

Hybrid CVD-MBE Er:Y₂O₃ thin films for on-chip quantum technologies

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Introduction

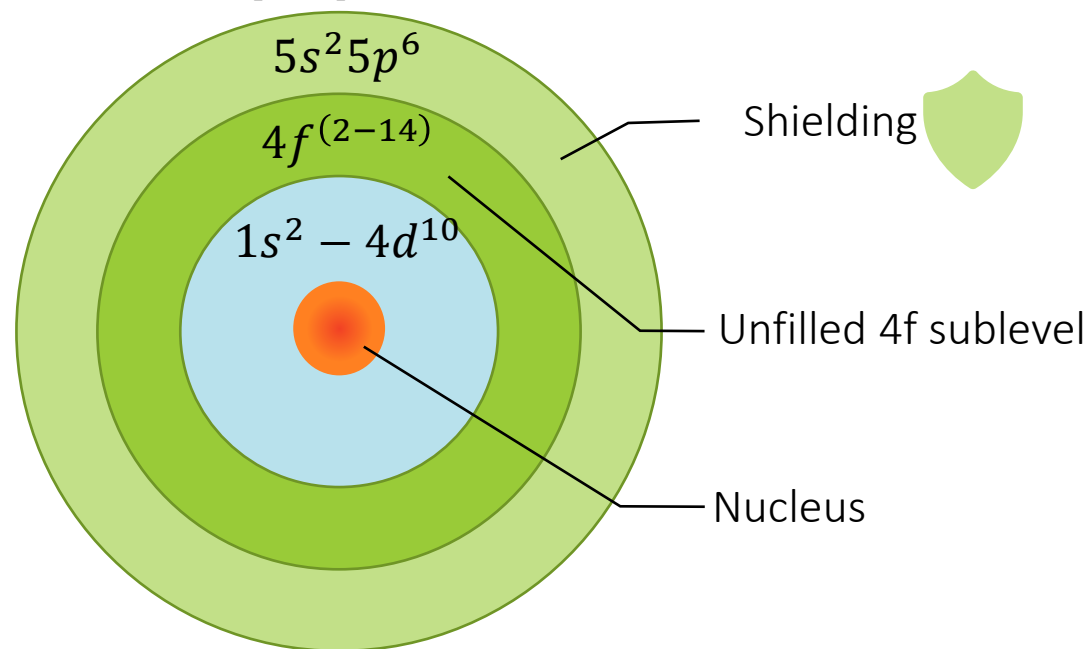
Introduction

The rare-earth ions

The lanthanides- [Ln]

57 La Lanthanum	58 Ce Cerium 140	59 Pr Praseodymium 141	60 Nd Neodymium 144	61 Pm Promethium	62 Sm Samarium 150	63 Eu Europium 152	64 Gd Gadolinium 157	65 Tb Terbium 159	66 Dy Dysprosium 162.5	67 Ho Holmium 165	68 Er Erbium 167	69 Tm Thulium 169	70 Yb Ytterbium 173	71 Lu Lutetium 175
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➤ Lanthanide ions $[Ln^{3+}]$: $1s^2 2s^2 2p^6 3s^2 3p^6 4s^2 3d^{10} 4p^6 5s^2 4d^{10} 5p^6 4f^n$

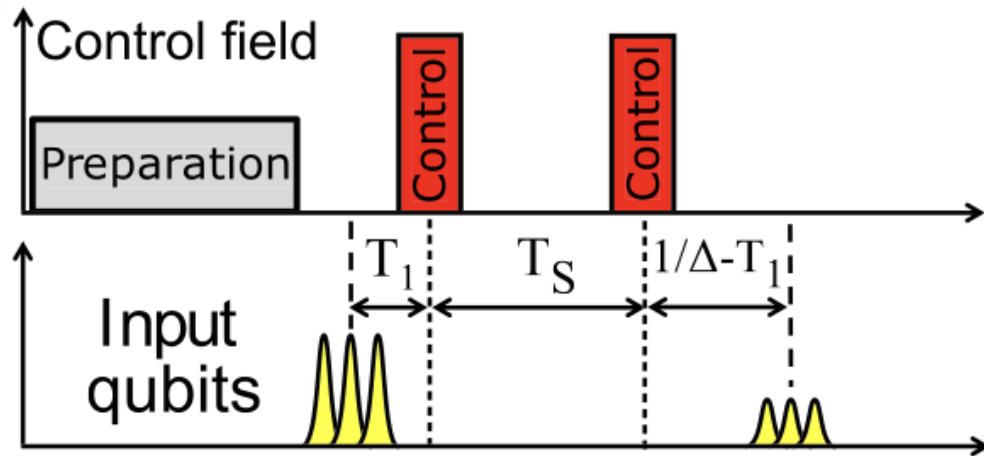


- Shielding by the electron of the $5s^2$ and $5p^6$ sublevels
- Narrow transitions, almost unaffected by the crystal environment
- Long optical and spin coherence time at low temperature

Rare earth Lanthanide ion $[Ln^{3+}]$ atomic structure

Introduction

The rare-earth ions for quantum applications : quantum memories

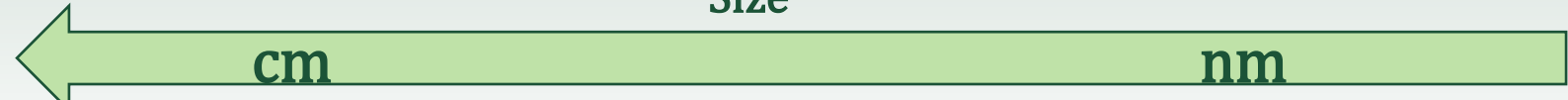


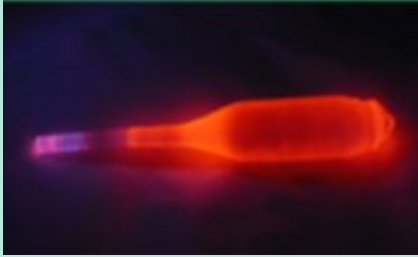

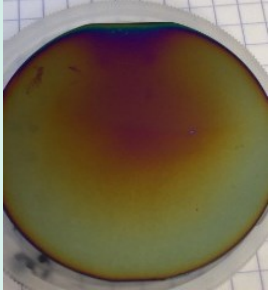
- Businger et al., *Non-classical correlations over 1250 modes between telecom photons and 979-nm photons stored in $^{171}\text{Yb}^{3+}:\text{Y}_2\text{SiO}_5$* , *Nat. Commun.* 13, 6438 (2022)
- Askarani et al., *Long-Lived Solid-State Optical Memory for High-Rate Quantum Repeaters*, *Phys. Rev. Lett.* 127(22), 220502 (2021).
- Kutluer et al. *Time Entanglement between a Photon and a Spin Wave in a Multimode Solid-State Quantum Memory*, *Phys. Rev. Lett.* 123 (2019)
- Zhong et al. *Nanophotonic rare-earth quantum memory with optically controlled retrieval*, *Science* 357 (2015)
- Heinze et al., *Stopped light and image storage by electromagnetically induced transparency up to the regime of one minute*, *Phys. Rev. Lett.* 111 (3), (2013)

