

Eu³⁺ or Er³⁺ doped Y₂O₃ thin films grown by ALD with optimized properties for quantum technologies

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Plan

Introduction

I. Towards ALD deposition of ultra-thin films and optical properties

II. Alternative ALD depositions: buffer layer and different substrates

Conclusions and perspectives

Plan

Introduction

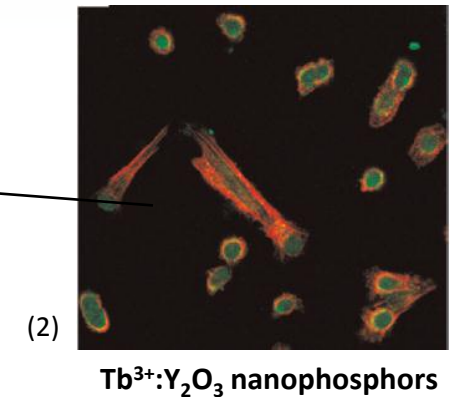
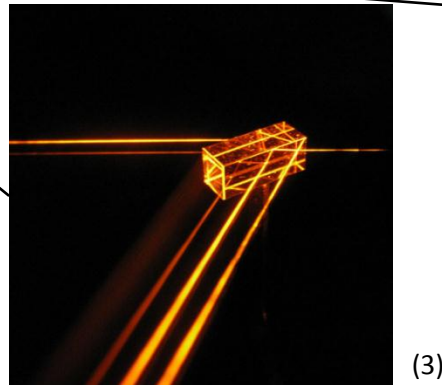
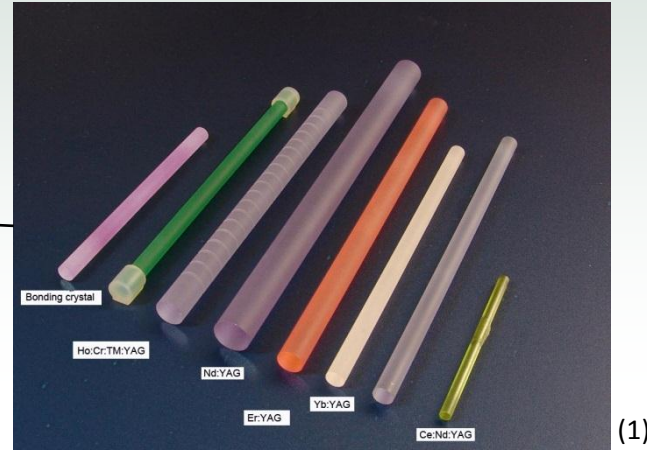
I. Towards ALD deposition of ultra-thin films and optical properties

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Rare-Earth Doped Materials for Photonics

- Luminophores: $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$ (red color)
- Laser: $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Nd}^{3+}$ (YAG)
- Scintillator: $\text{YSO}:\text{Ce}^{3+}$
- Bioimaging (nanoparticles)
- Quantum information

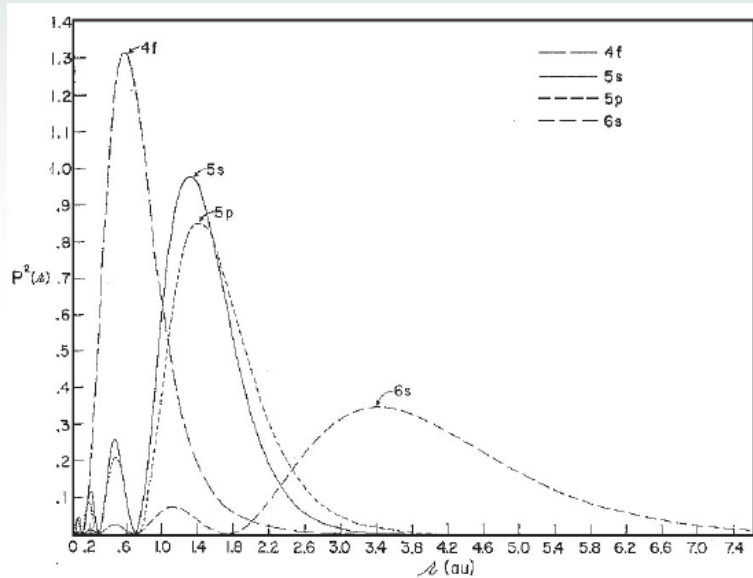


=> Specially designed high-grade materials

(1) <http://keywordhelp.cn.com/vs-yag>
(2) Sotiriou et al, *ACS Nano* 6, 5 (2012).
(3) Courtesy J. Bartolomew

Rare Earth Ions Properties

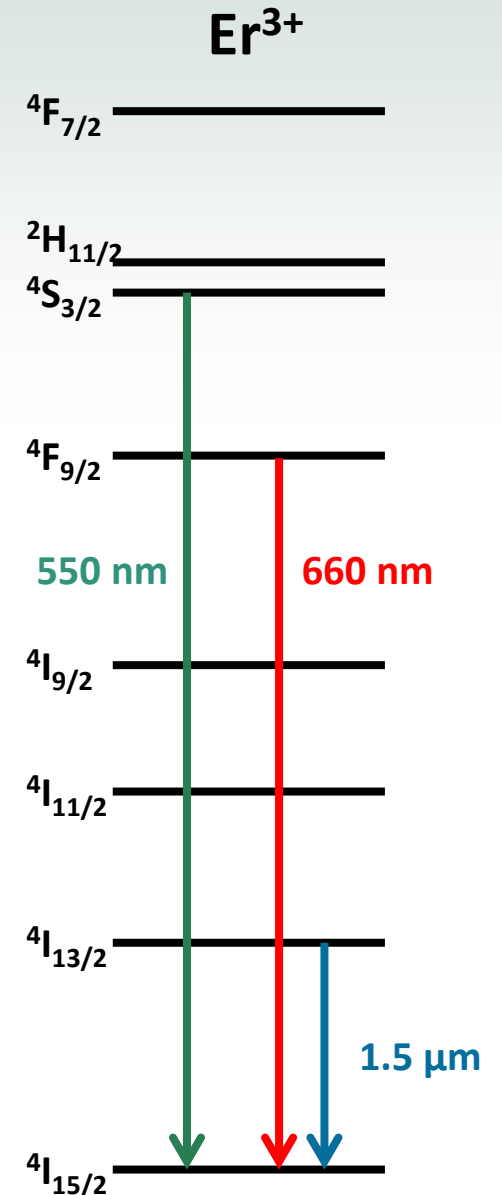
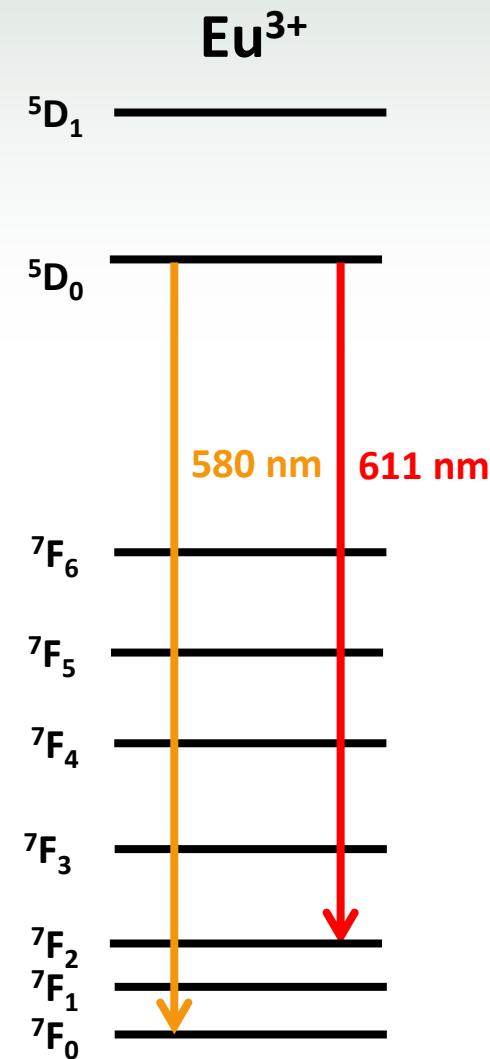
- Electronic configuration Ln^{3+} ion: $[\text{Kr}]4f^n(5s^25p^6)$



