

# Rare-earth doped oxide thin films deposited by DLICVD for quantum technologies applications



Nao Harada, Alexandre Tallaire, Alban Ferrier, Diana Serrano, Philippe Goldner

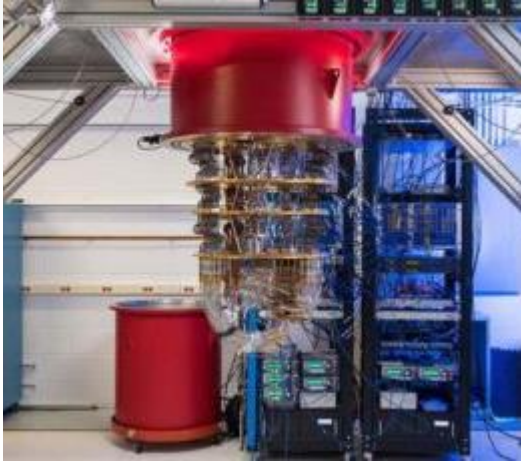
Institut de Recherche de Chimie Paris, Chimie ParisTech, CNRS, Université PSL, Paris, France

*GDR Epidox November 2021*

# 1. Introduction on quantum technologies

# Quantum technologies

## Quantum computing



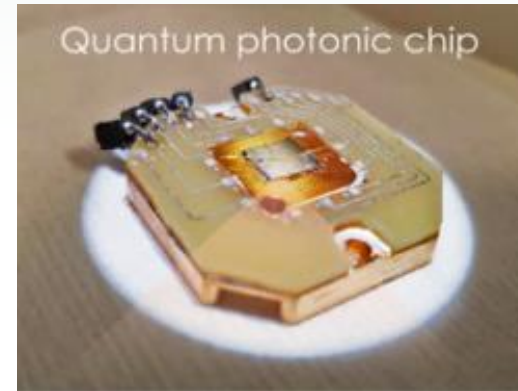
Google

## Quantum cryptography



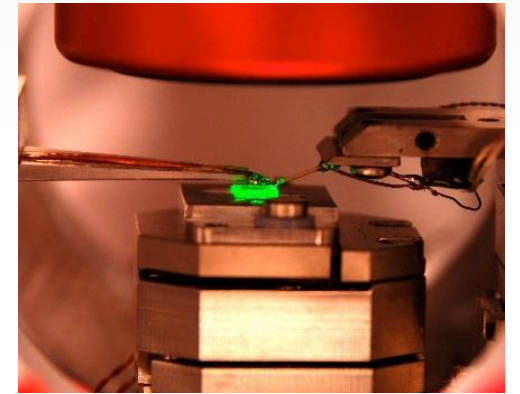
IDquantique

## Quantum communications



Quandela

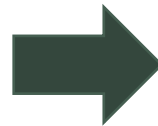
## Quantum sensing and metrology



QNami



QUANTUM  
FLAGSHIP



ASTERIQS

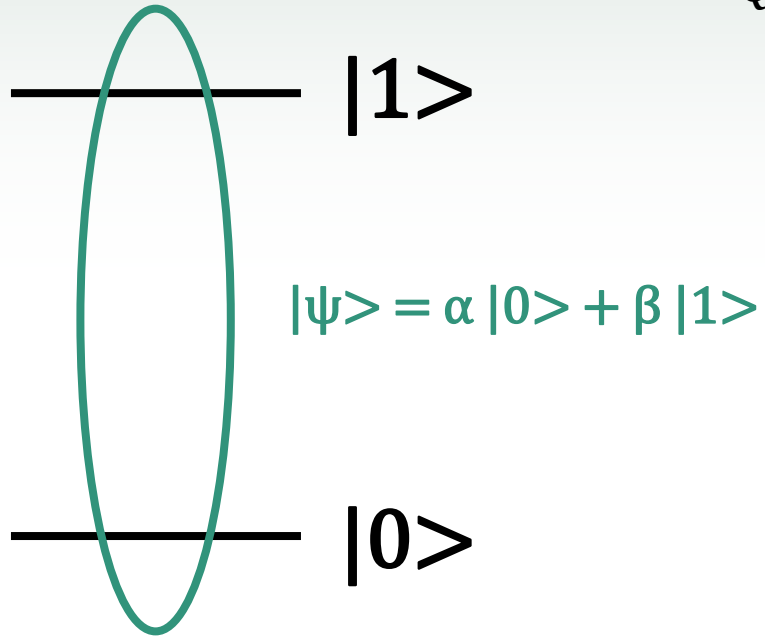
<https://www.asteriqs.eu/>

SQUARE

<http://square.phy.kit.edu/>

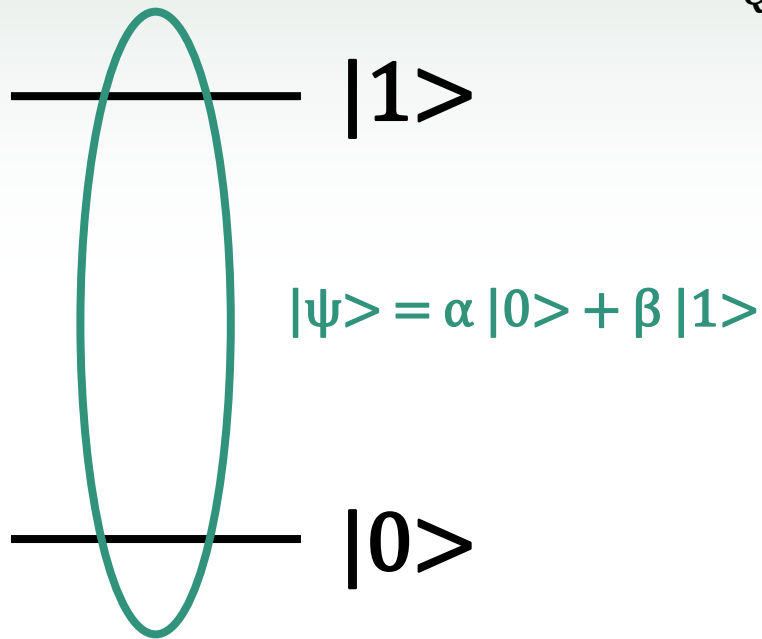
# Need for stable qubits

*Qubit*

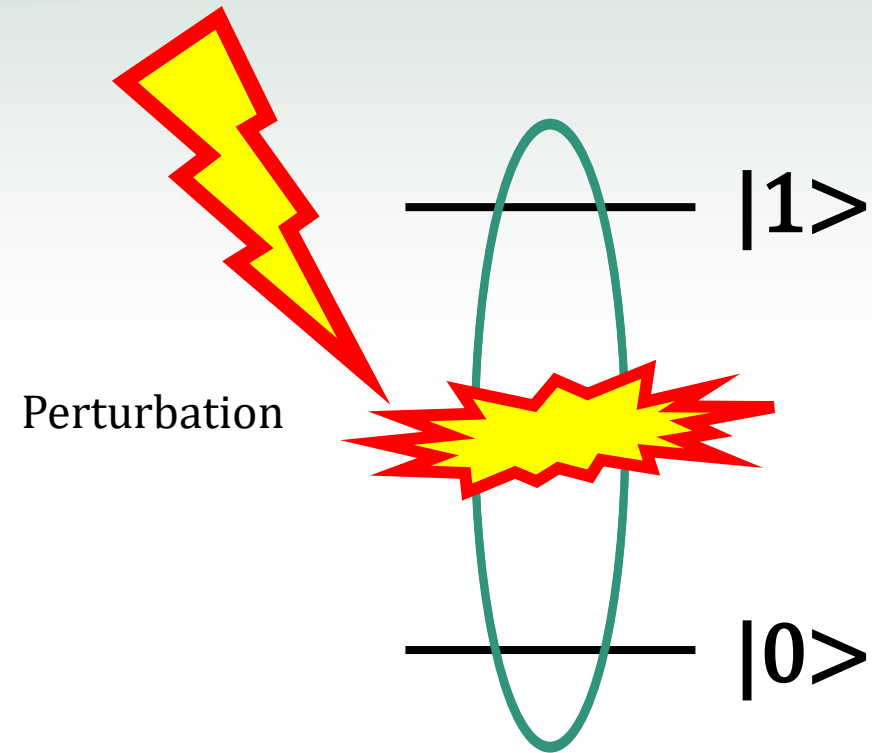


Quantum state superposition

# Need for stable qubits



Quantum state superposition