Introduction

Quantum applications

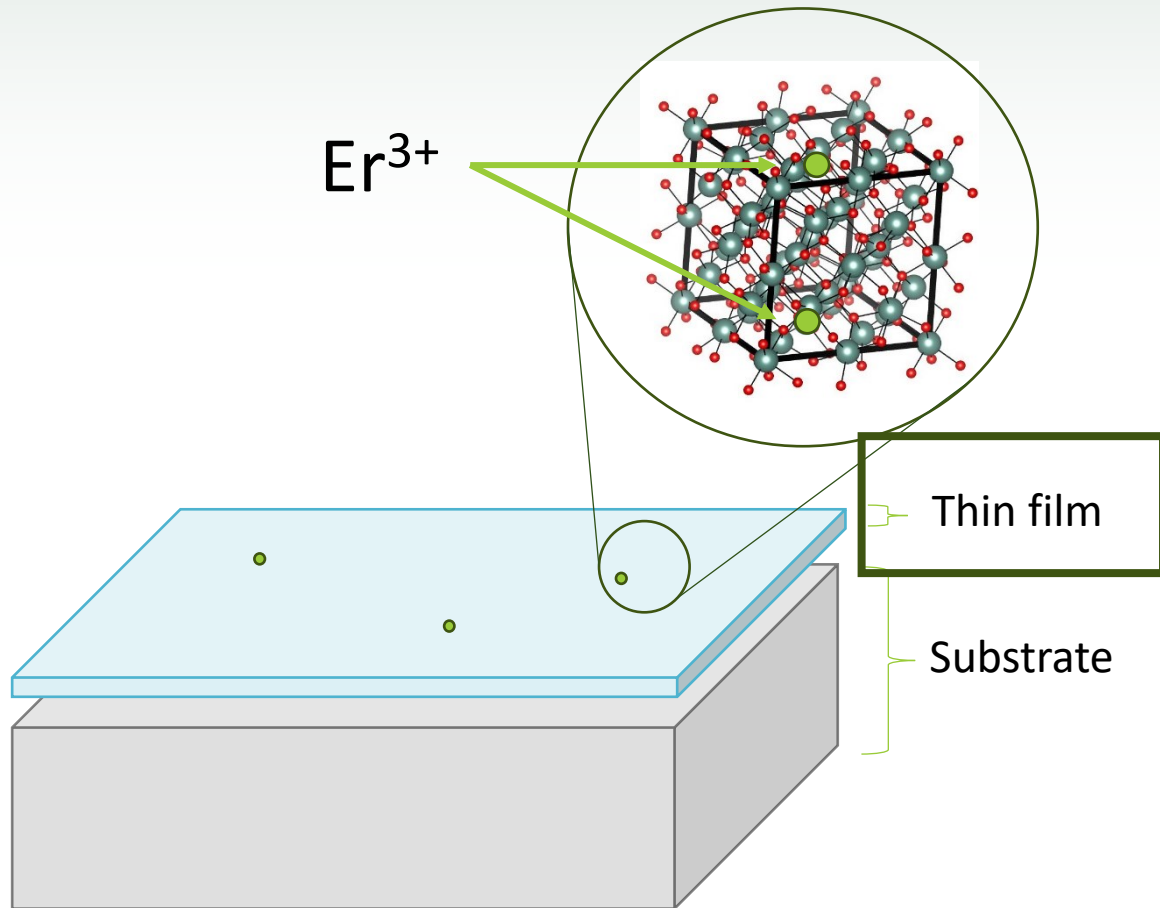
Size



System	Bulk material (YSO)	Nano-particules (Y_2O_3)	Thin films (Y_2O_3)
	 <p>Monocrystal (under UV lamp)</p>	 <p>NPs (100 nm) (under UV lamp)</p>	 <p>DLI-CVD films 200 nm on Si</p>
RE doping	0,3 %Eu	0,3 %Eu	2 %Eu
Inhomogeneous linewidth	5 GHz	11 GHz	90 GHz
Homogeneous linewidth	600 Hz	56 kHz	12 MHz
=	=	-	-
Optical T_2	510 μ s	5,67 μ s	0,027 μ s
References	G. P. Flinn et al. ; <i>Phys. Rev. B</i> , 49 , 5821 (1994)	S. Liu et al. , <i>ACS Nano</i> , 14 , 9953–9962 (2020)	G. A. West & K. W. Beeson, <i>J. Mater. Res.</i> , 5 , 1573–1580 (1990)

Introduction

Material



Thin films:

- Grown by Direct Liquid Injection CVD
- Possible integration : resonators, waveguides
- Research to optimize coherence properties

Y_2O_3 :

- Chemical compatibility with RE
- Binary compound
- IR transparency
- Long coherence time possible (spin and optical)

Er^{3+} :

- Emission wavelength in the telecom C-band
- Paramagnetic with an electronic spin
- Substitution of Y: two possible sites C_2 and C_{3i}

Introduction

Different substrates and their impact on epitaxy

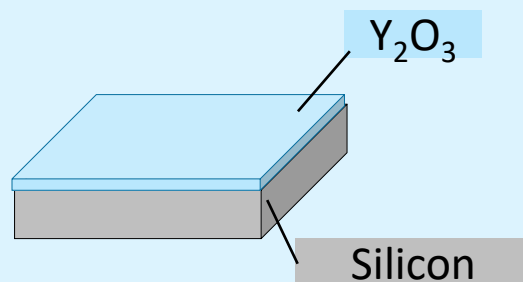
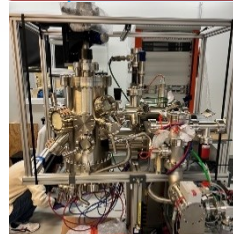
Silicon :

- Si(100)
- Si(111)



- Commercially available ✓
- Possible to remove the substrate from the film ✓
- Direct epitaxial growth of Y_2O_3 very difficult ✗

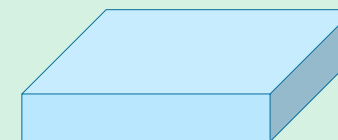
MBE template :



- Interface more favorable for epitaxy ✓
- Possible to remove the substrate from the film ✓

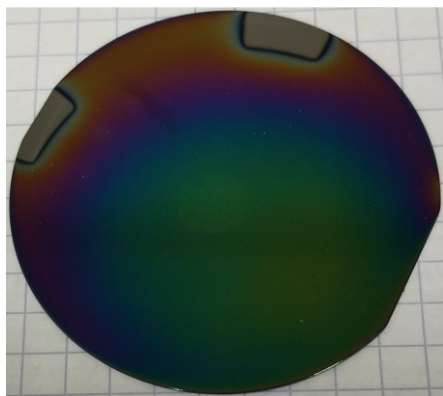
Oxides :

- Sapphire
- Quartz
- YSZ (Yttria Stabilized Zirconia)

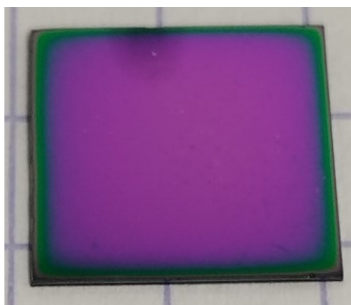


- Interface more favorable for epitaxy ✓
- Difficult substrate removal ✗

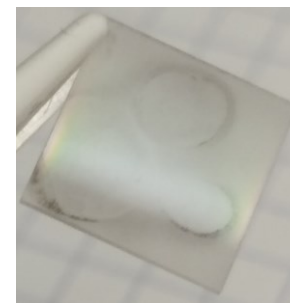
Results



Er³⁺ : Y₂O₃ on Si(100)



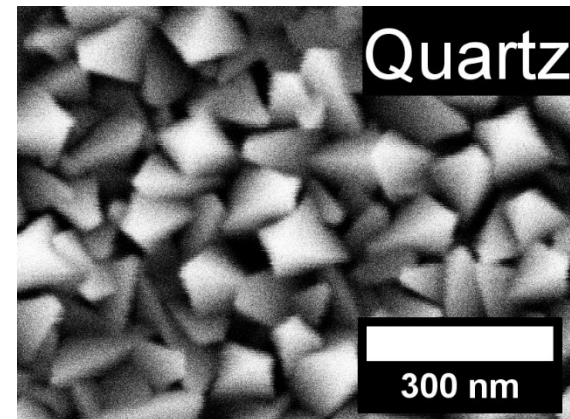
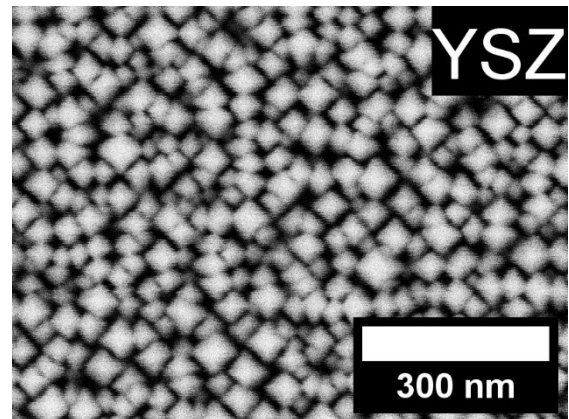
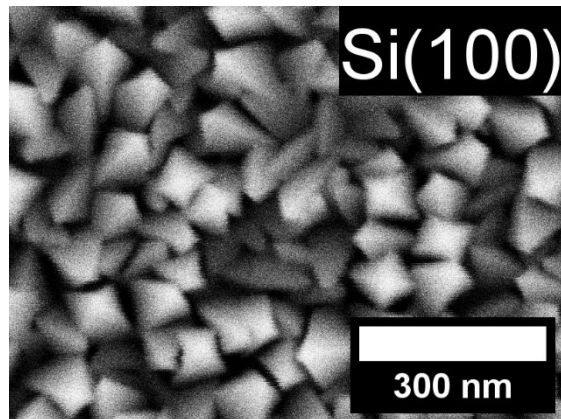
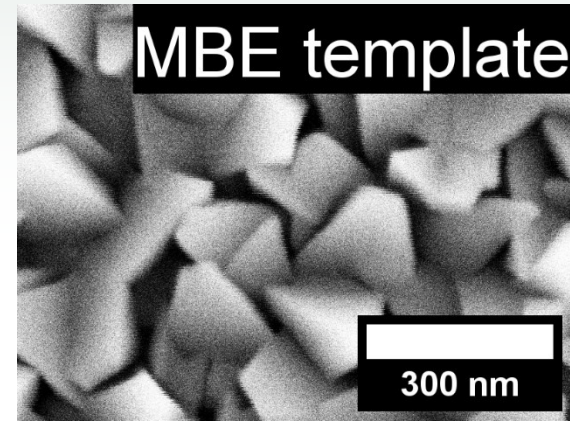
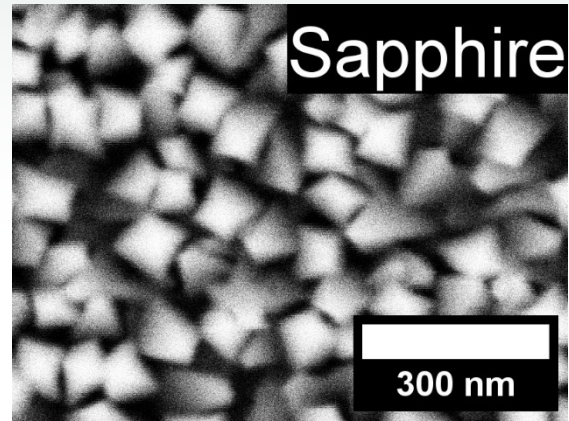
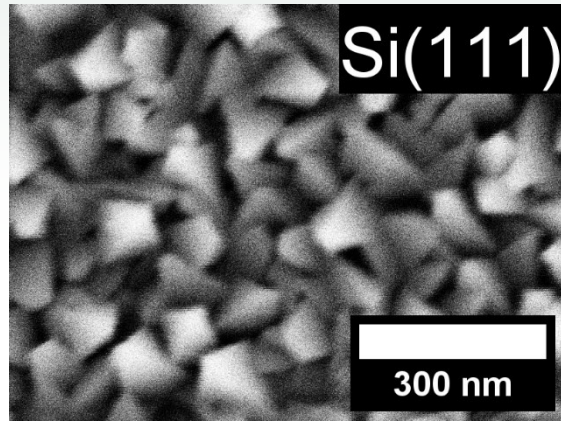
Er³⁺ : Y₂O₃ on
MBE template