(1)

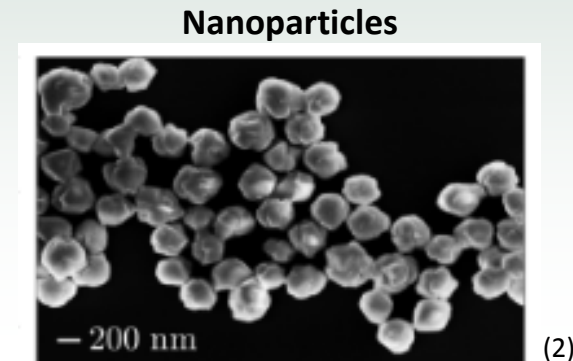
- 4f layer protected from the outside, behave like a gas atom
 → **Long coherence time for optical and spin transitions**
- Wide range of frequencies from MHz to THz

Need for a host material



Objectives

- **Host material:** Y_2O_3 = sesquioxide, cubic phase (ambient T and P)
- $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$ = material with long coherence time:
 - Monocrystal: $T_2 \approx 1 \text{ ms}$
 - Transparent ceramic: $T_2 \approx 100 \mu\text{s}$ (1)
 - Nanoparticles: $T_2 \approx 7 \mu\text{s}$ (2)



- Development of rare earth doped nanostructures for hybrid quantum systems
→ Thin films $\text{Eu}^{3+}:\text{Y}_2\text{O}_3$ or $\text{Er}^{3+}:\text{Y}_2\text{O}_3$ on Si(100) substrates
- **Final goal:** obtain a high quality **ultrathin film** with properties as close as possible to the single-crystal
→ Different couplings possible (photonic crystals, cavity, mechanical resonator...)

Ultra-thin films with optimized optical properties => Atomic Layer Deposition (ALD)

(1) N. Kunkel et al, *APL Materials*, 3 (2015).

(2) J. G. Bartholomew et al, *Nano. Lett.*, 17 (2017).

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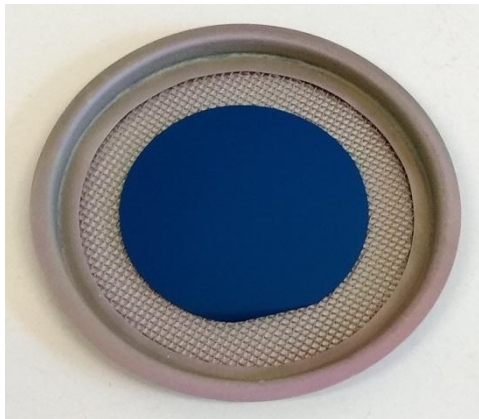
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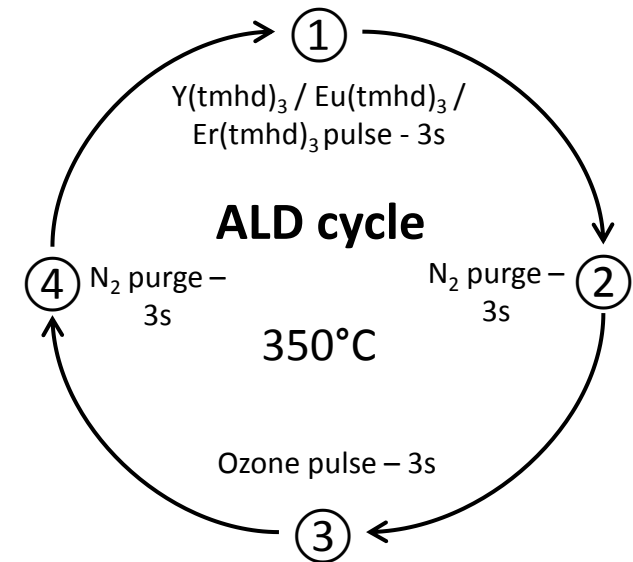
Conclusions and perspectives

I. Towards ultra-thin films – ALD deposition

- Picosun Sunale R200 vertical flow type reactor
- $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$ and $\text{Y}_2\text{O}_3:\text{Er}^{3+}$ thin films (5 nm – 400 nm)
- $\text{Y}(\text{tmhd})_3$, $\text{Eu}(\text{tmhd})_3$ and $\text{Er}(\text{tmhd})_3$ precursors and O_3
- Optimization of ALD parameters (deposition and precursors temperatures, ozone and purge pulse time,...)

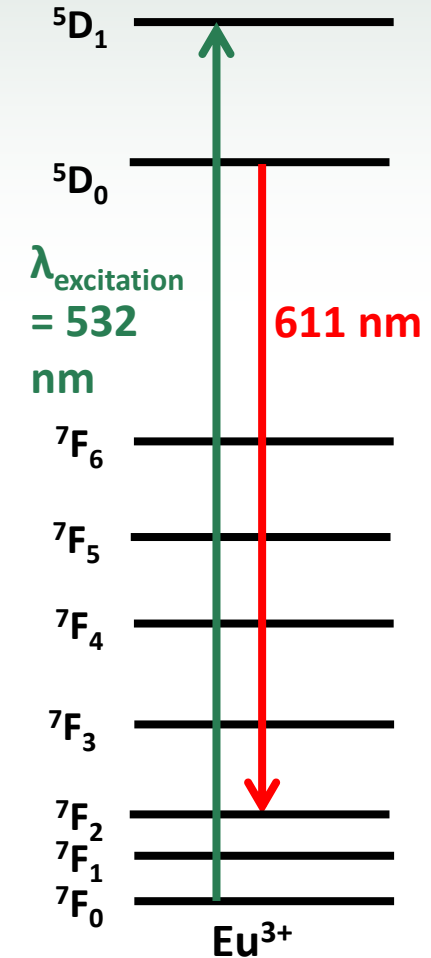
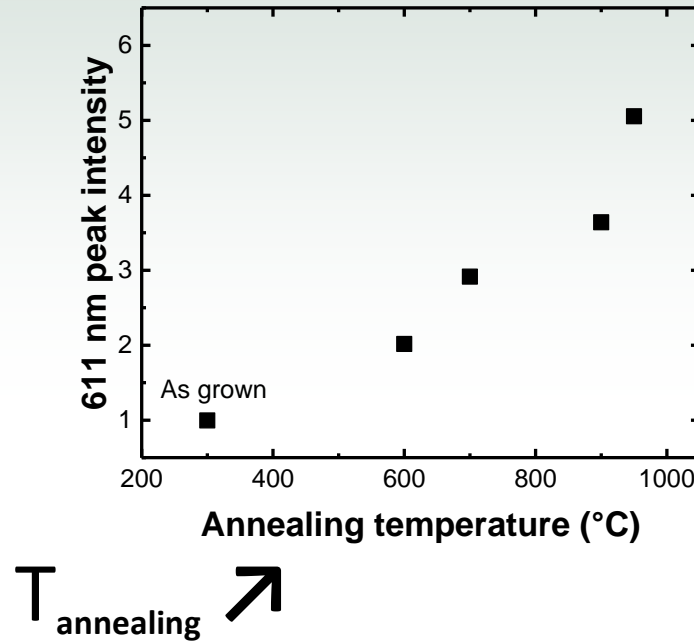
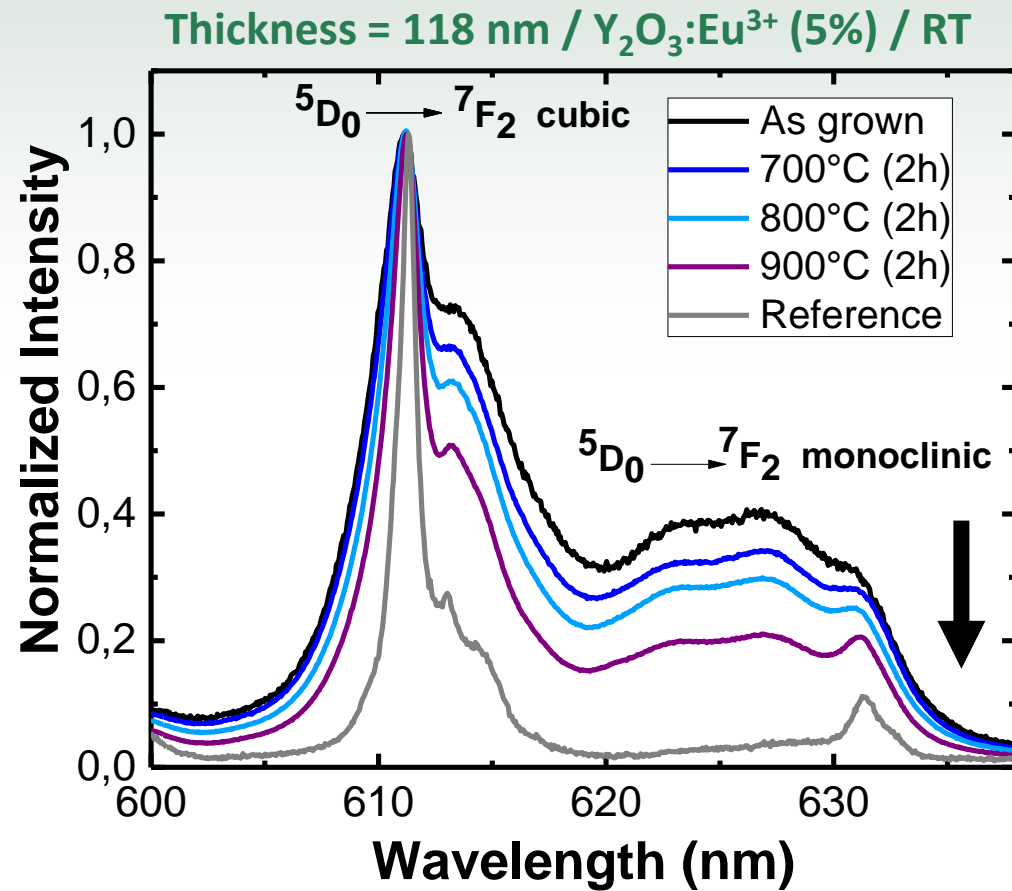