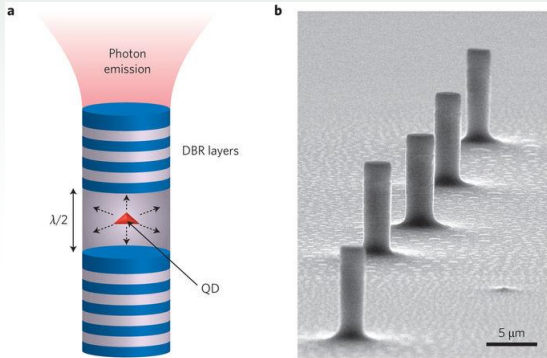


Decoherence induced by external perturbation

Coherence time  $T_2$

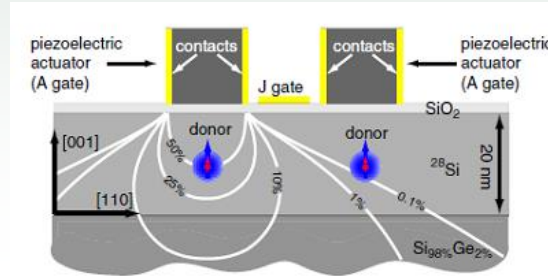
# Specially engineered solid-state materials

## InAs quantum dots



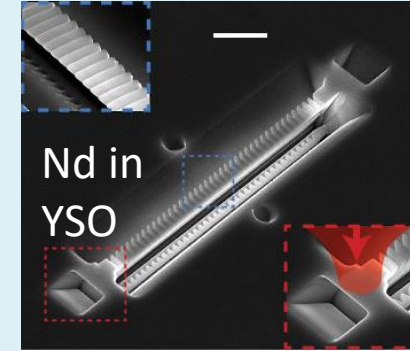
Nowak et al. Nat. Com. 2014

## Single P donors in Si



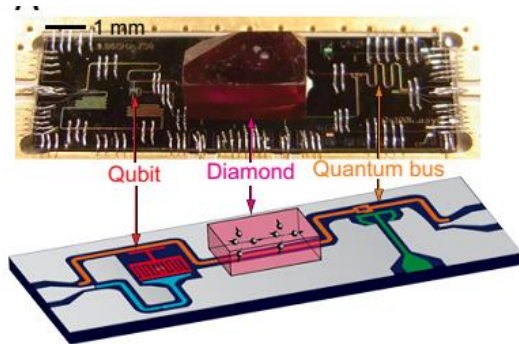
Dreher et al. PRL2011

## Rare earth ions in crystals



Zhong Nat Com 2015

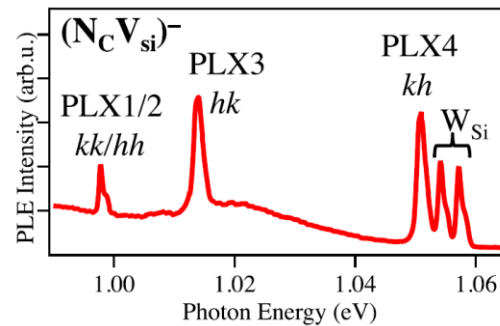
## NVs in diamond



Kubo et al. PRL2010

## Defects in semiconductors

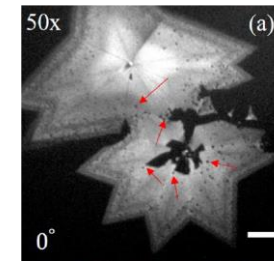
### 4H-SiC



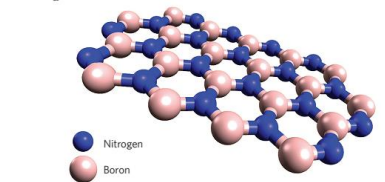
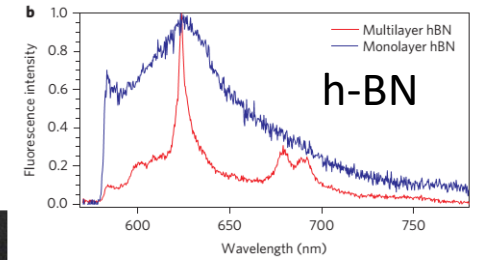
Zargaleh et al. PRB2016

## 2D materials

### WS<sub>2</sub>



Cao et al. 2D Mater 2017



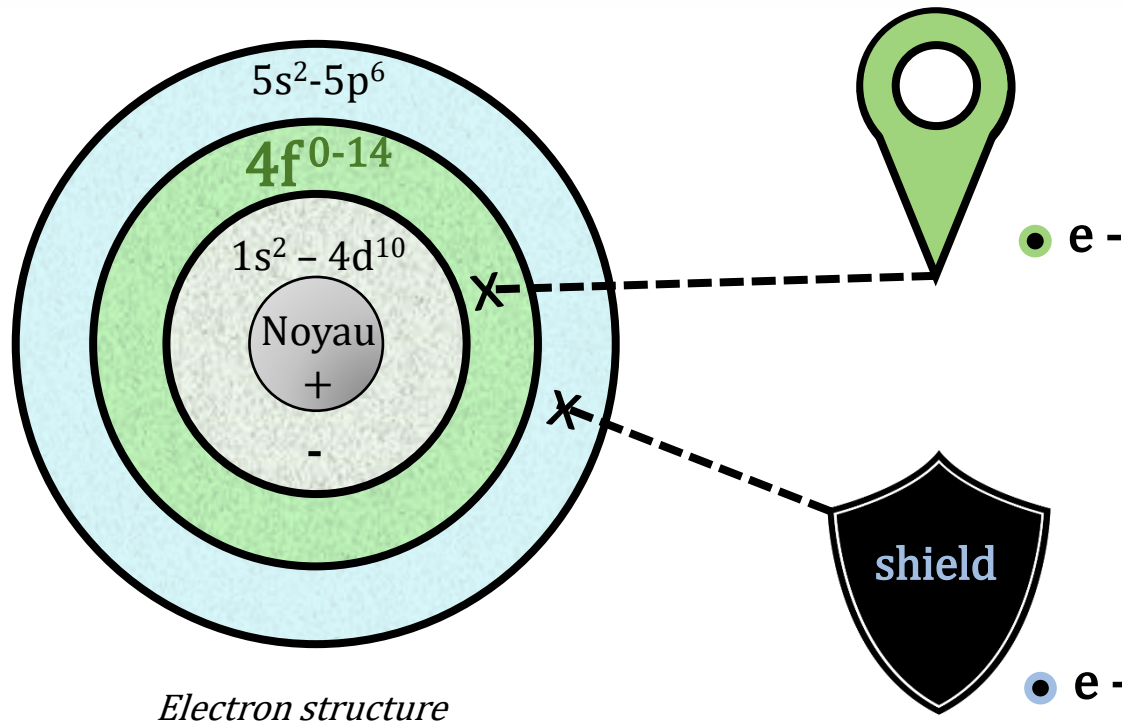
Ahoronovich Nature Nanotech 2016

G. Zhang, Y. Cheng, J.-P. Chou, A. Gali, Material platforms for defect qubits and single-photon emitters, Applied Physics Reviews. 7 (2020) 031308.