Er³⁺ : Y₂O₃ on YSZ

Results

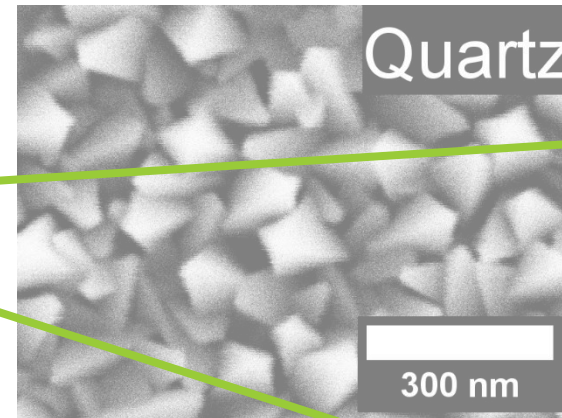
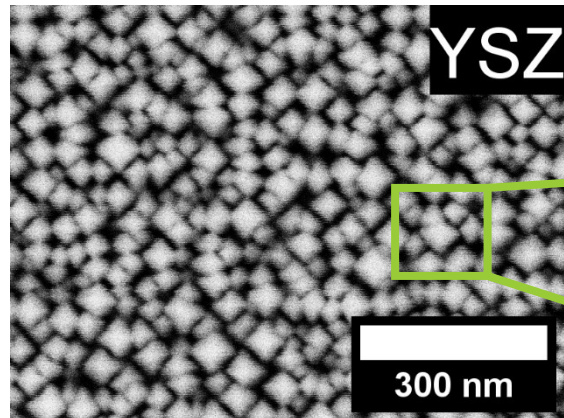
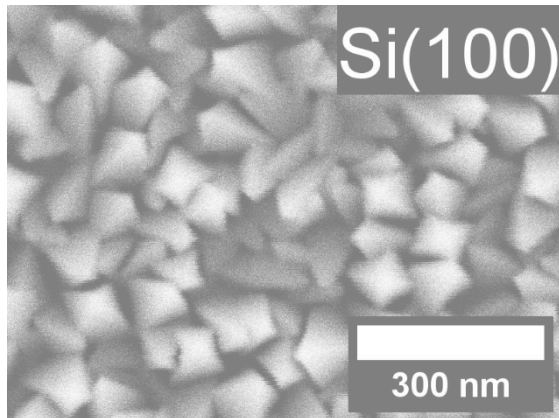
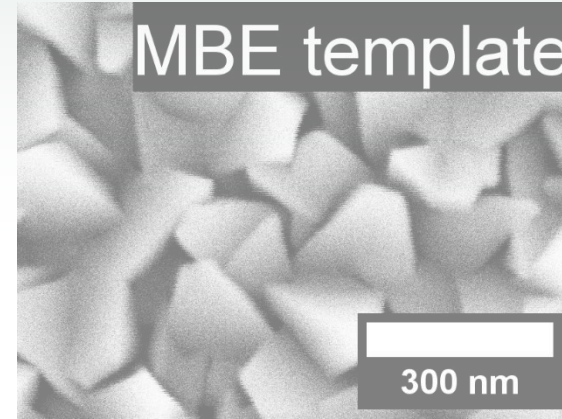
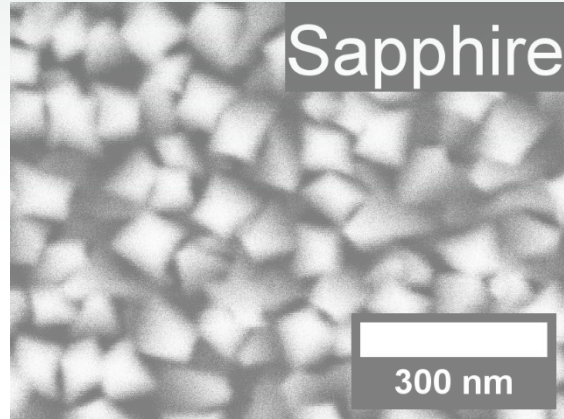
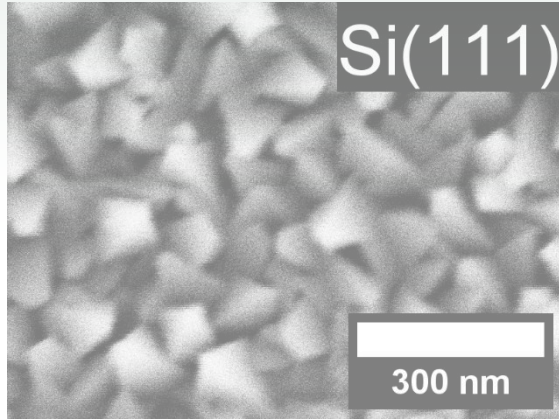
Morphological properties: SEM images



