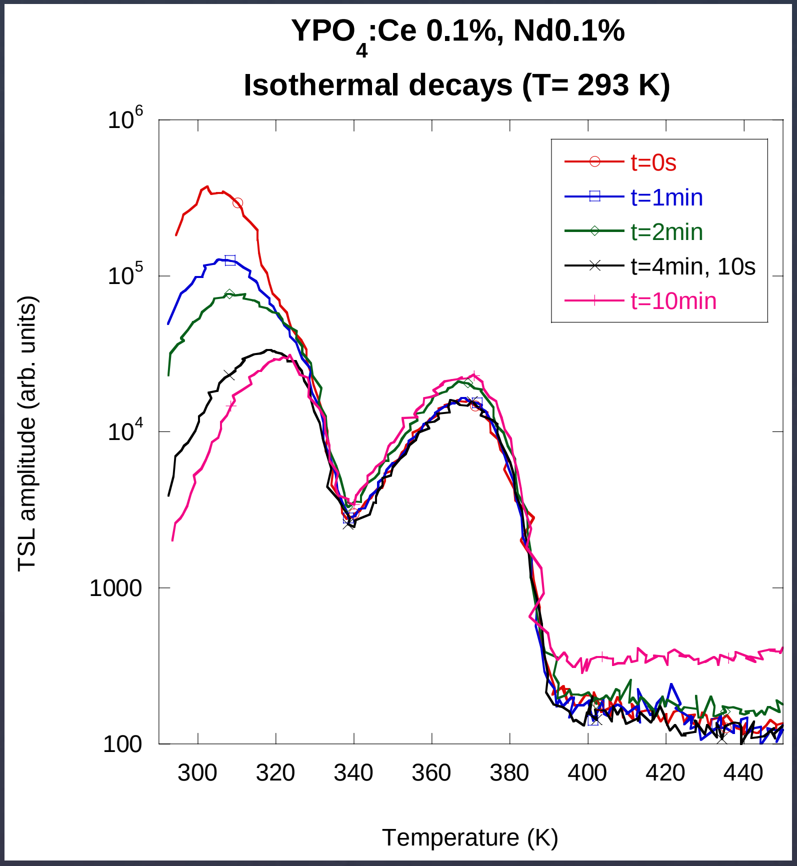
TSL spectra clearly show different emission intensity ratio between Ce^{3+} and Nd^{3+} at the main glow peaks



YPO$_4$:Ce,Nd High Temperature TSL

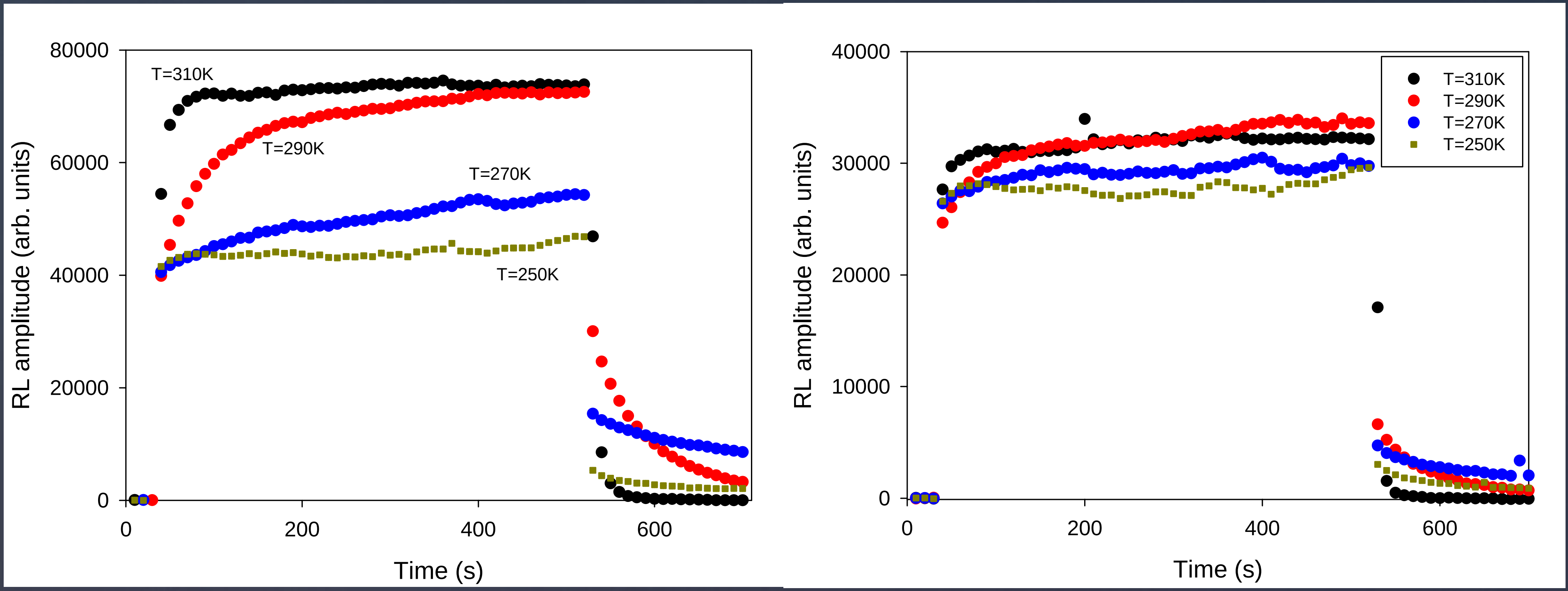


- Nd-related glow peak (at 300K) clearly evident for the two highest contents. Again, shift in its position. It is substantially unstable at room temperature
- Wavelength resolved measurements show only the Ce^{3+} emission
- Other 4/5 glow peaks are clearly visible



RL intensity vs irradiation time

Ce^{3+} emission (300-370nm) | Nd^{3+} emission (861-917nm)



For T=310, 290K: well evident increase at the measurement beginning
For T=270, 250K: lack of initial increase. Possible not complete trap emptying during heating at 300K

Next steps and Conclusions

We plan to:

- Complete the characterization of the TSL traps.
- Obtain new and more reliable RL intensity vs irradiation time growth curves by modifying the irradiation conditions and the measurement temperature, as well as considering also the samples with lower Nd concentration (and other co-dopants).
- Evaluate the goodness of the model on the new experimental data and, in case, improve it in order to make it more realistic.

In conclusion

The first results appear promising (both from the simulation and the experimental point of view): this simple model is able to describe the RL intensity dependence on the accumulated dose, as well as irradiation conditions (temperature, dose rate) and sample related parameters.

YPO_4:Ce, RE seems to be a good test material: the RE-related traps give rise to glow peaks which are, at least for sufficiently high concentrations, much more evident than those related to intrinsic defects. The RL intensity growth is well evident and behaves according to the model.

Thank you for your attention!