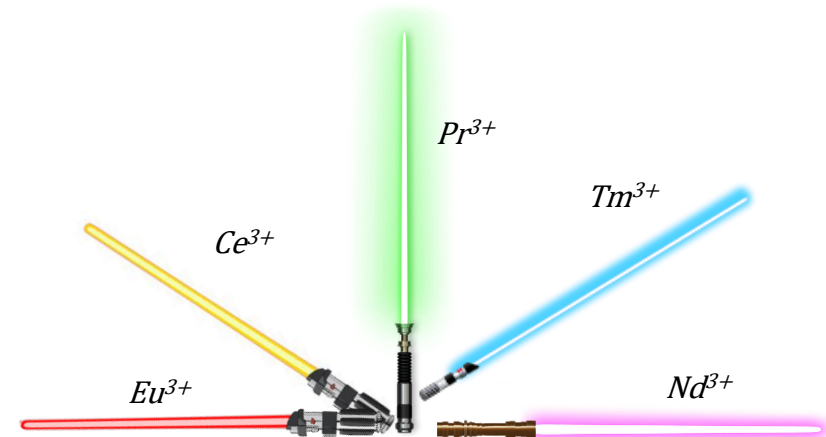
# Rare-earth ions

lanthane 57 La 138,90547	cérium 58 Ce 140,116	praséodyme 59 Pr 140,90765	néodyme 60 Nd 144,242	prométhium 61 Pm [144,9127]	samarium 62 Sm 150,36	europium 63 Eu 151,964	gadolinium 64 Gd 157,25	terbium 65 Tb 158,92535	dysprosium 66 Dy 162,500	holmium 67 Ho 164,93032	erbium 68 Er 167,259	thulium 69 Tm 168,93421	ytterbium 70 Yb 173,04	lutécium 71 Lu 174,967
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lanthanides  $[Ln^{3+}]$ :  $1s^2 2s^2 2p^6 3s^2 3p^6 4s^2 3d^{10} 4p^6 5s^2 4d^{10} 5p^6 4f^n$

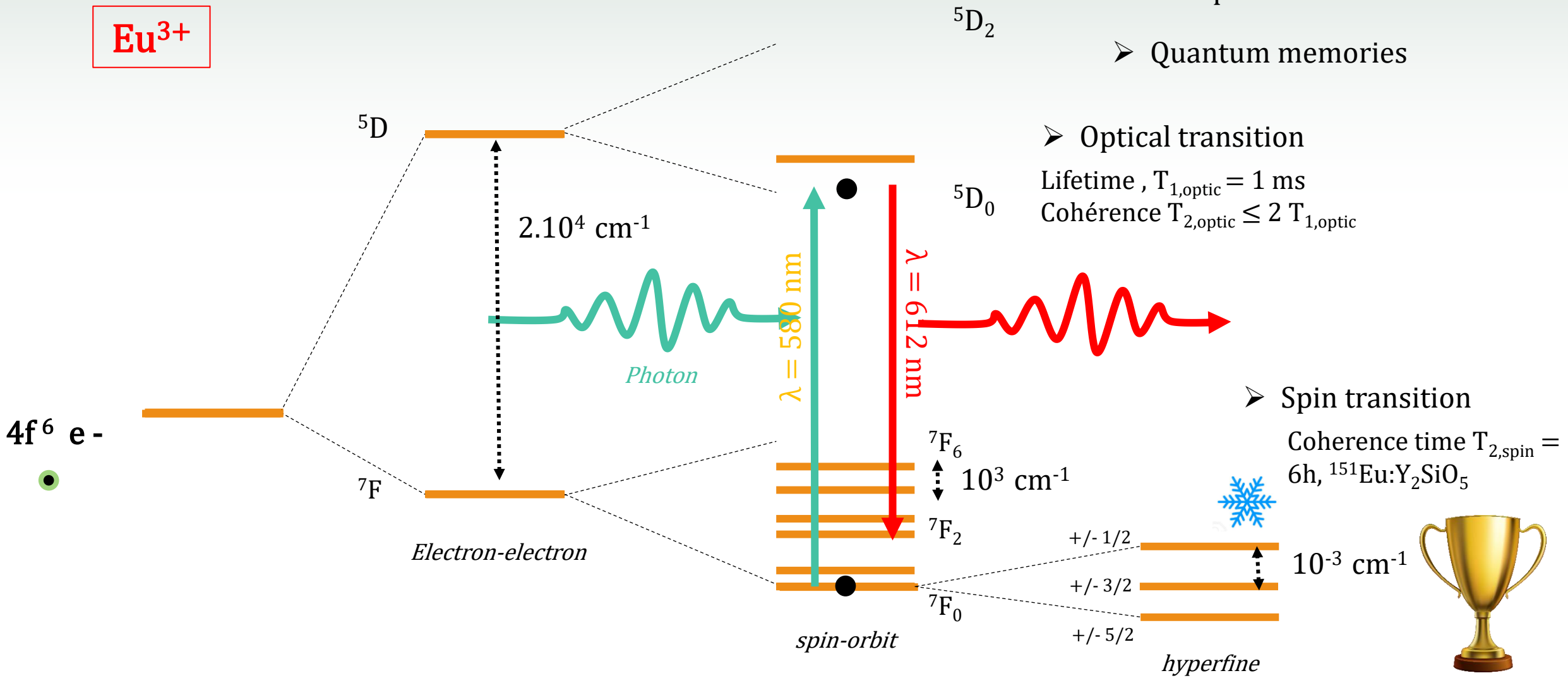


- Transitions involving **4f electrons**
- Screening by orbitals:  $5s^2 5p^6$
- Narrow optical lines



# Rare-earth ions

**Eu<sup>3+</sup>**



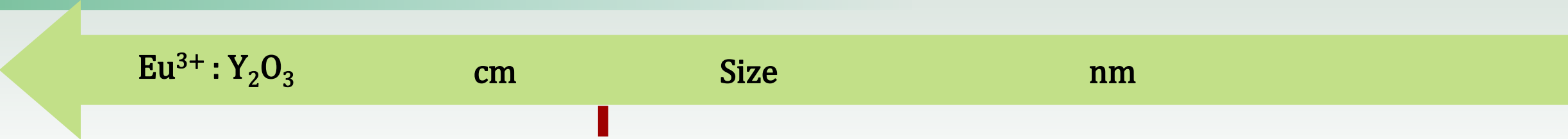
- MW-photon interface
- Quantum memories

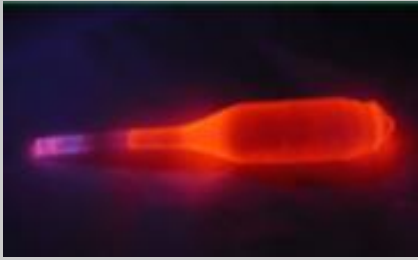

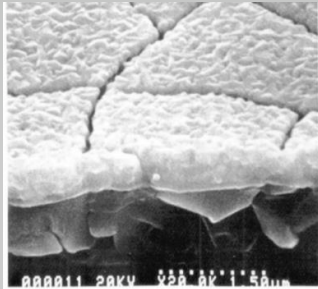

➤ Optical transition  
 Lifetime,  $T_{1,\text{optic}} = 1 \text{ ms}$   
 Coherence  $T_{2,\text{optic}} \leq 2 T_{1,\text{optic}}$

➤ Spin transition  
 Coherence time  $T_{2,\text{spin}} = 6\text{h}$ ,  $^{151}\text{Eu}:\text{Y}_2\text{SiO}_5$



# Material systems available



	cm	Size	nm	
Photo				
	Single crystal (YSO)	Nano-particles	CVD film 2μm	ALD film 100nm
Homog. Linew.	600 Hz	56 kHz	12 MHz	/
Optical T <sub>2</sub>	510 μs	5,67 μs	0,027 μs	/
Coherence	++	+	--	--
Integrability	--	-	++	++