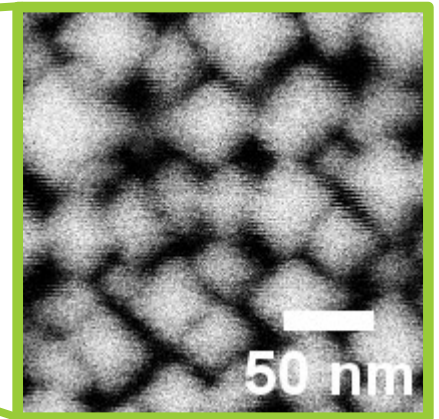
- Crystallized films

Results

Morphological properties: SEM images

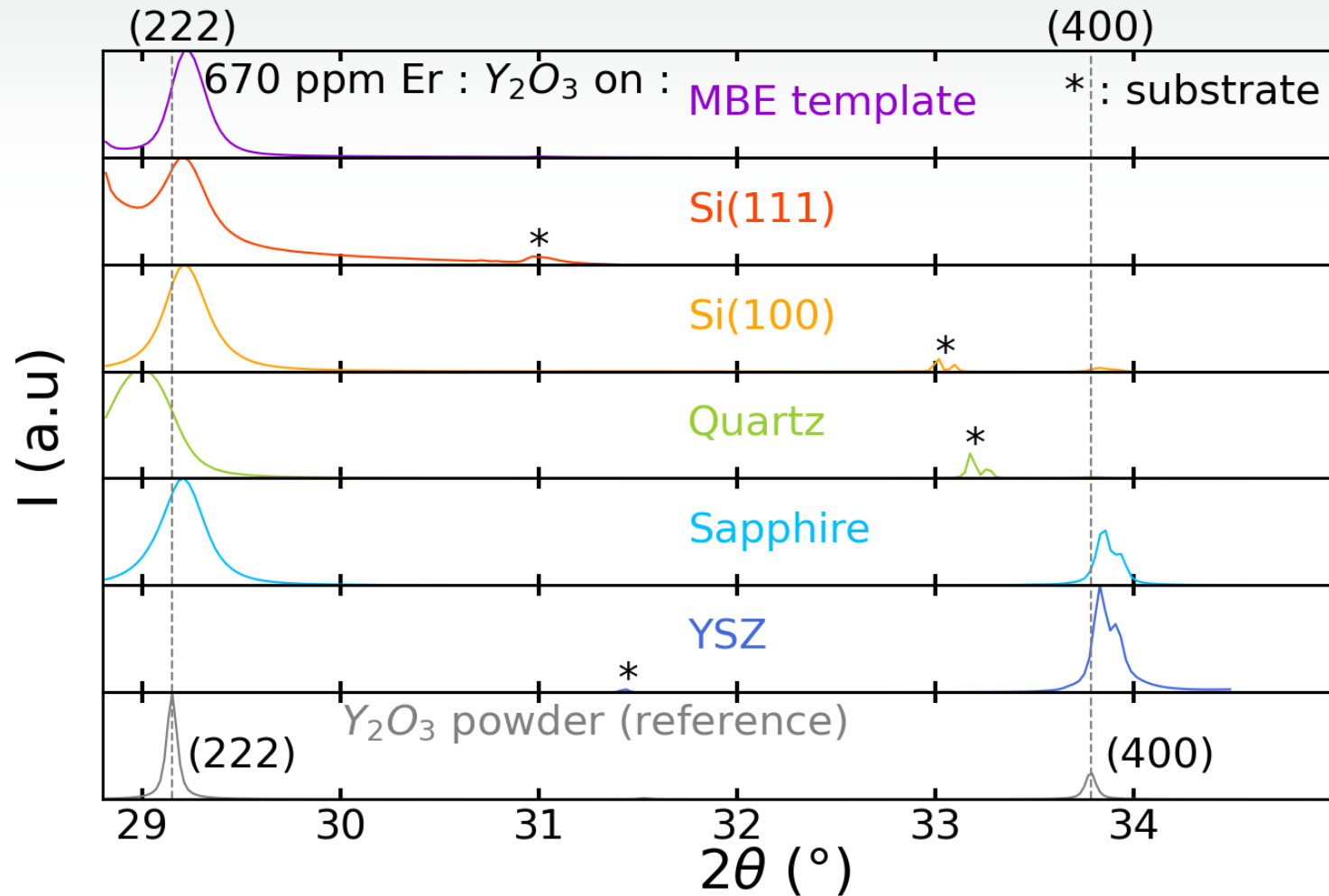


- Crystallized films
- YSZ: grains with square shape, smaller and aligned → Epitaxy ?



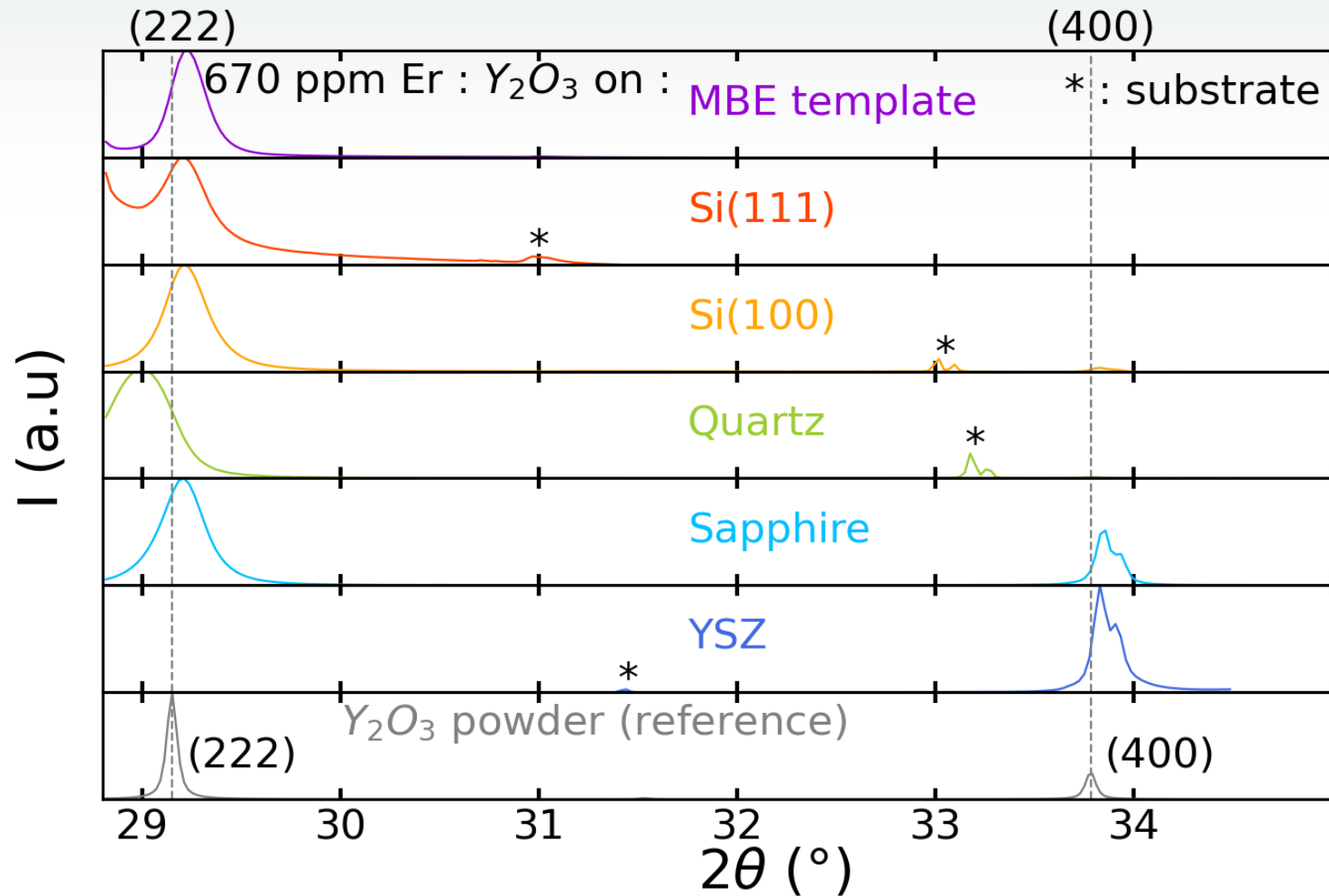
Results

Structural properties: X-ray diffraction



Results

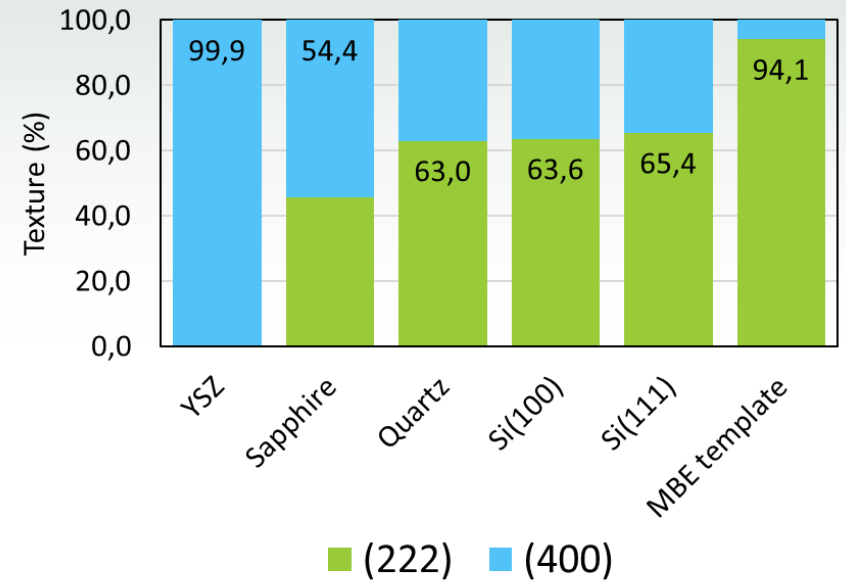
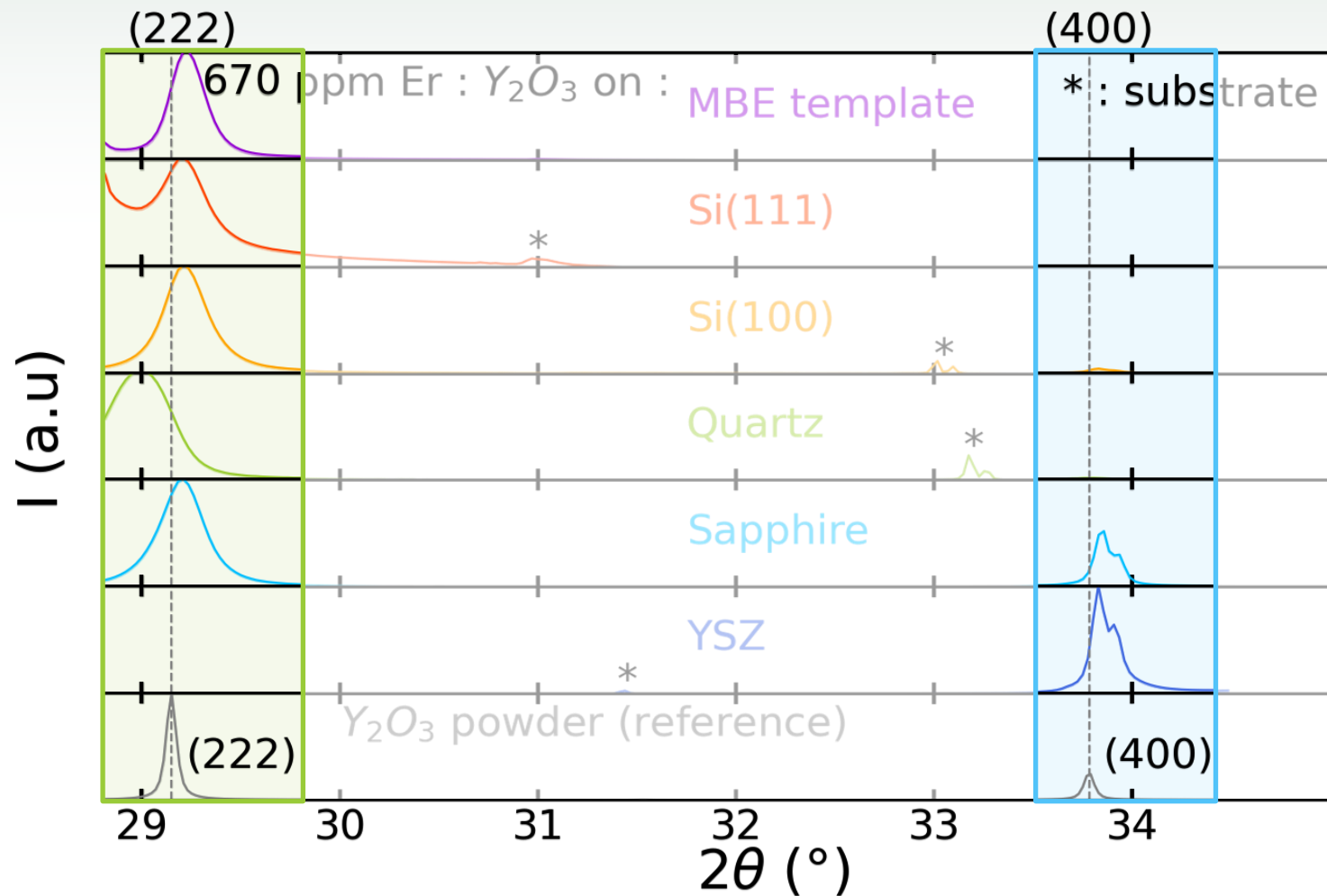
Structural properties: X-ray diffraction