Yttrium oxide thin film doped with Eu^{3+} (thickness of 100 nm) on Si(100) elaborated by ALD



Scarafagio et al, *J. Phys. Chem. C*, 123 (2019).

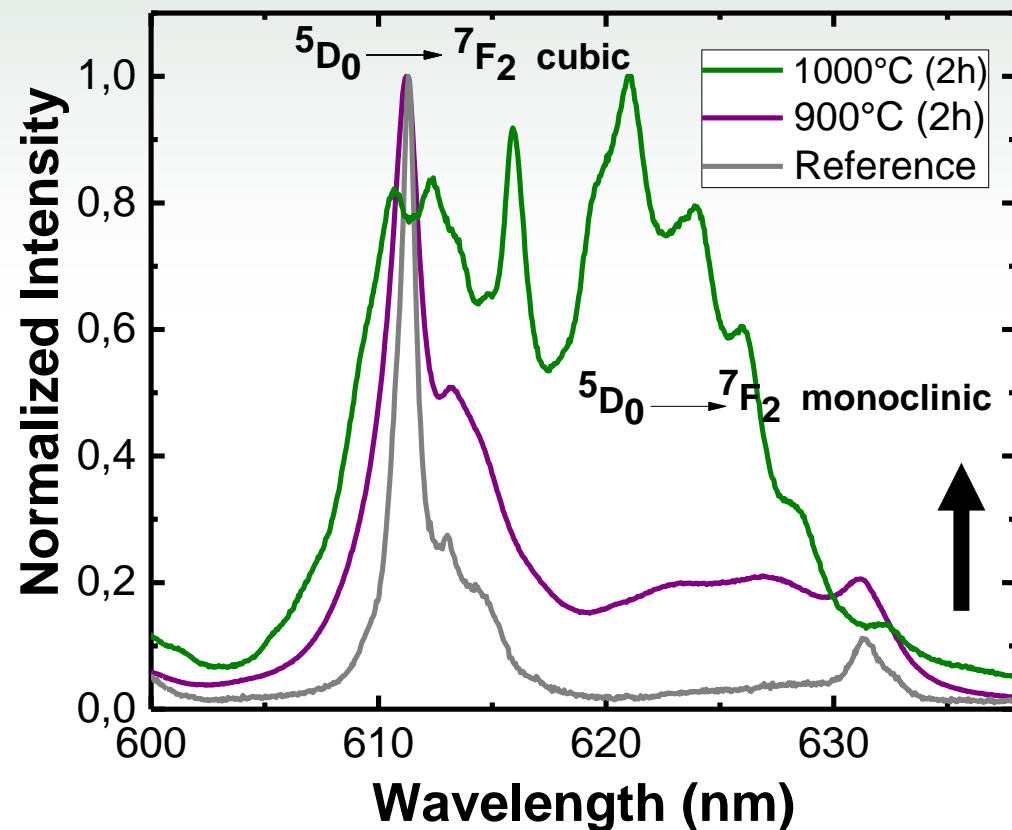
I. Room Temperature Photoluminescence



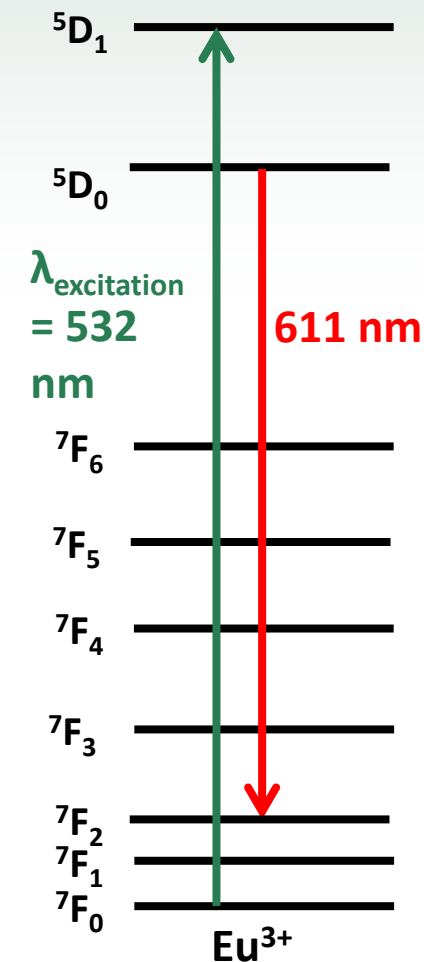
Scarafagio et al, *J. Phys. Chem. C*, 123 (2019).