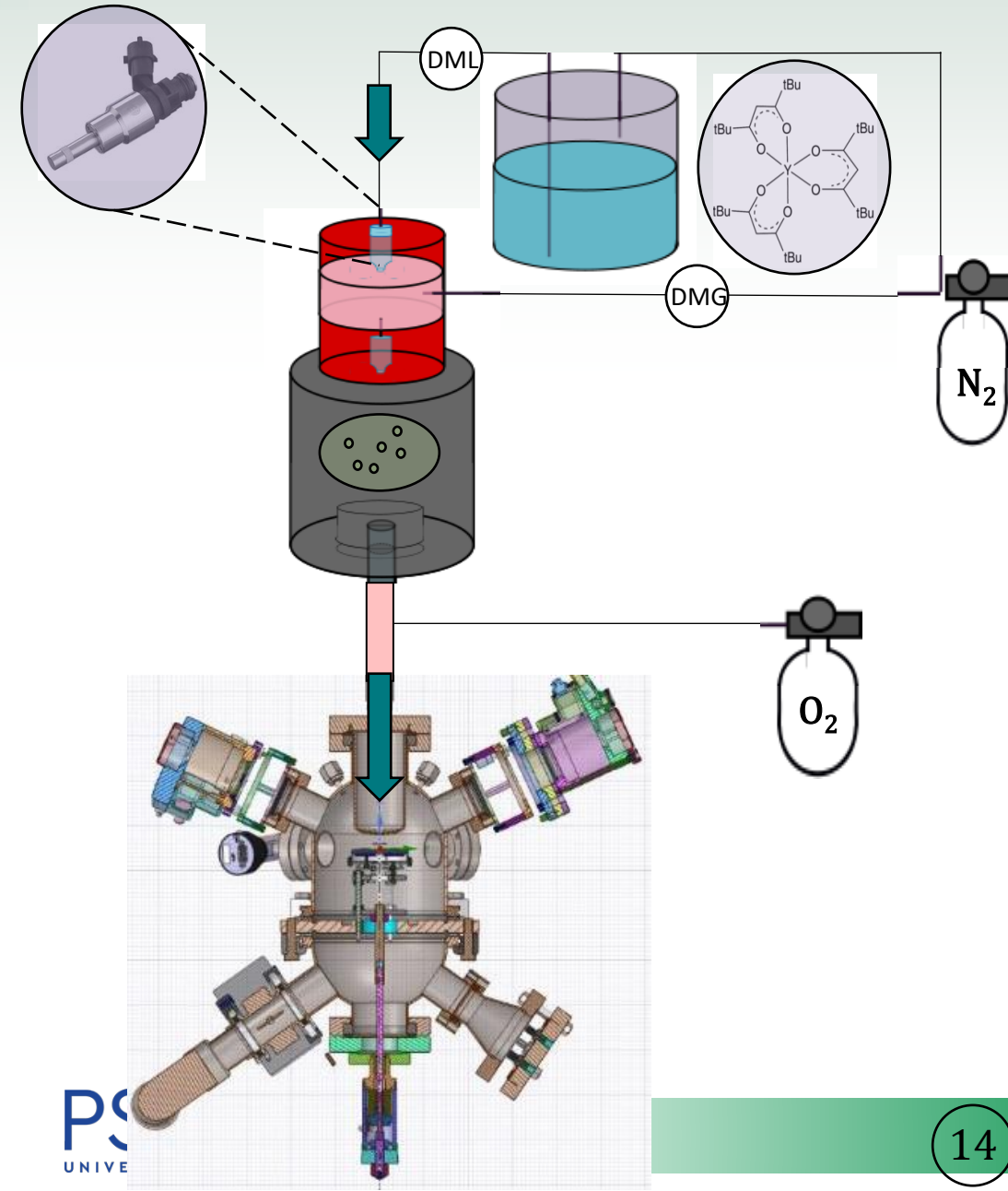
**Challenge:** evaluate and optimize coherent optical properties of rare-earth in CVD thin films

T. Zhong, P. Goldner, *Emerging rare-earth doped material platforms for quantum nanophotonics, Nanophotonics. 8 (2019) 2003–2015.*

## 2. Thin oxide films by direct liquid injection CVD

# DLI-CVD technique

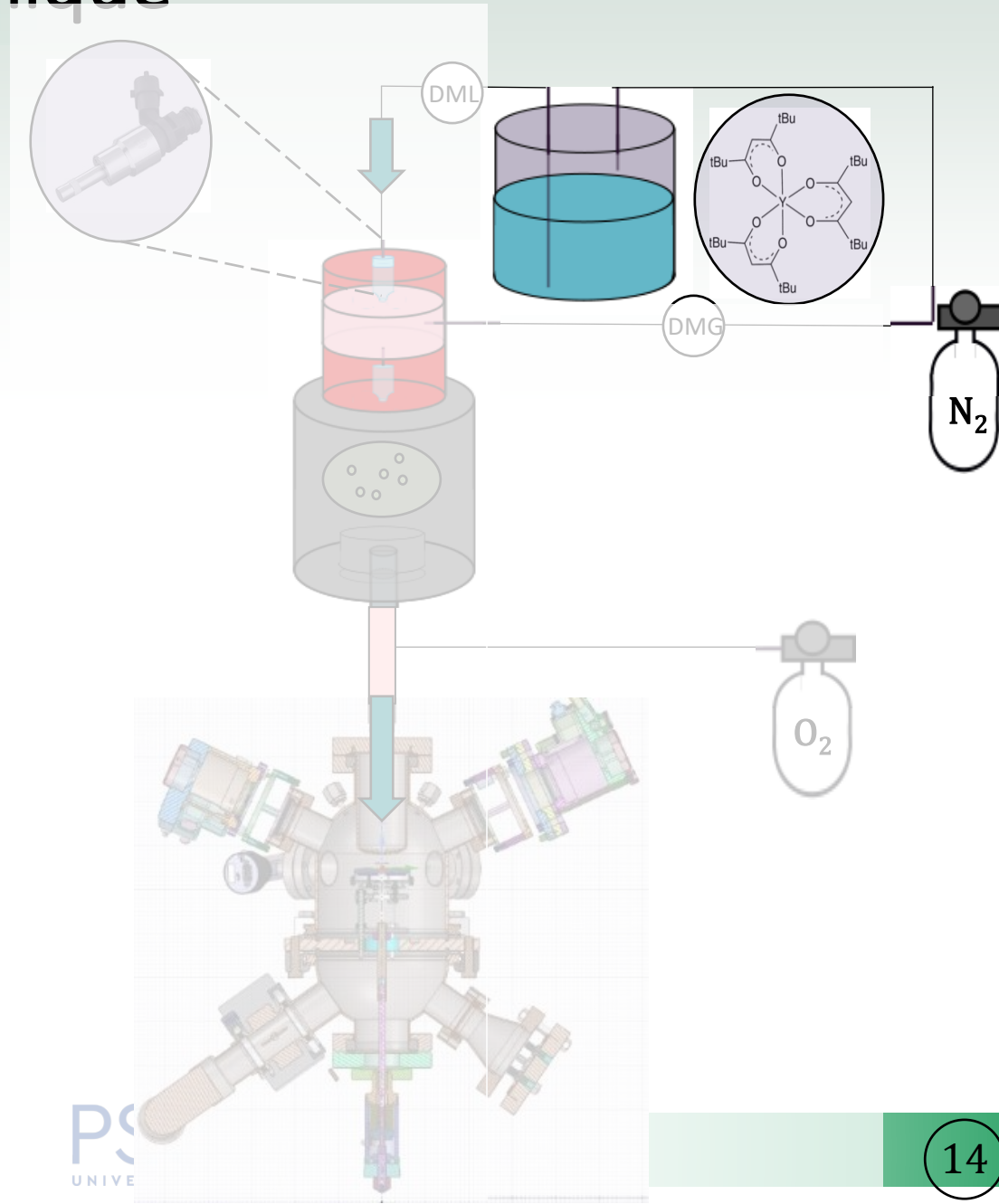
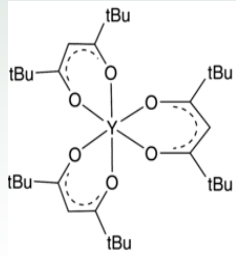
Eu:Y<sub>2</sub>O<sub>3</sub>



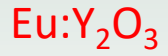
# DLI-CVD technique

$\text{Eu:Y}_2\text{O}_3$

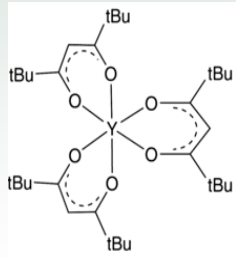
- B-diketonate Y or  $\text{Eu}(\text{thd})_3$