- (222) and (400) peak size varies with **substrate**

Results

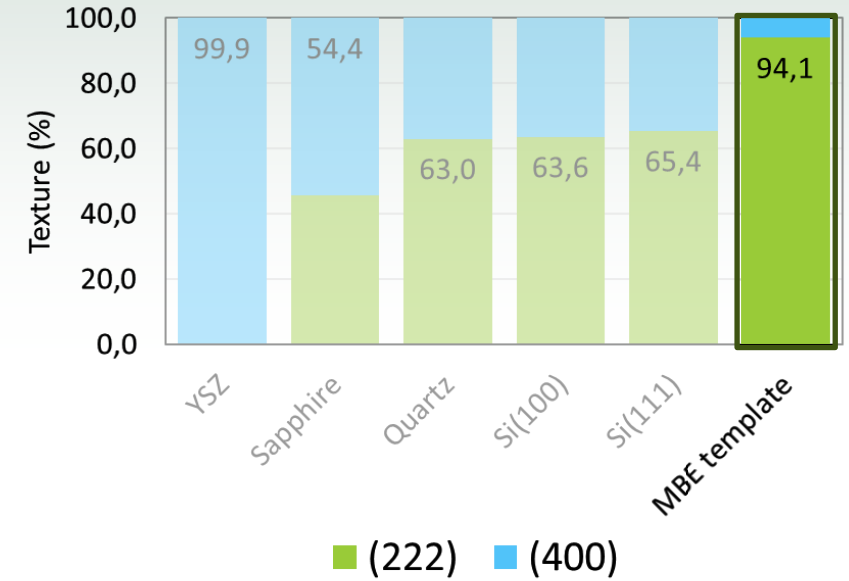
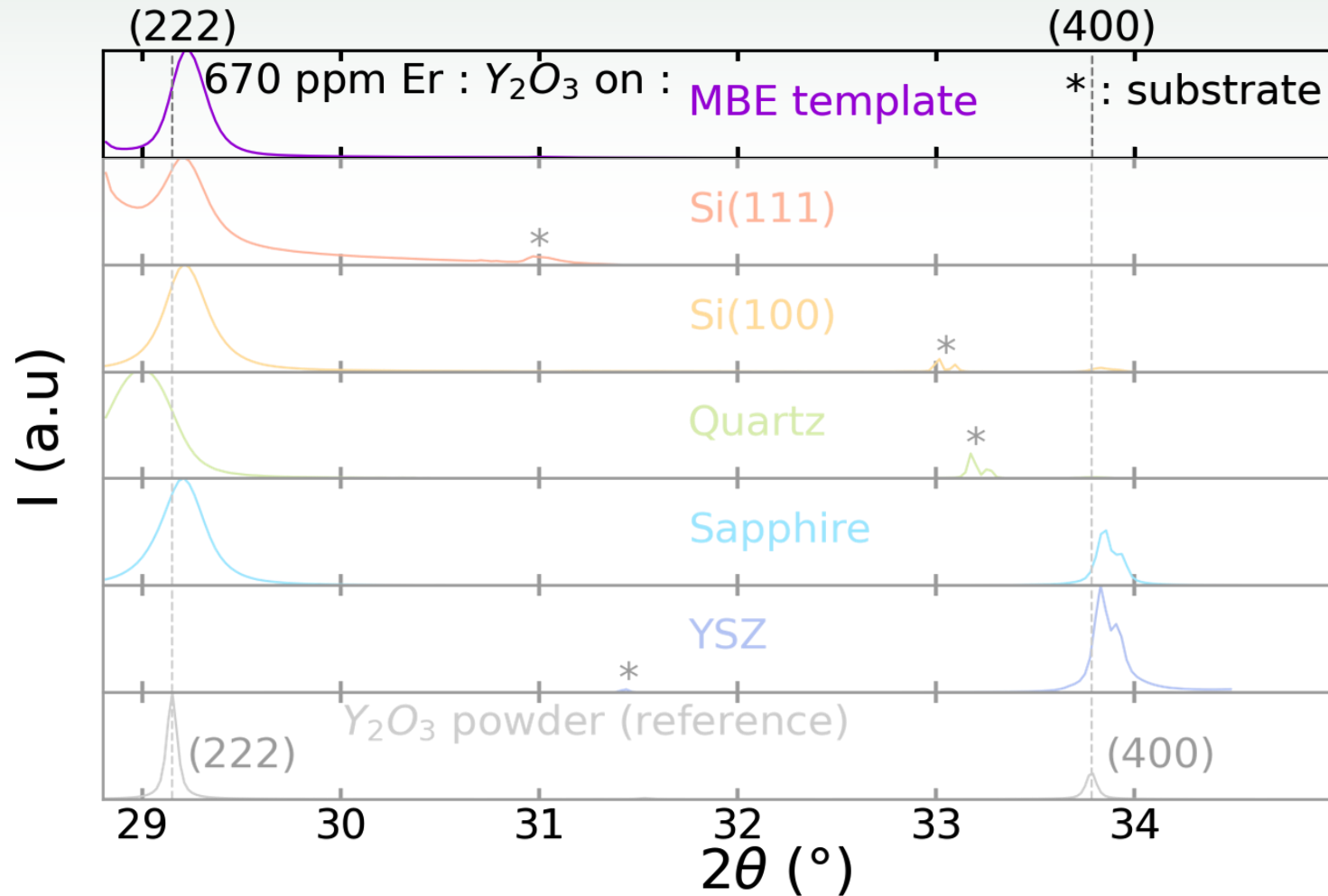
Structural properties: X-ray diffraction



$$\%_{texture} = 100 \times \left(1 - \frac{\left(\frac{I_{400}}{I_{222}}\right)}{\left(\frac{I_{400}}{I_{222}}\right)_{powder}} \right)$$