I. Room Temperature Photoluminescence

Thickness = 118 nm / $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$ (5%) / RT

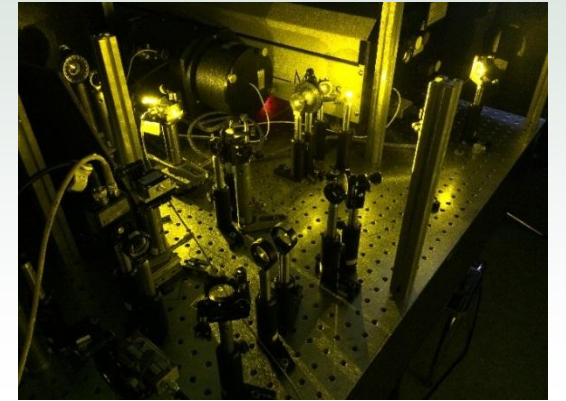
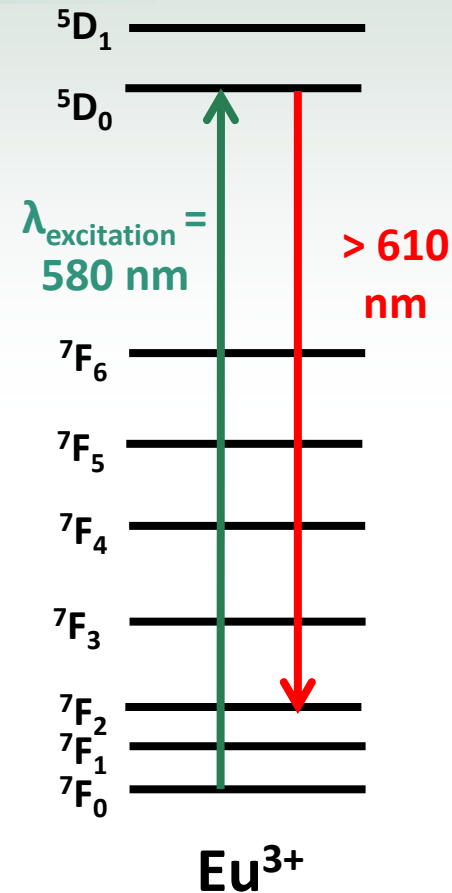
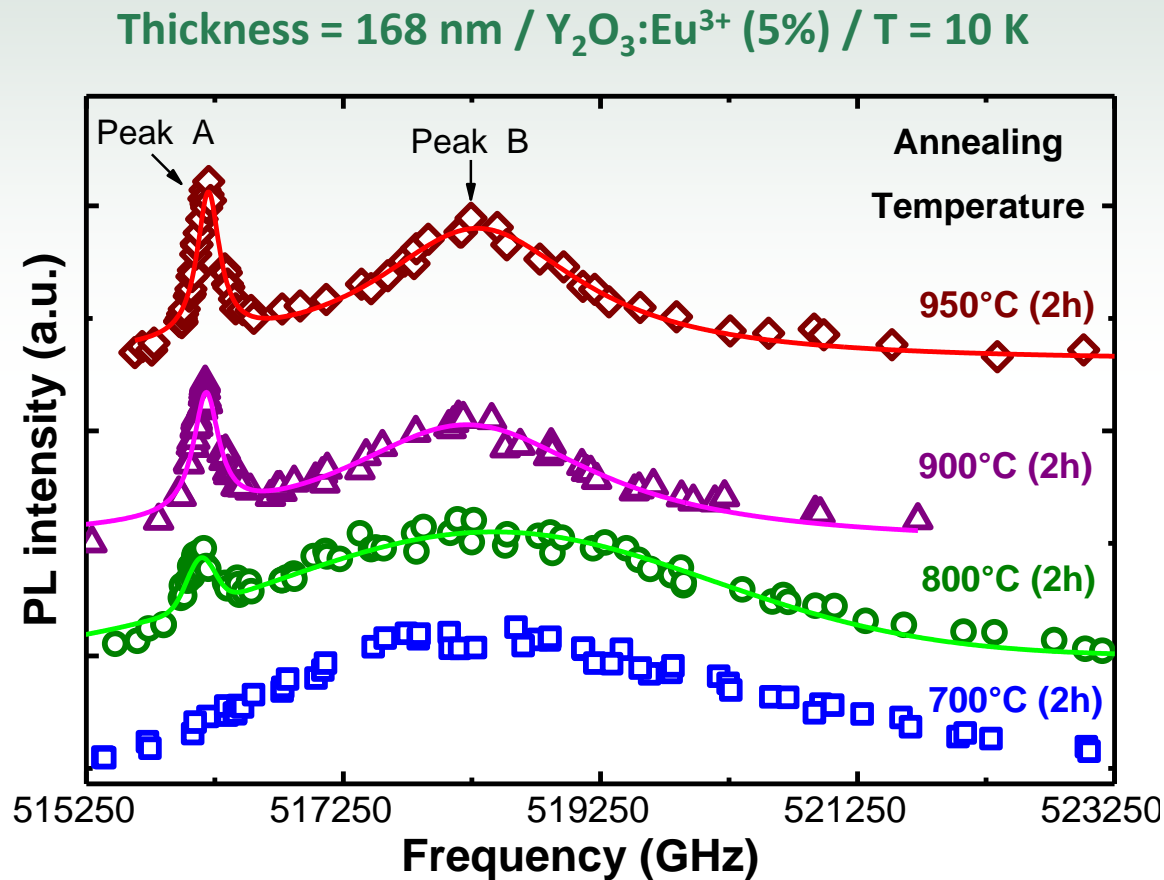


But above 1000°C new phases are formed (interfacial silicate)



Scarafagio et al, *J. Phys. Chem. C*, 123 (2019).

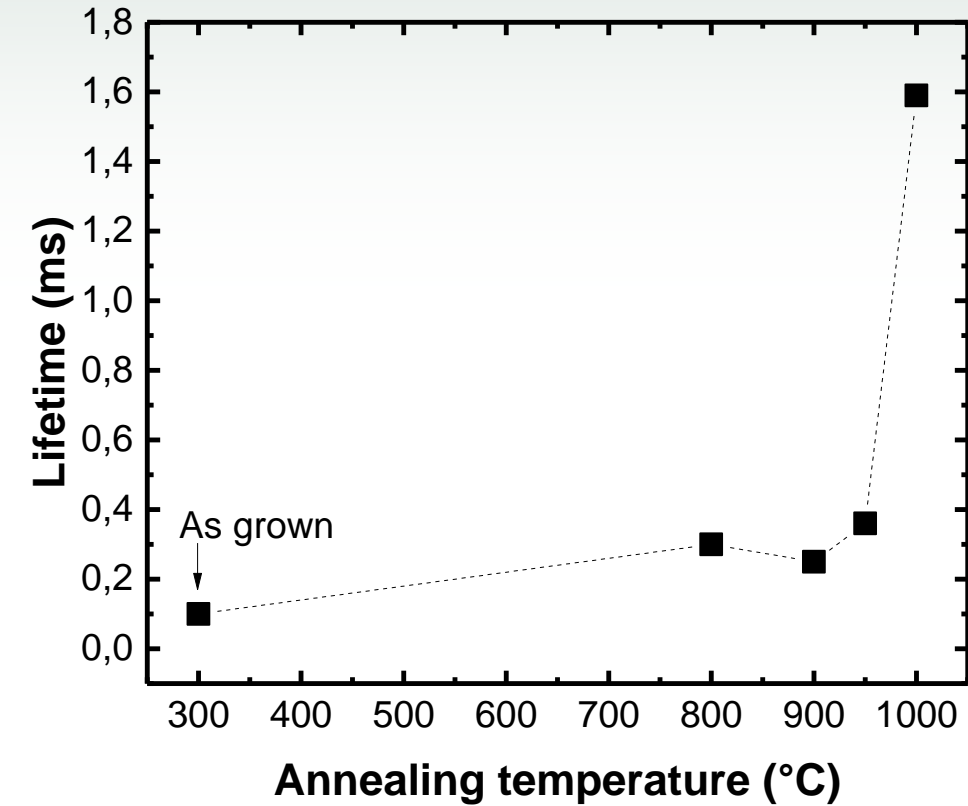
I. Inhomogeneous linewidth (PLE)