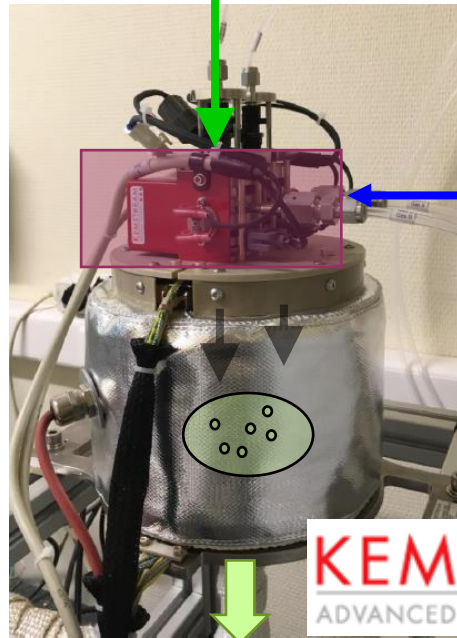
# DLI-CVD technique



- B-diketonate Y or  $\text{Eu}(\text{thd})_3$



Liquid

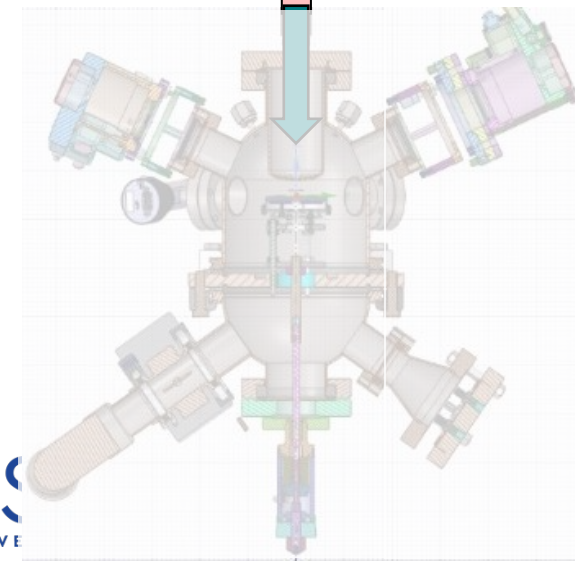
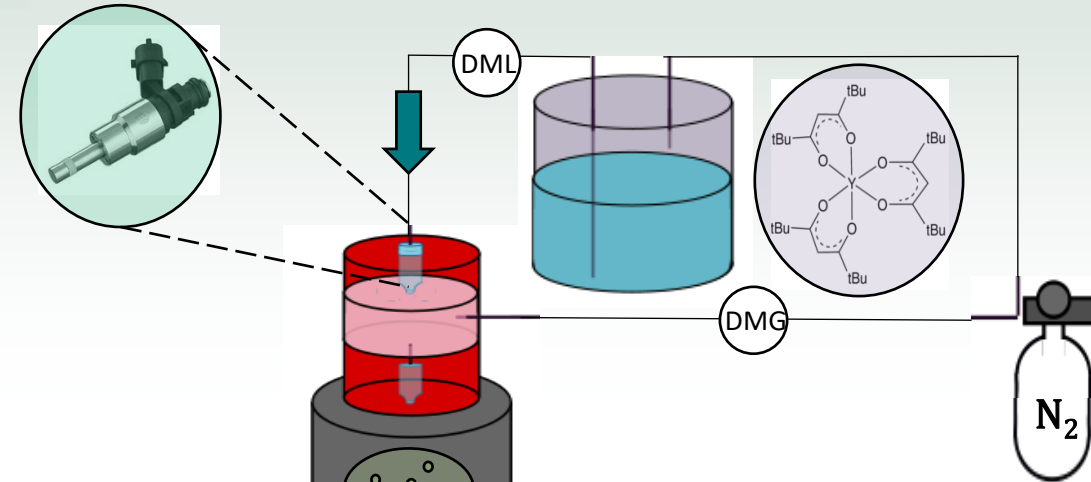


Injection

Gas

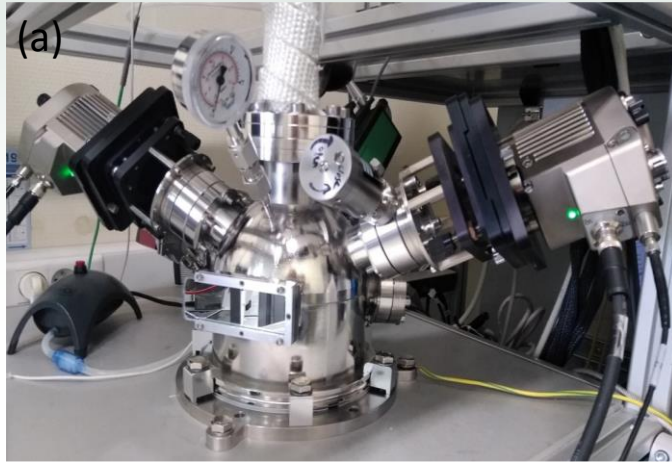
Vaporisation @  
200 °C

**KEMSTREAM**  
ADVANCED VAPORIZERS

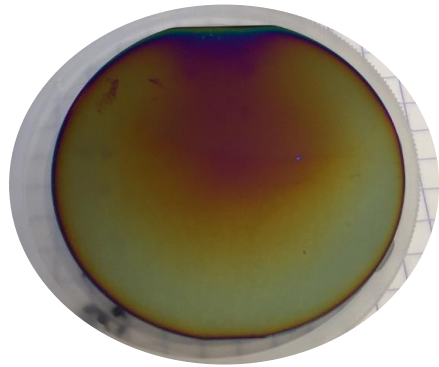


V. Astié, C. Millon, J. M. Decams, A. Bartasyte, Direct liquid injection chemical vapor deposition, *Chemical Vapor Deposition for Nanotechnology*, IntechOpen, 2019 pp. 2–23.

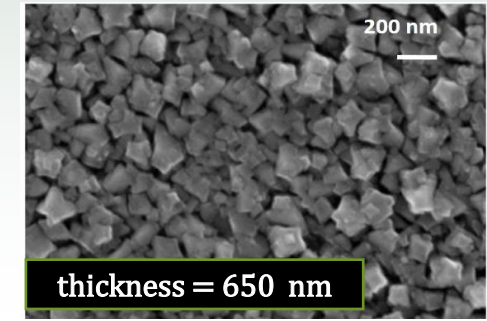
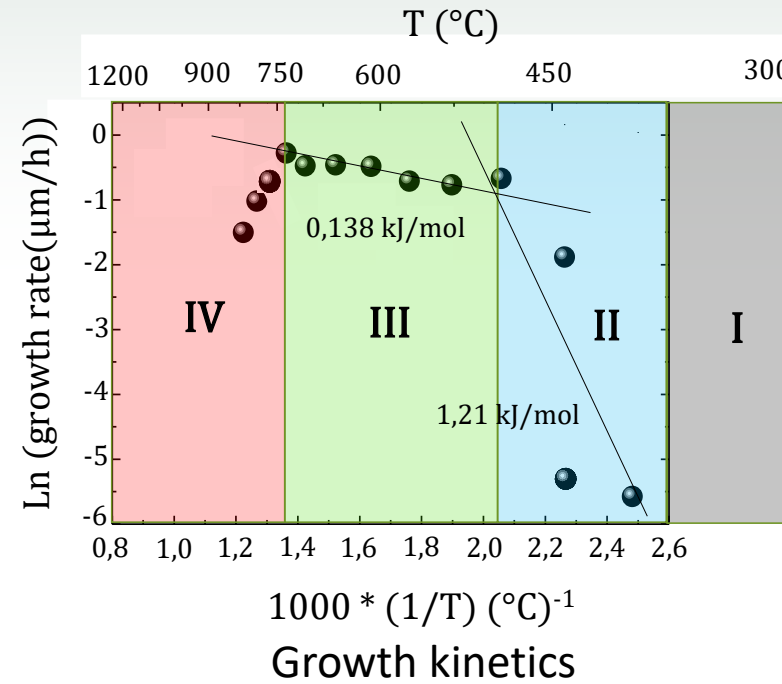
# Growth of thin Eu:Y<sub>2</sub>O<sub>3</sub> films



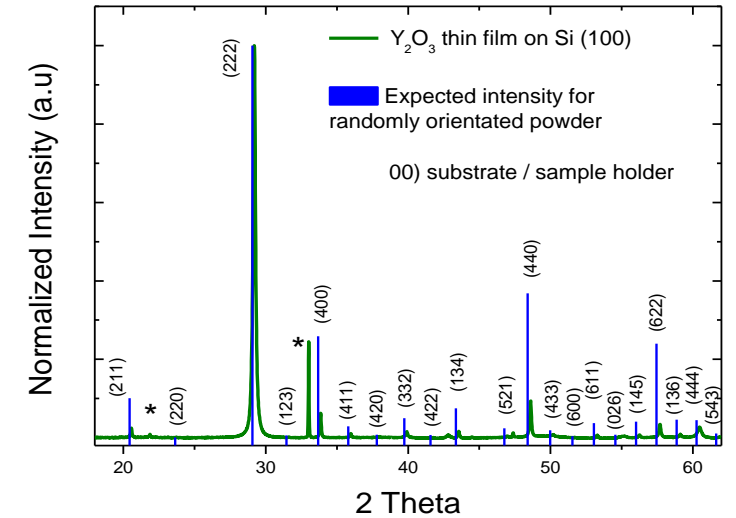
DLI-CVD reactor with in-situ ellipsometer