Results

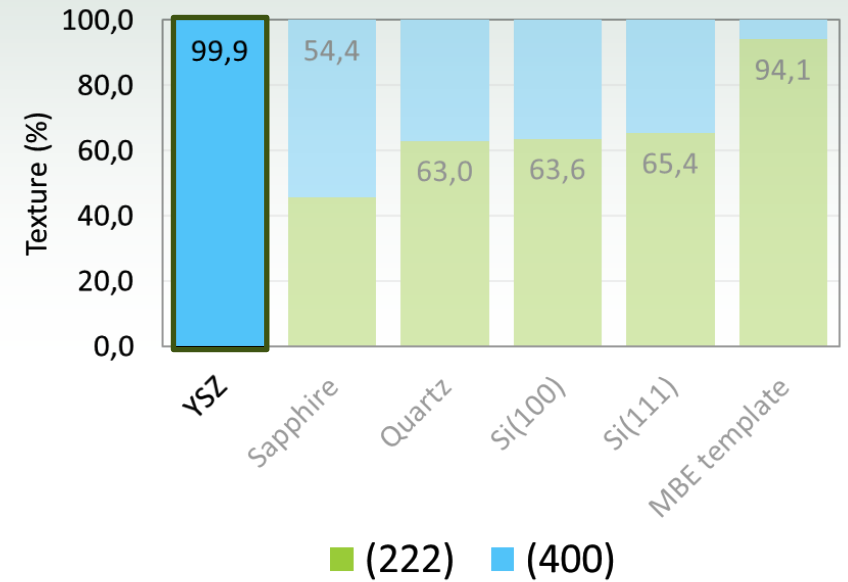
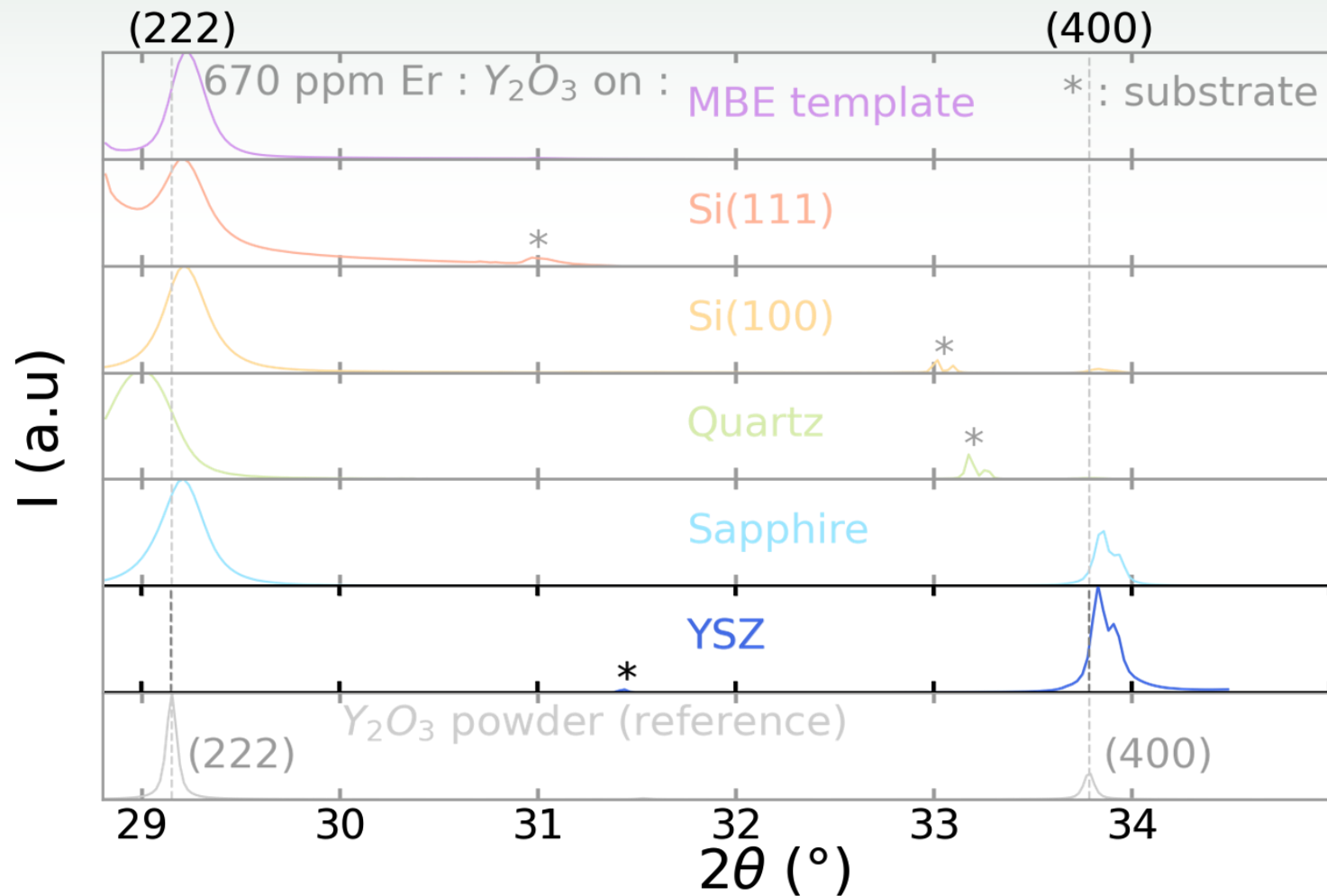
Structural properties: X-ray diffraction



- (222) and (400) peak size varies with **substrate**
- Increase of preferential orientation with **MBE template**

Results

Structural properties: X-ray diffraction



- (222) and (400) peak size varies with **substrate**
- Increase of preferential orientation with **MBE template**
- **YSZ**: orientation almost 100 % → Epitaxy ?

Optical properties

Optical properties: Photoluminescence spectra

$$\mathcal{H} = H_{IL} + H_{CC} + H_{Spin}$$

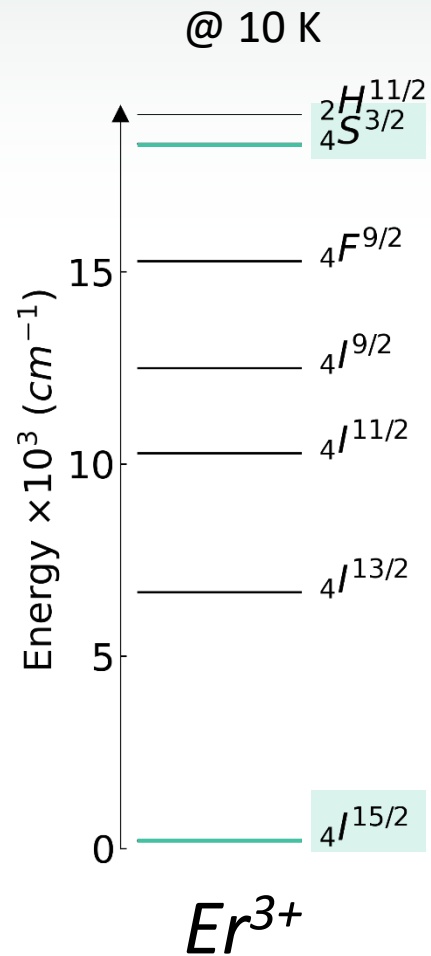
H_{IL} : Free ion Hamiltonian

H_{CC} : Crystal field Hamiltonian

H_{Spin} : Spin Hamiltonian

Results

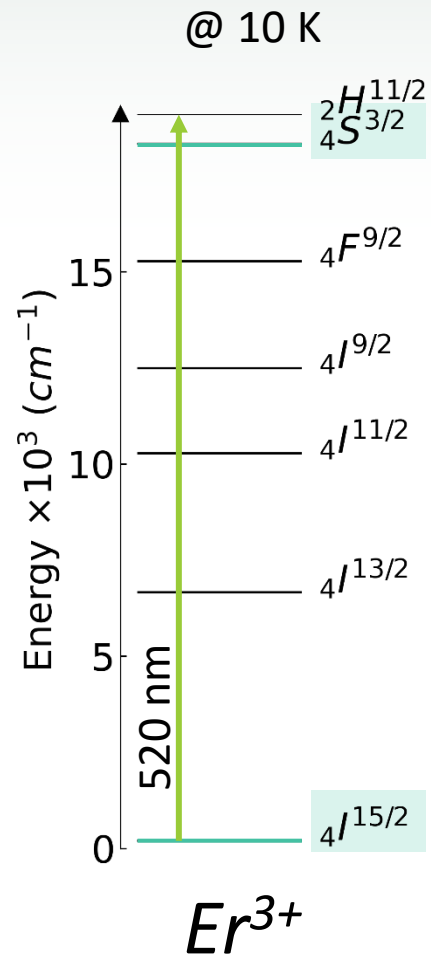
Optical properties: Photoluminescence spectra



$$\mathcal{H} = H_{IL} + H_{CC} + H_{Spin}$$

Results

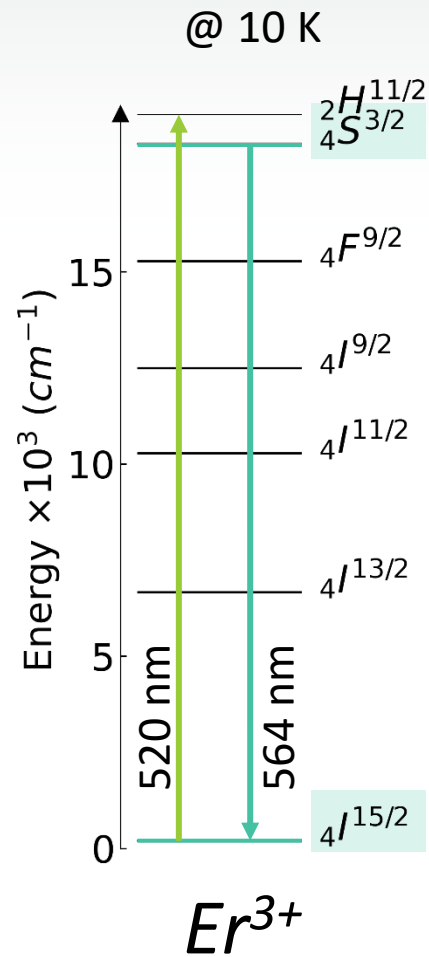
Optical properties: Photoluminescence spectra