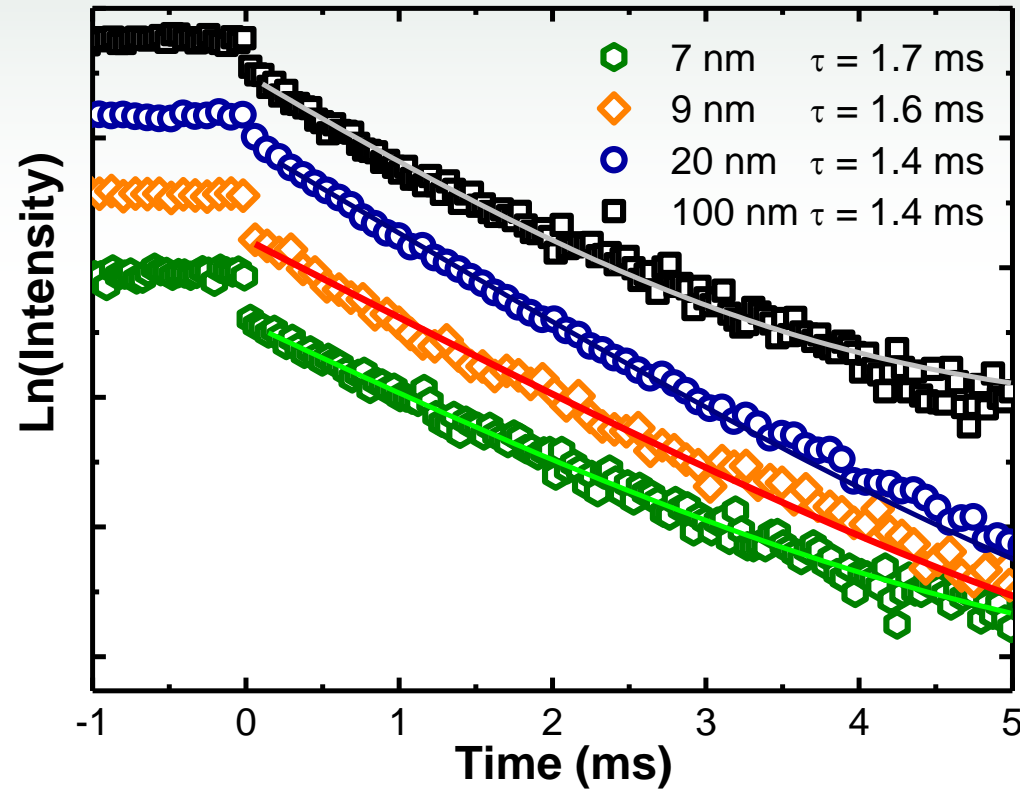
- **Peak A** \rightarrow $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$ cubic phase; **Peak B** \rightarrow disorder + monoclinic phase
- Films: $\Gamma_{\text{inh}} \approx 200 \text{ GHz}$ and Ceramic (reference): $\Gamma_{\text{inh}} \approx 100 \text{ GHz} \rightarrow$ Impurities, defects and stress

I. Ultra-Thin Films: 1.5 μm Er^{3+} Lifetime Measurements

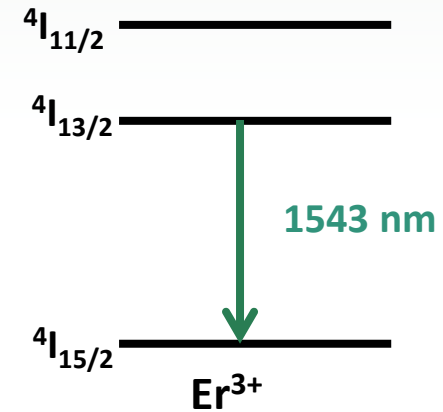
Thickness = 14 nm / $\text{Y}_2\text{O}_3:\text{Er}^{3+}$ (2%) / RT



$\text{Y}_2\text{O}_3:\text{Er}^{3+}$ (2%) / RT / Annealing = 950°C (2h)



ICFO^R
The Institute of Photonic
Sciences



- Improvement with annealing step
- Lifetime **still long** (> 1 ms) when the thickness decreases
- **Reference:** 76 μs for $\text{Y}_2\text{O}_3:\text{Er}^{3+}$ (1 mol %) nanoparticles with a diameter of 100-150 nm (1)

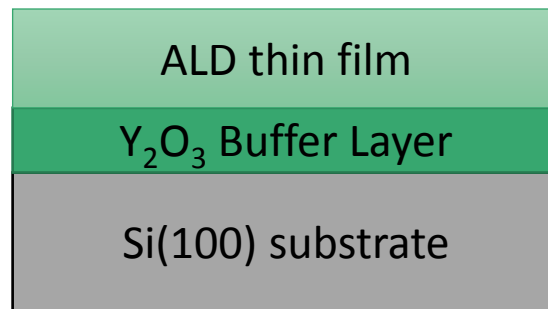
I. Towards Ultra-Thin films – Summary

- Promising results but **limitations** with Si(100) substrates:
 - SiO₂ growth after annealing step at 900°C and above
 - New phases formation at 1000°C and above

Two alternative strategies

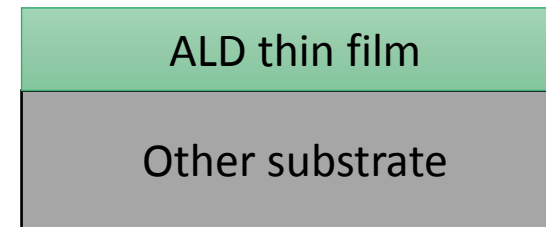
Buffer Layer

- Increase the **distance** between the **emitting ions** and the **substrate**
- **Ex:** Y₂O₃ grown by ALD, Y₂O₃ grown by CVD...