2 inch Si (111) wafers



Polycrystalline on Si



Highly (111) textured

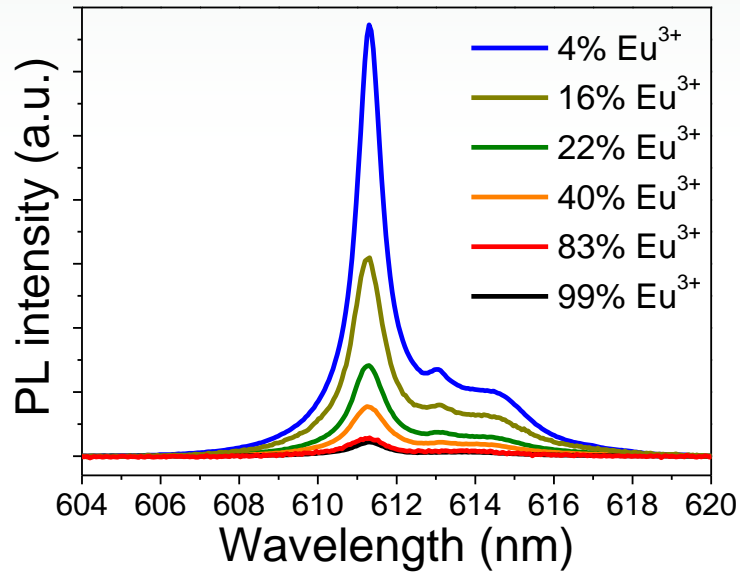
*N. Harada et al. Chemically vapor deposited Eu<sup>3+</sup>:Y<sub>2</sub>O<sub>3</sub> thin films as a material platform for quantum technologies, Journal of Applied Physics. 128 (2020) 055304*

# 3. Assessment of optical properties using advanced spectroscopy

# Optical properties

## PL emission

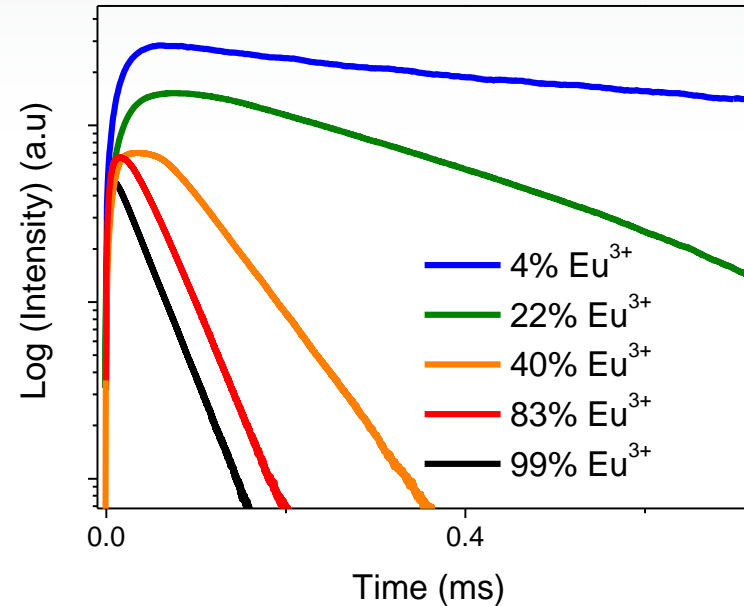
$\lambda_{\text{excitation}} = 532 \text{ nm}$



Successful doping 0-99%  
**PL drops above 6%**  
 (concentration quenching)

## Decay rate

$\lambda_{\text{excitation}} = 465 \text{ nm}$

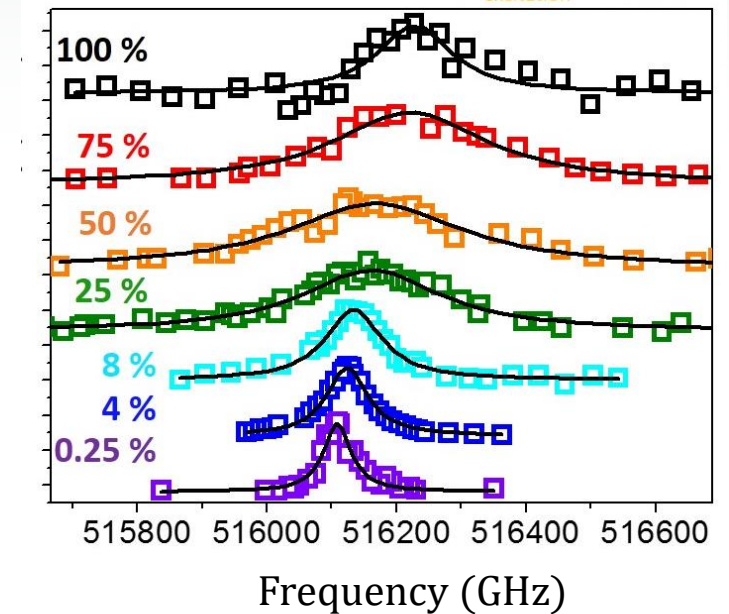


**Optical  $\tau_1 = 1 \text{ ms}$**   
 Comparable to bulk  
 Decrease with doping

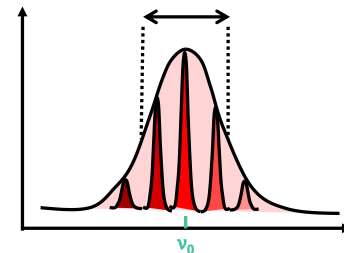
## Inhomogeneous broadening

$\lambda_{\text{excitation}} \sim 580.88 \text{ nm}$

@ 12K

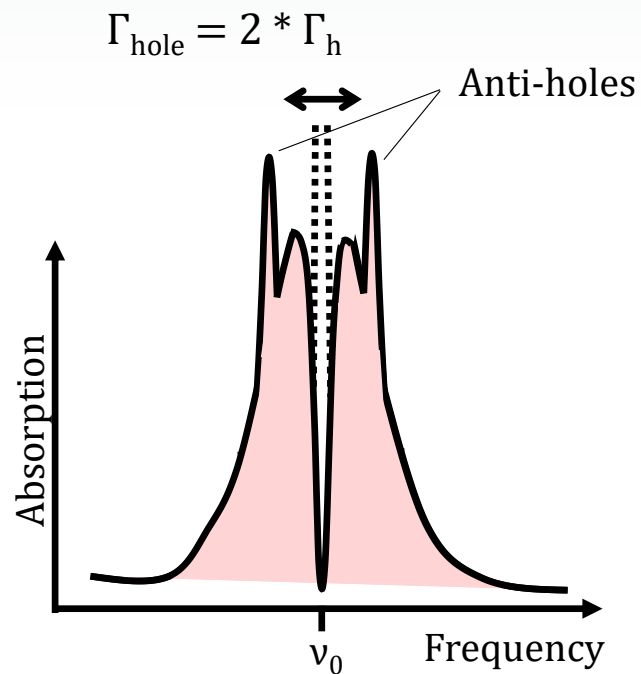
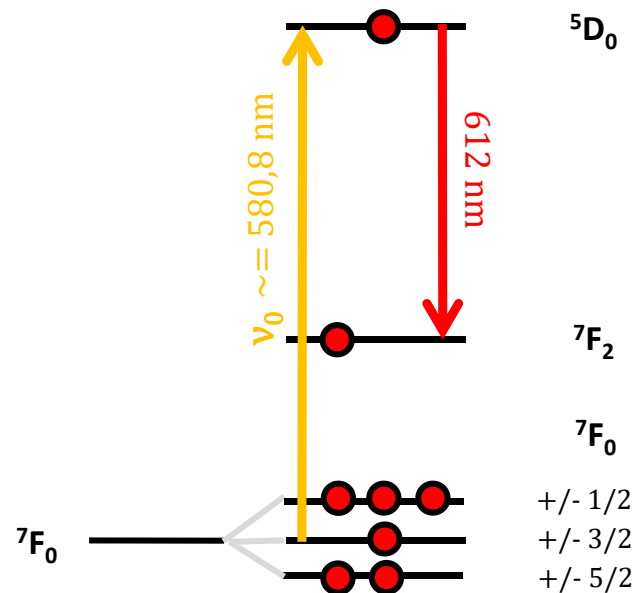
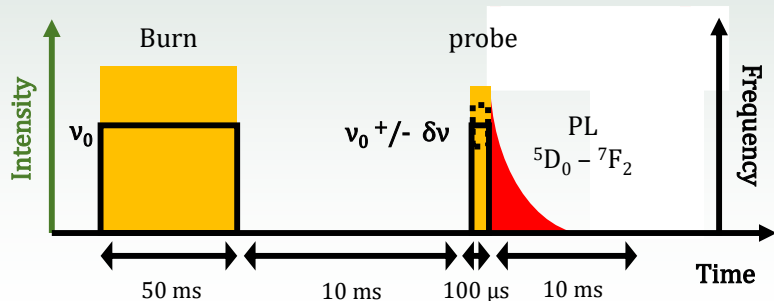