$$\mathcal{H} = H_{IL} + H_{CC} + H_{Spin}$$

Results

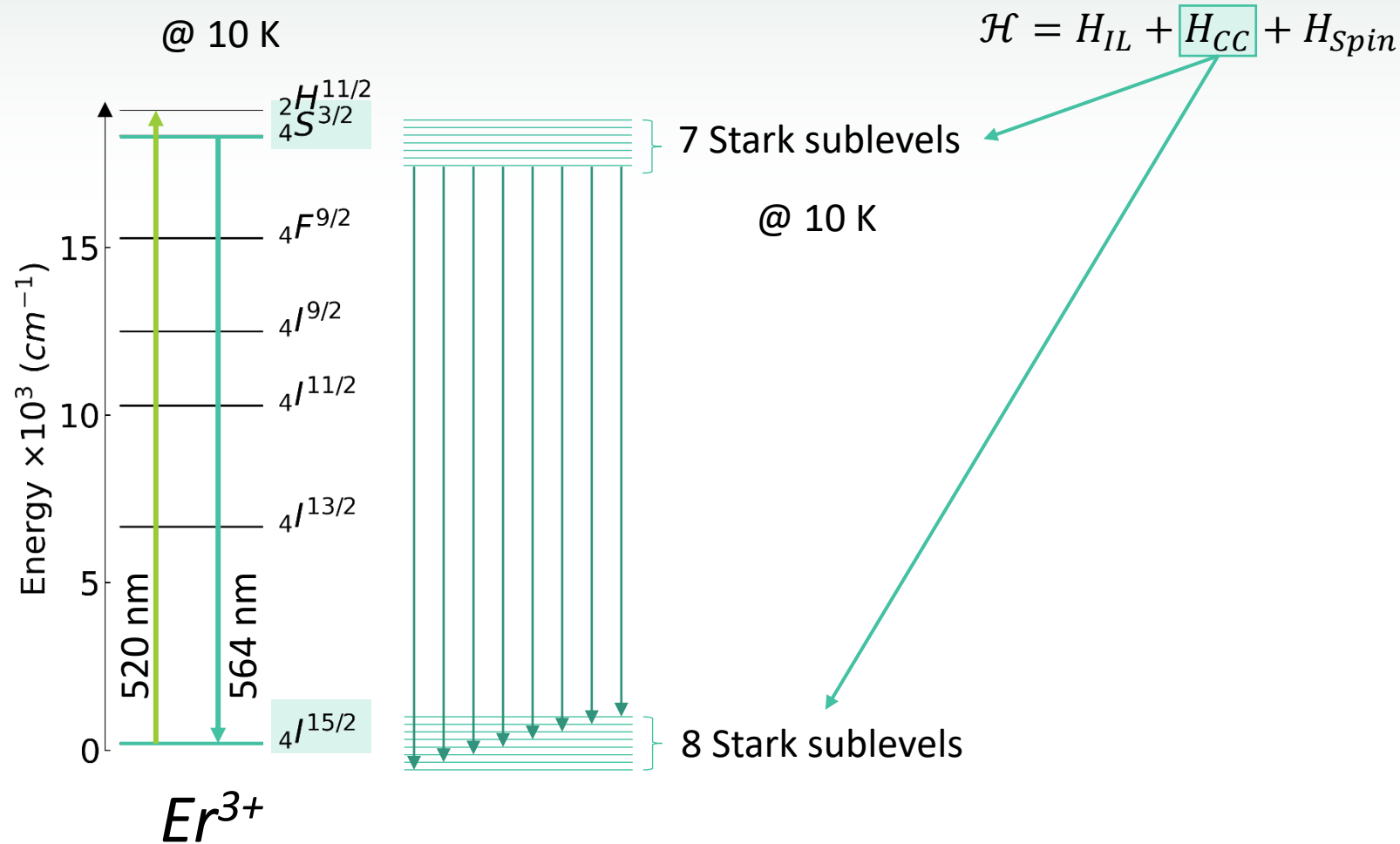
Optical properties: Photoluminescence spectra