Other substrates

- **Refractory** substrates
- **Ex:** sapphire, transparent ceramic Y₂O₃



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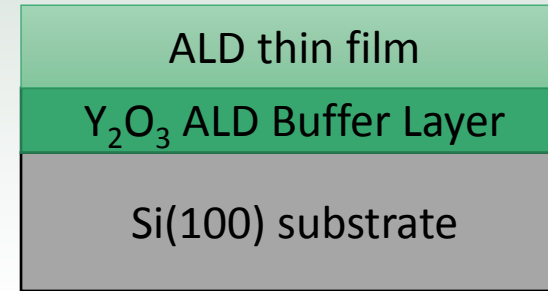
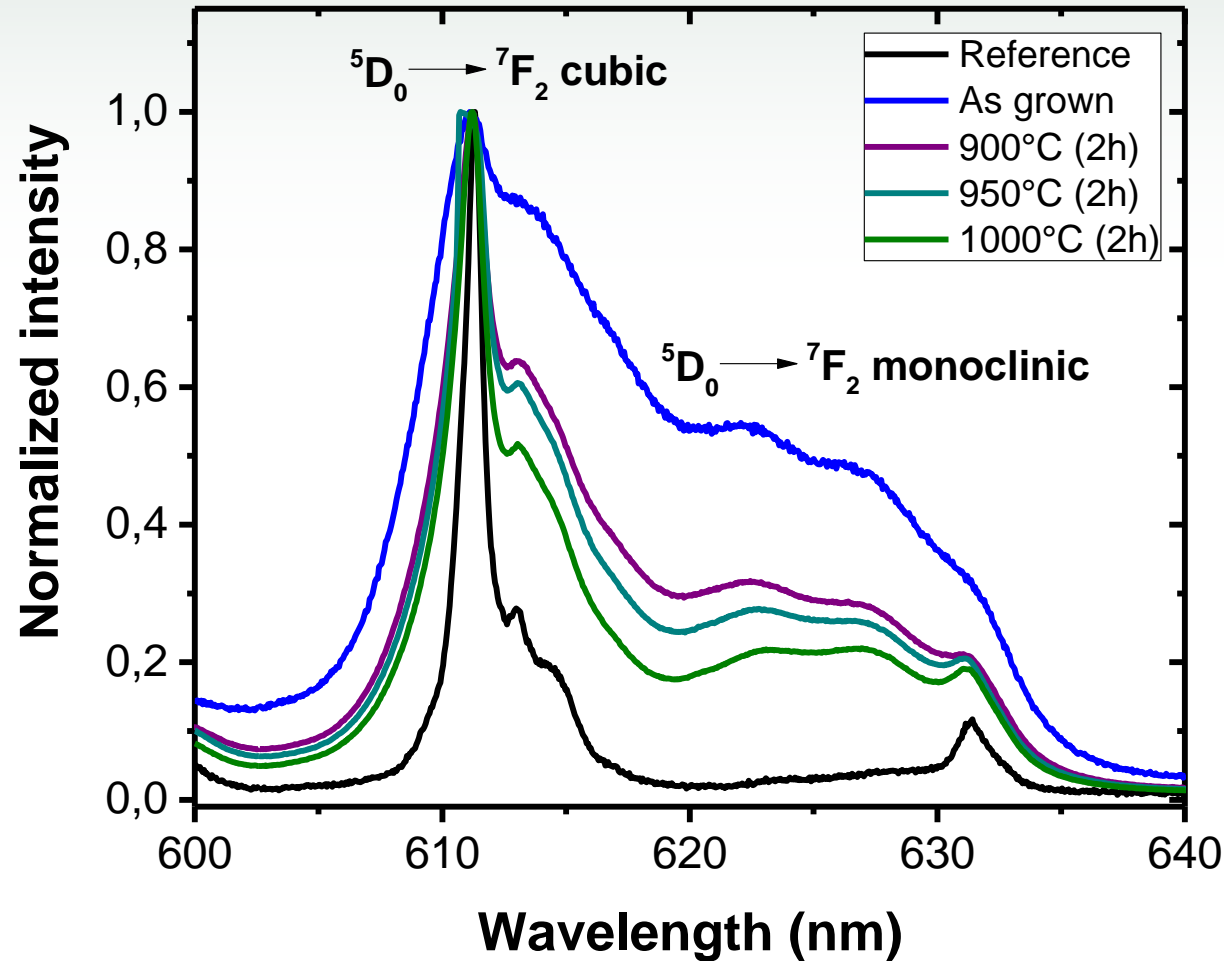
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II. Alternative Depositions – ALD Buffer Layer

- Buffer layer = Y_2O_3 grown by ALD



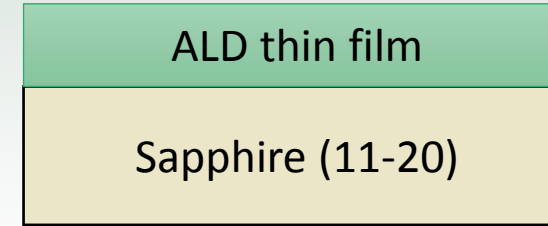
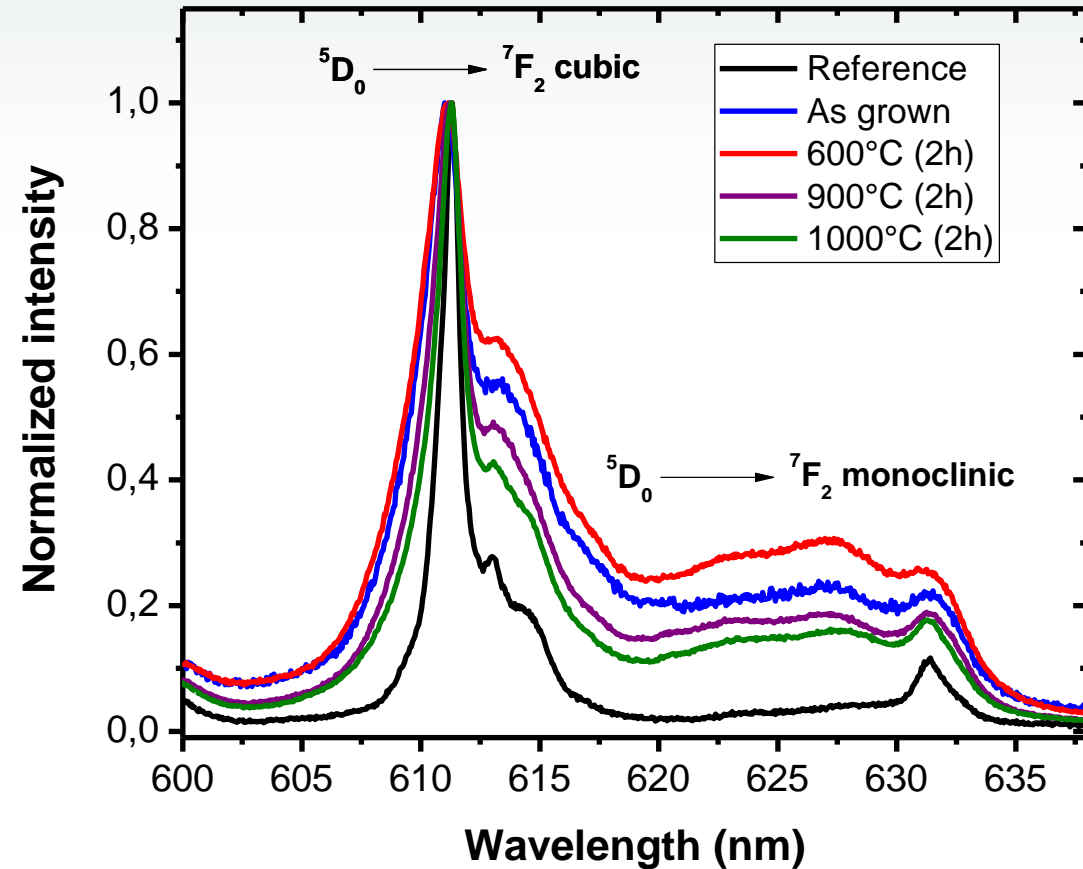
Thickness Film = 358 nm
 $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$ (5%)
+ buffer Y_2O_3

- Improvement of PL spectra when the annealing temperature increases
- No parasitic phases at 1000°C**

The ALD buffer layer opens the way to higher annealing steps

II. Alternative depositions – Other substrates

- Substrate = **Sapphire (11-20)** (a-plane = blue plane)

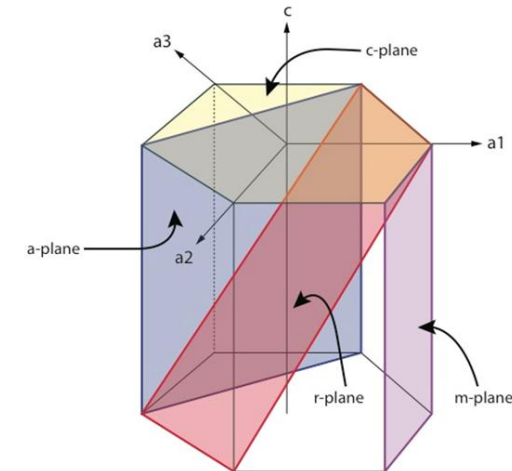


Thickness = 83 nm
 $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$ (5%)

- Improvement** of PL spectrum when the temperature increases
- No additionnal phases** at 1000°C

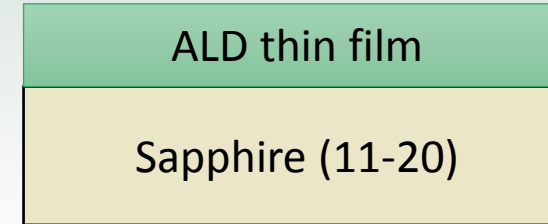
Promising PL properties at
1000°C => Higher temperature ?