**$\Gamma_{\text{inh}} = 50 \text{ GHz @ 2\%$**   
 10x that of bulk



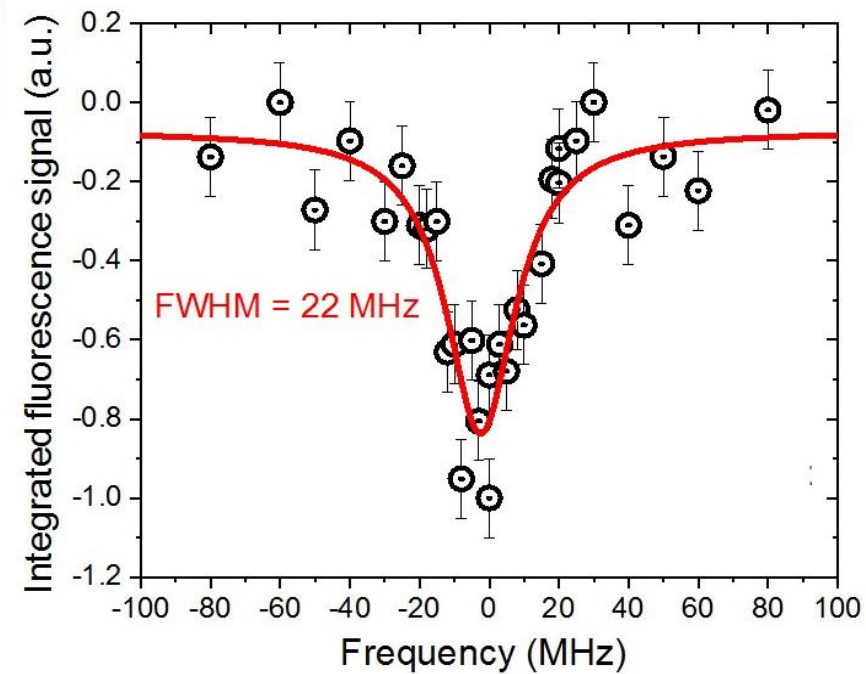


# Spectral hole burning



Working principle of SHB

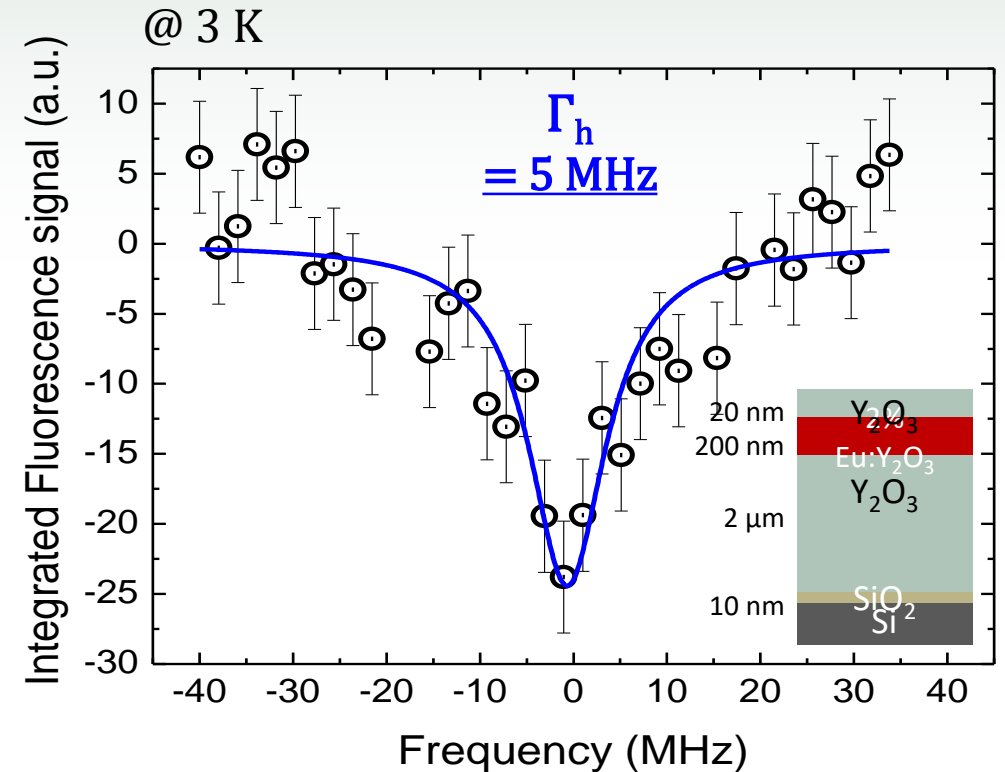
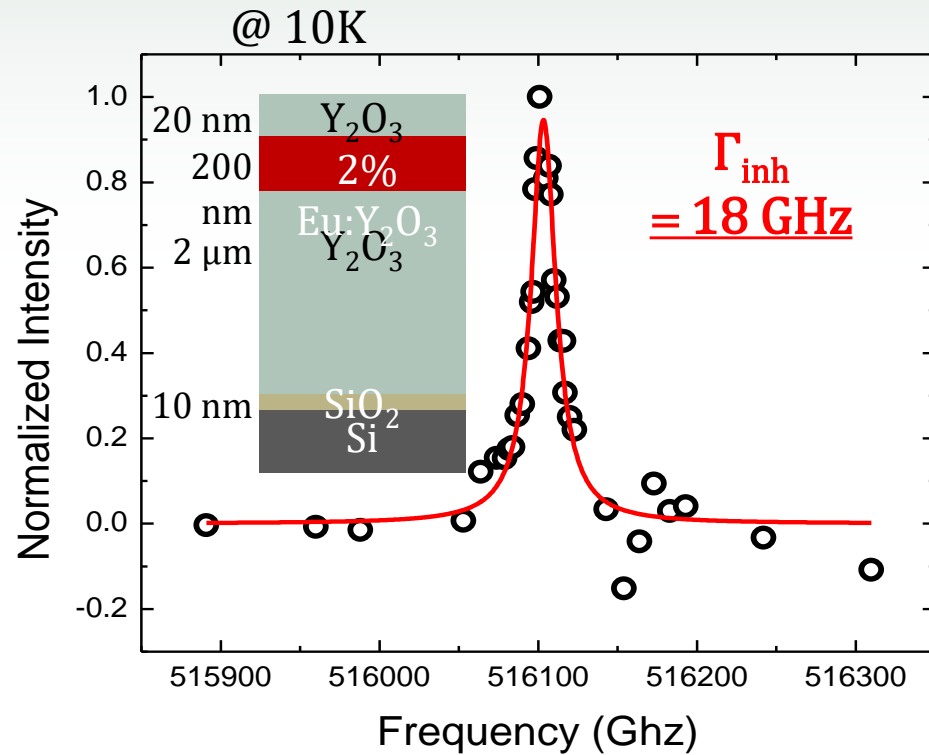
SHB for a 200nm Eu:Y<sub>2</sub>O<sub>3</sub> thin film on Si @3K