Hexagonal cell of sapphire

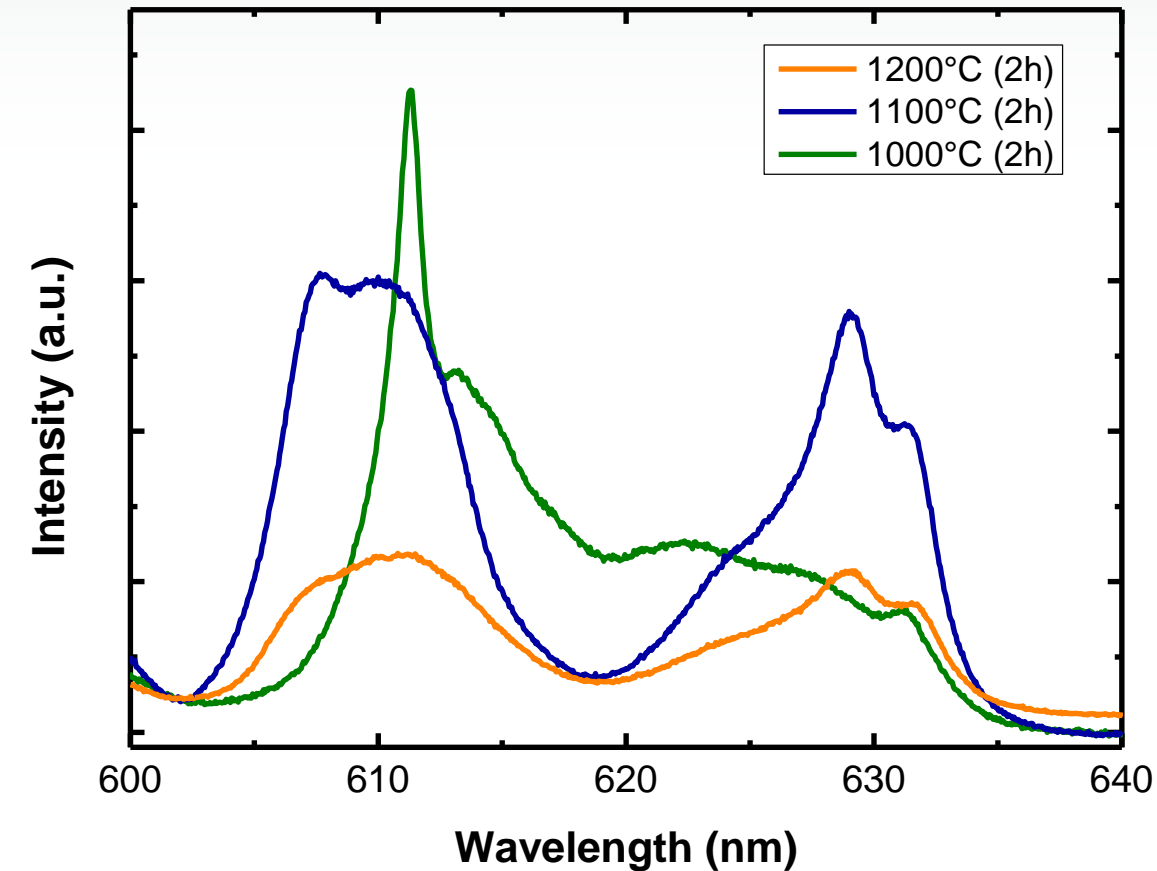


II. Alternative depositions – Other substrates

- ALD thin films on sapphire substrates annealed at 1100°C and 1200°C for 2 hours



Thickness = 83 nm
 $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$ (5%)



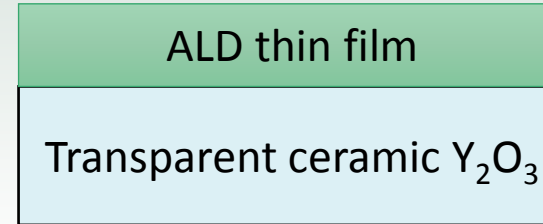
- Deformation of PL spectrum at **1100°C** and **1200°C**
- Formation of **new phases** at the interface

Maximum temperature for annealing step:
1050°C

II. Alternative depositions – Other substrates

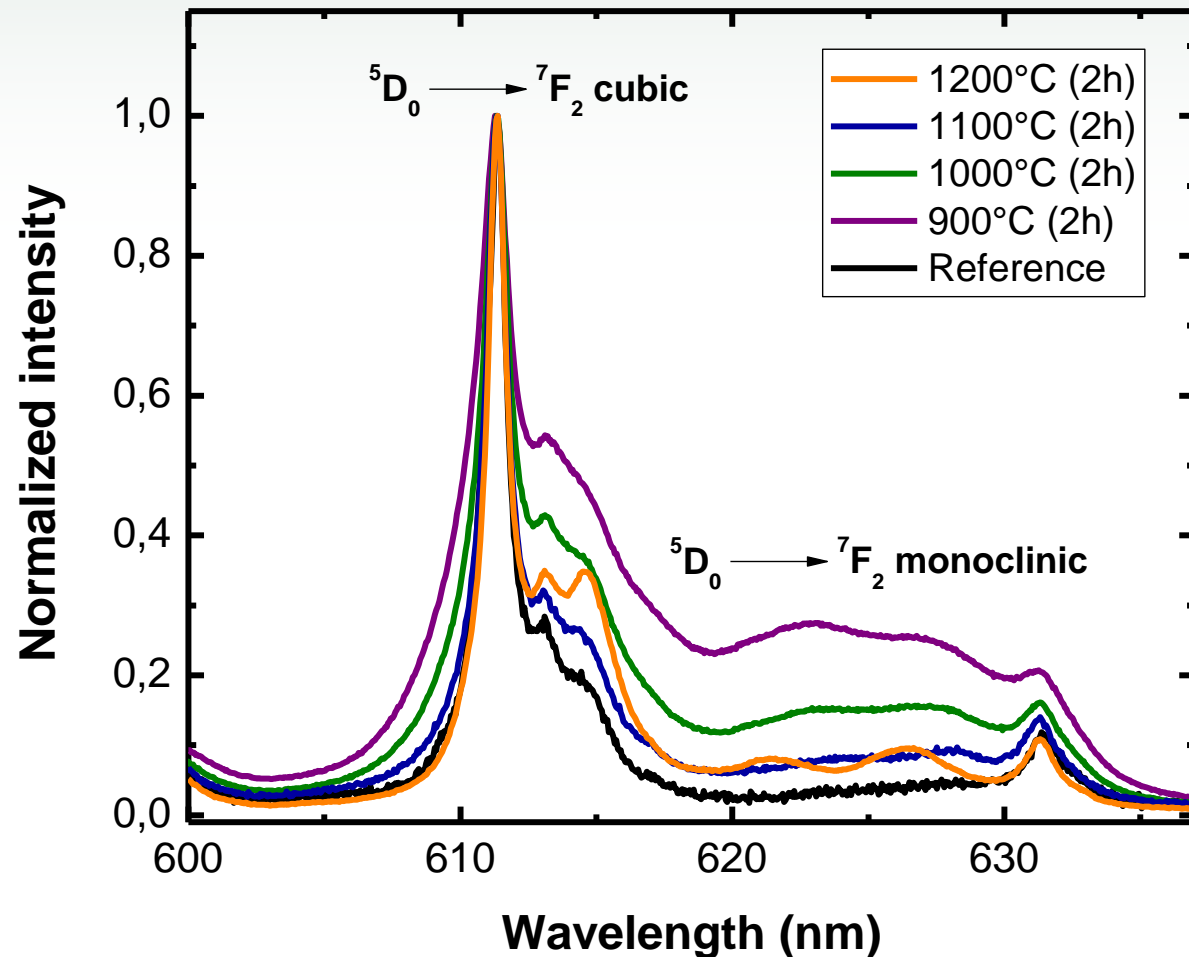
- Substrate = transparent ceramic Y_2O_3

Thickness = 83 nm
 $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$ (5%)



- Improvement of PL spectrum
- For $T_{\text{annealing}} = 1100^\circ\text{C}$ and 1200°C , PL spectrum is **very close** to the reference
- No formation of parasitic phases

Promising PL properties with transparent ceramic Y_2O_3 substrates



Conclusions and perspectives

- Annealing step after the deposition → improved optical spectroscopic properties
- Limitation to 950°C on Si substrates → parasitic phases
- First measurement of narrow inhomogeneous linewidth on films ($\text{Eu}^{3+} \text{ } ^5\text{D}_0 \rightarrow ^7\text{F}_0$ 200 GHz)
- Other substrates → higher annealing temperatures up to 1200°C
- Sapphire or transparent ceramic Y_2O_3 → promising substrates

ON PROGRESS:

- **Lifetime measurements:**
 - On other substrates
 - Improvement of lifetime up to 8 ms for 11 nm thick film (ICFO)
- Study of the emitting ions Eu^{3+} localisation on the surface
- Coherent experiment currently on thin films: **measurement of T_2**

Thank you for your attention !



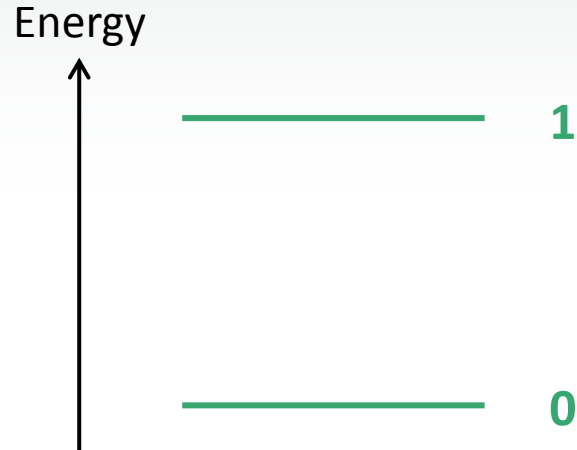
<http://www.nanoqtech.eu/>



This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 712721.

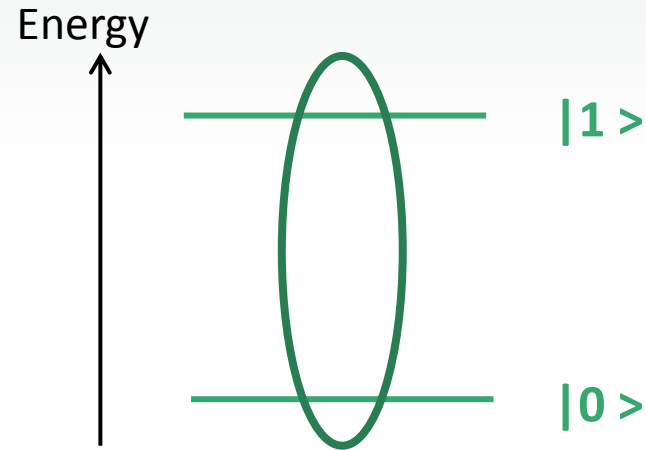
Introduction – Quantum Properties

Classical system (bit):



=> States: 0 and 1

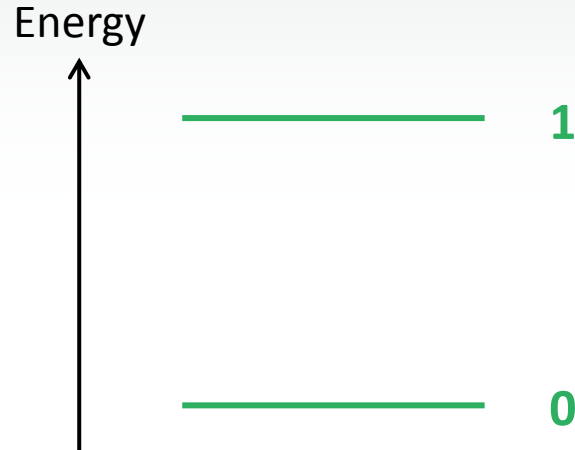
Two-level quantum system (= Qubit):



=> Superposition states: $\Psi = a|0\rangle + b|1\rangle$

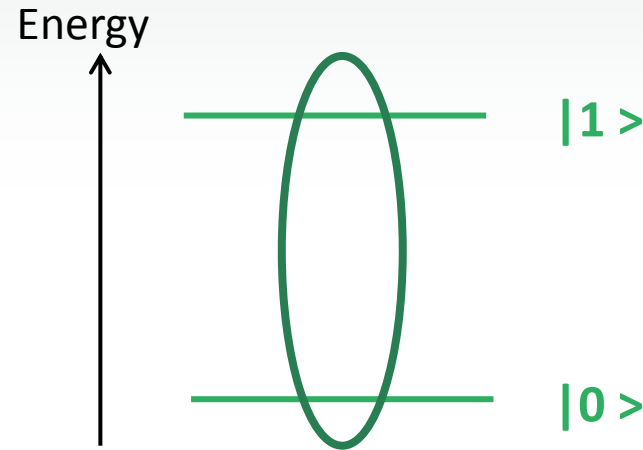
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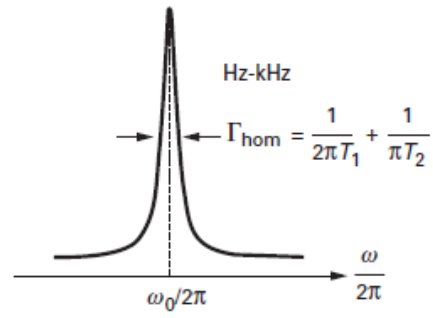


=> Superposition states: $\Psi = a|0\rangle + b|1\rangle$

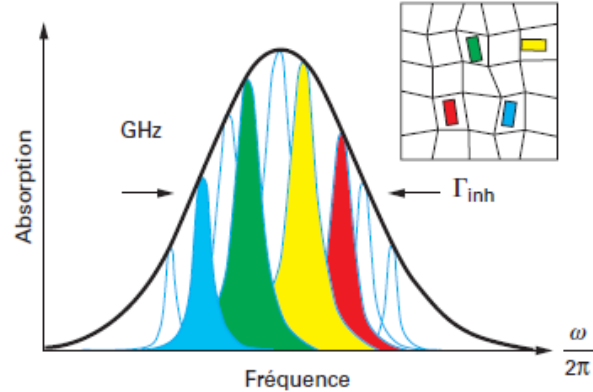
- Quantum information: **coherent superposition** of two quantum states $|0\rangle$ et $|1\rangle$
- **Problem** = interactions with the environment => **decoherence** (loss of quantum information)
- **Goal** = systems with coherence as long as possible (high T_2)

=> Use of rare earth ions

Inhomogeneous Linewidth



- (a) élargissement homogène provoqué par l'interaction dynamique entre l'ion TR^{3+} optiquement actif et les différentes sources de décohérence



- (b) élargissement inhomogène induit par une variation d'environnement local autour de l'ion TR^{3+} . L'insert est une représentation schématique de l'environnement cristallin autour de quatre dopants différents

Figure 11 – Illustration des deux effets d'élargissement des raies optiques

(1) T. Chanelière, A. Louchet-Chauvet, A. Ferrier and P. Goldner, *Cristaux et dispositifs optiques pour le traitement de l'information quantique*, Techniques de l'ingénieur (2014).

Alternative depositions – ALD Buffer Layer

- Cathodoluminescence spectra:

Thickness Film = 11 nm
 $\text{Y}_2\text{O}_3:\text{Er}^{3+}$ (2%) + Buffer Y_2O_3 (50 nm)
Annealed at 950°C (2h)