$\Gamma_h = 11\text{MHz @2\%}$   
1000X higher than bulk

# SHB on optimized thin film

Thicker multilayer with 1000°C annealing

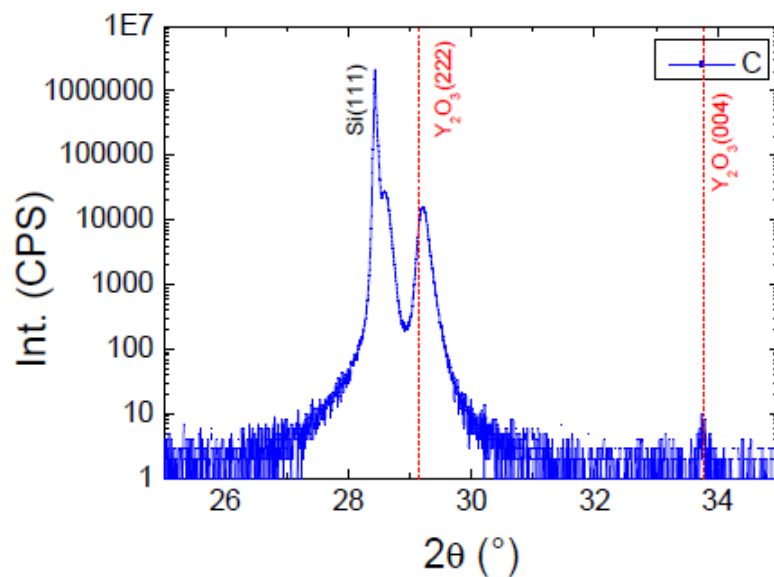
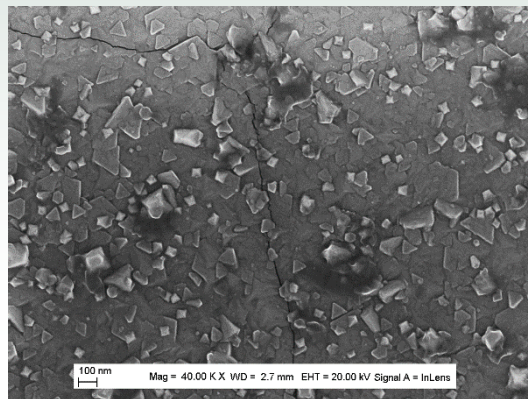
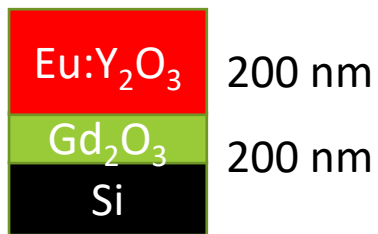


Improvement by a factor of 2 of both inhomogeneous and homogeneous linewidth.  
However still relatively broad compared to bulk and nanoparticles → more efforts needed

N. Harada et al. Controlling the interfacial reactions and environment of rare-earth ions in thin oxide films towards wafer-scalable quantum technologies, Mater. Adv. (2021).

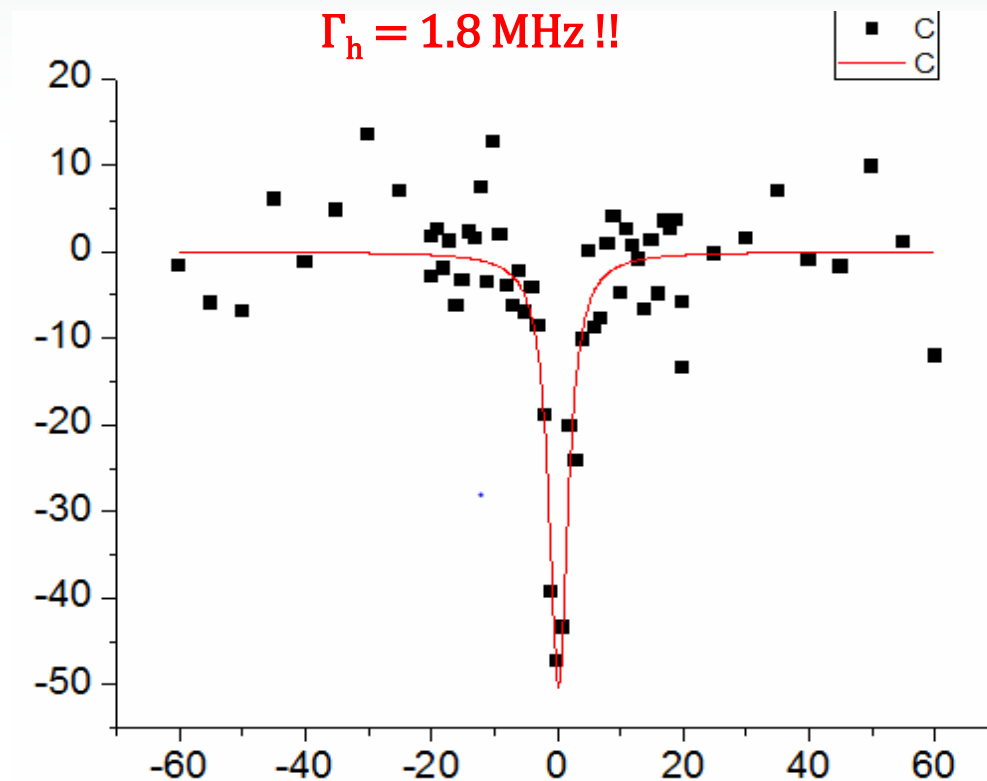
# Perspective: towards epitaxial layers

Growth on a commercial **epitaxial MBE template** (Gd<sub>2</sub>O<sub>3</sub>/Si)



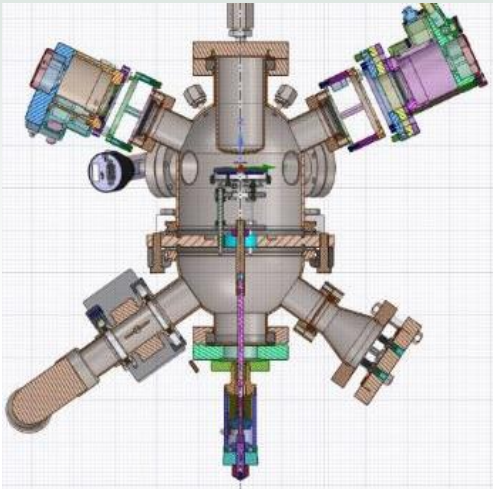
Collab. INL (R. Bachelet)

SHB measurement



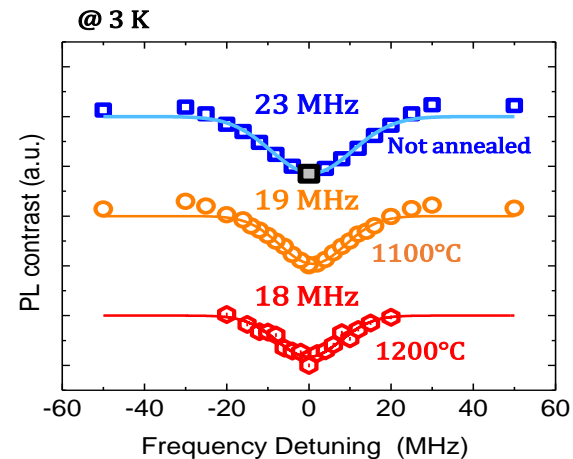
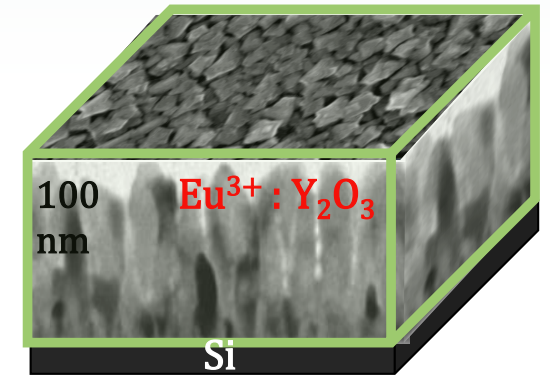
Very encouraging!

# Conclusion



Development of a new and versatile DLI-CVD process for RE-Y<sub>2</sub>O<sub>3</sub> thin films

Synthesis of Eu:Y<sub>2</sub>O<sub>3</sub> on Si with (111) texture and in a wide doping range



Assessment of optical properties within the context of quantum technologies → homogeneous linewidth of 1-5 MHz

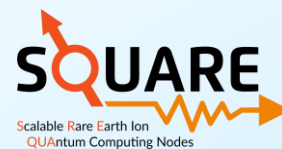
**A new potential platform for QTs with improved integrability**

Our team @IRCP: Crystals and quantum state dynamics



Nao Harada

Funding



**THANKS FOR  
YOUR ATTENTION!**