$$\mathcal{H} = H_{IL} + H_{CC} + H_{Spin}$$

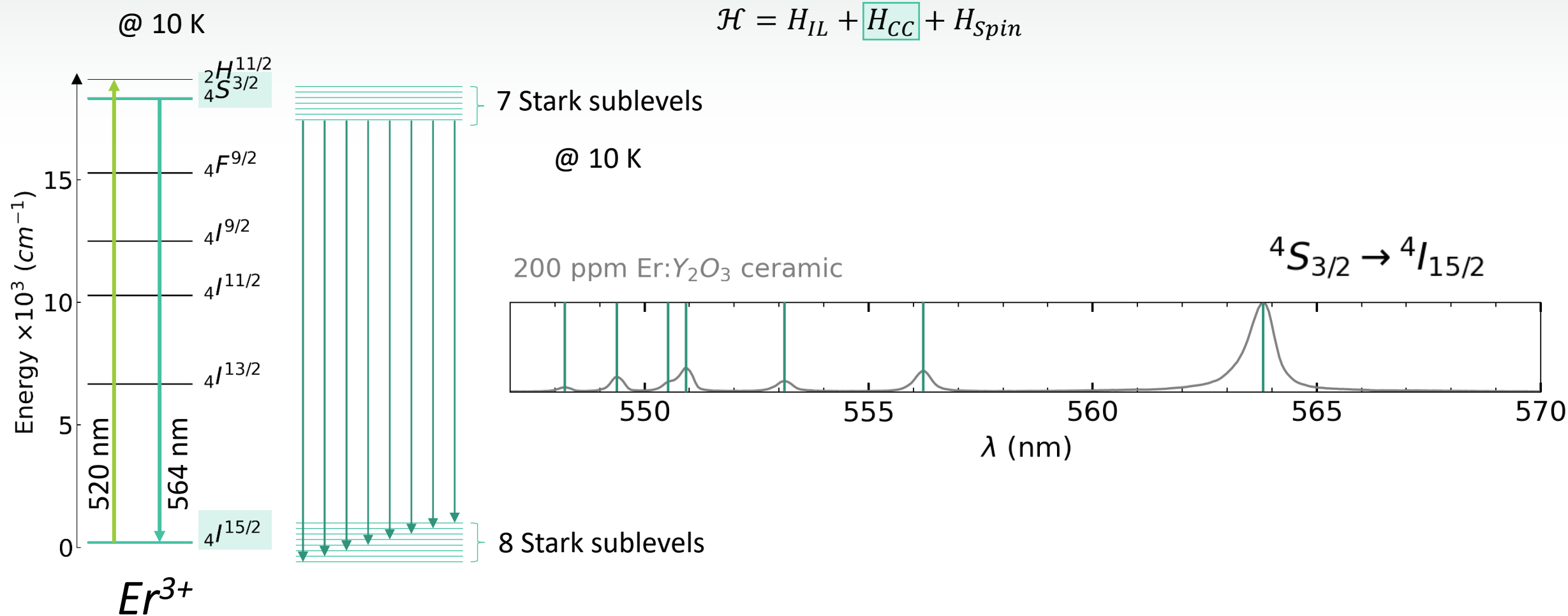
Results

Optical properties: Photoluminescence spectra



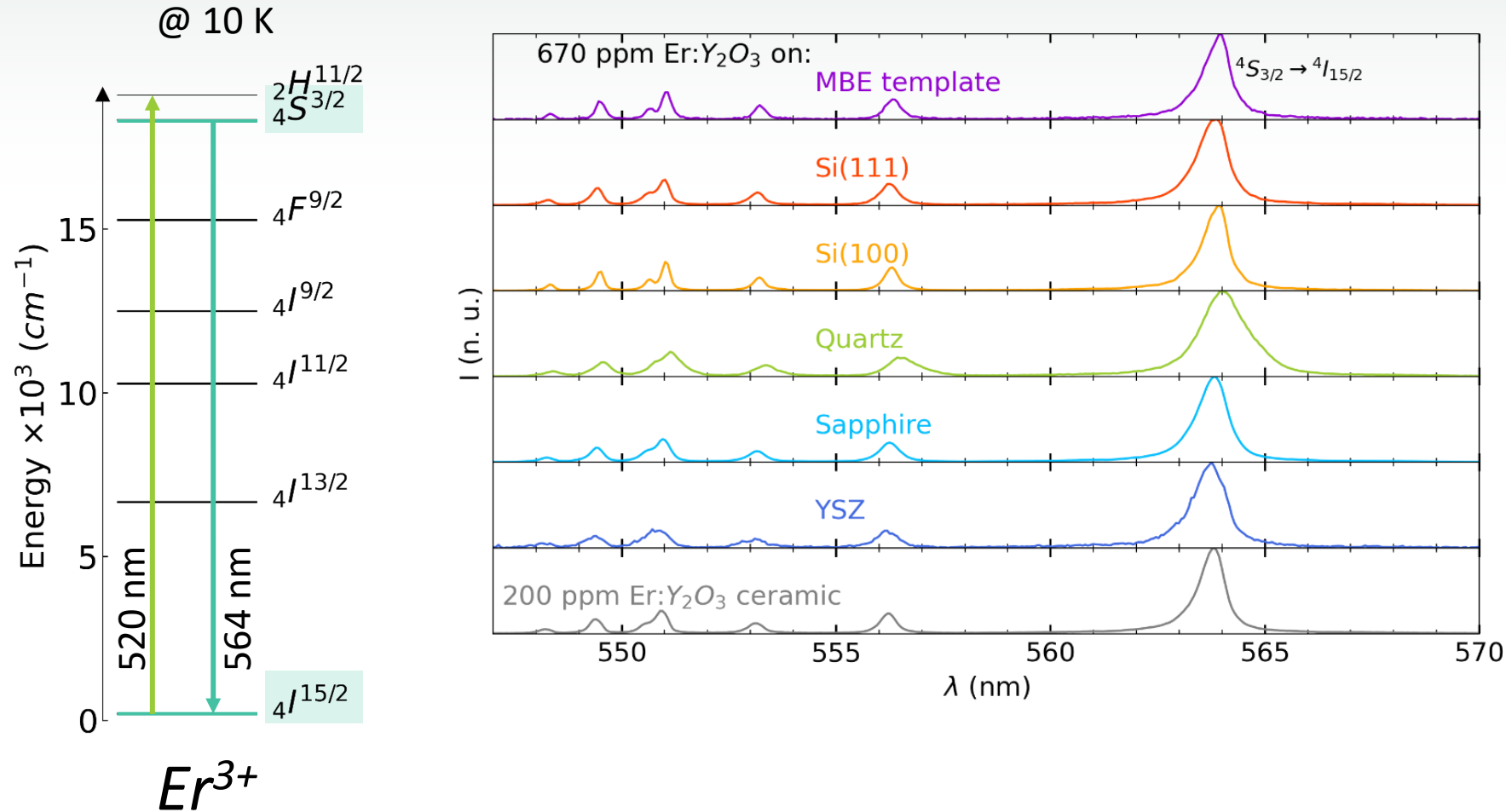
Results

Optical properties: Photoluminescence spectra



Results

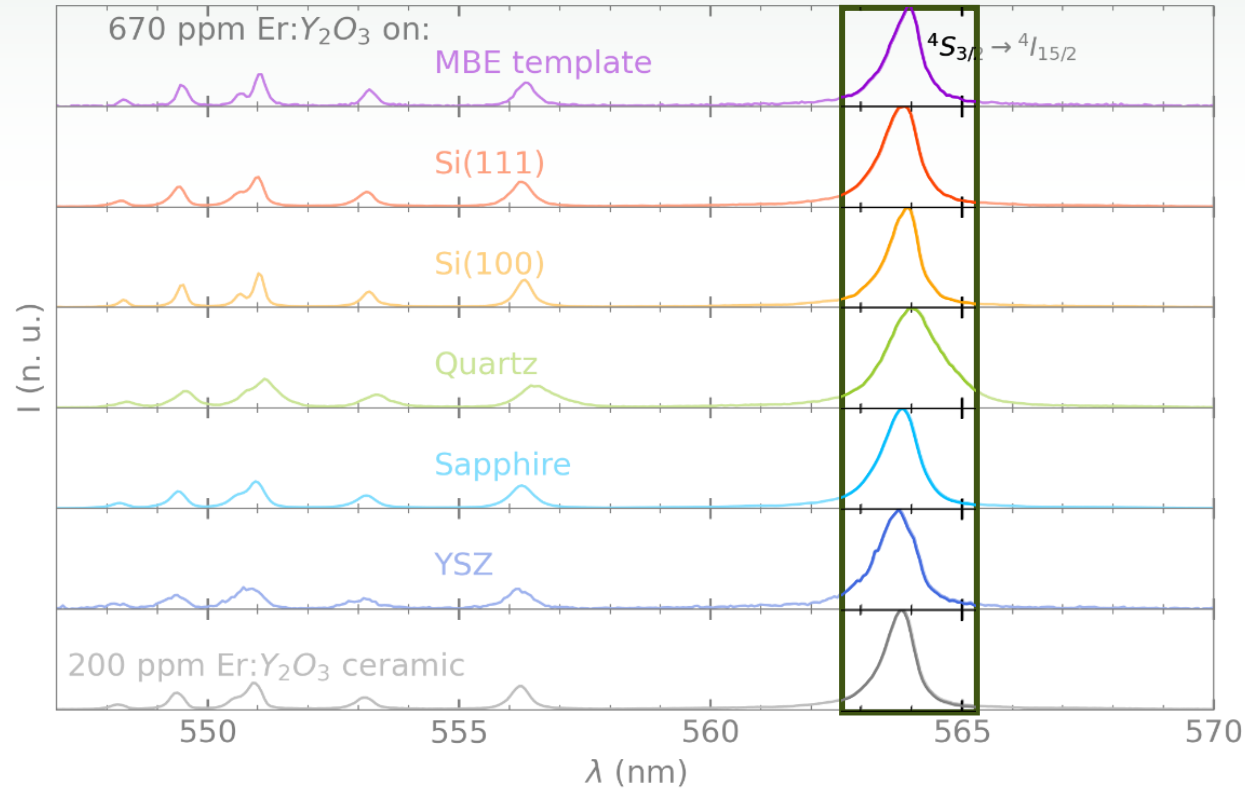
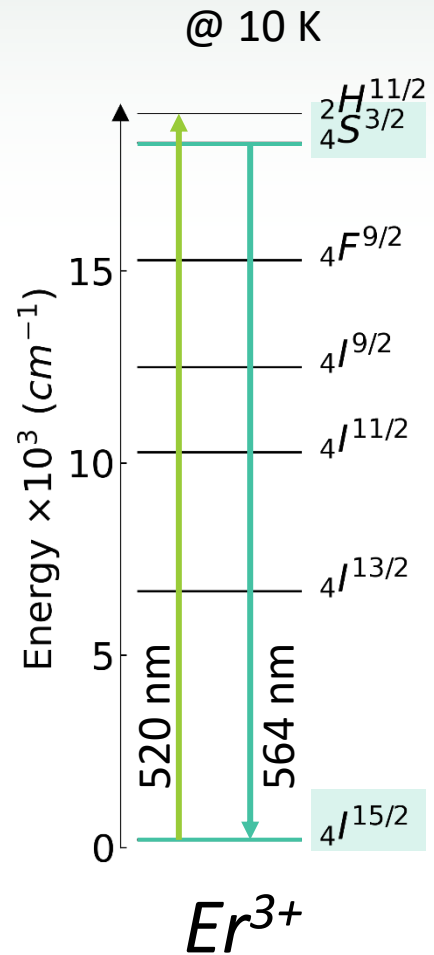
Optical properties: Photoluminescence spectra



On all the substrate, the Y_2O_3 film is crystallised

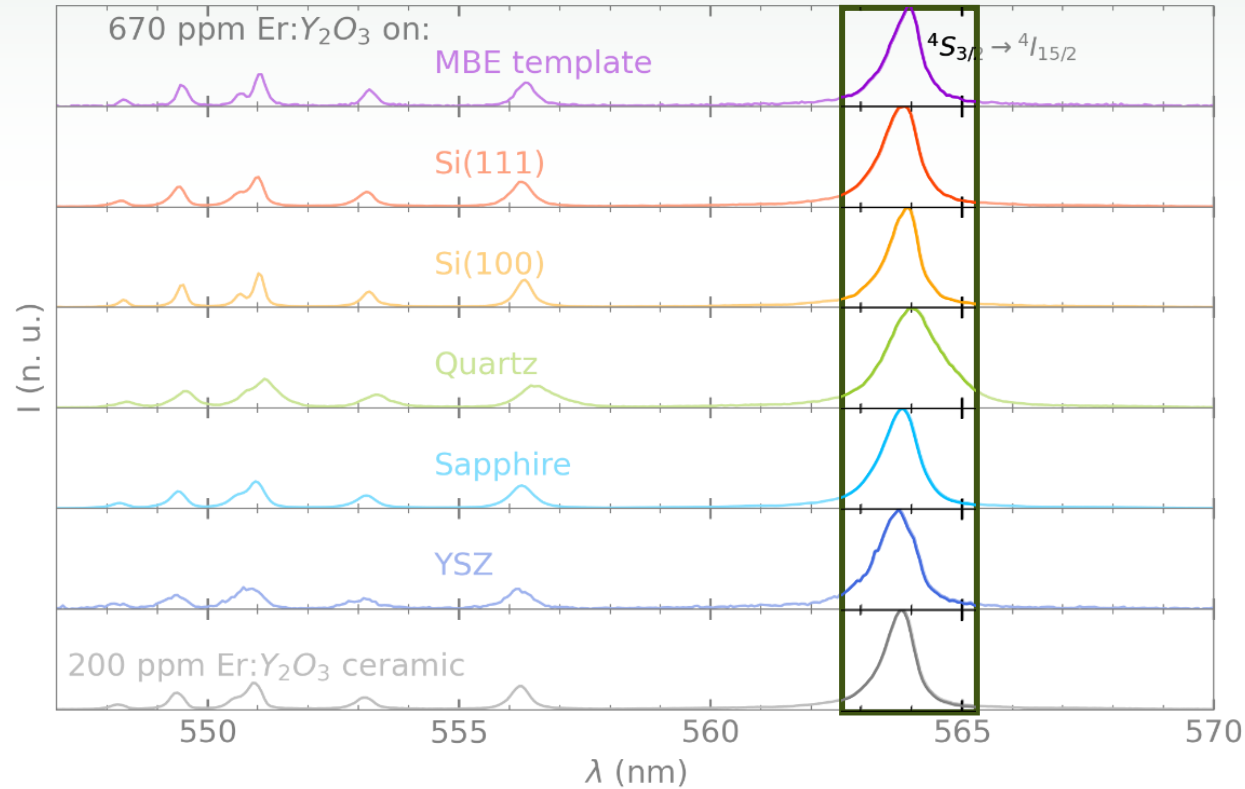
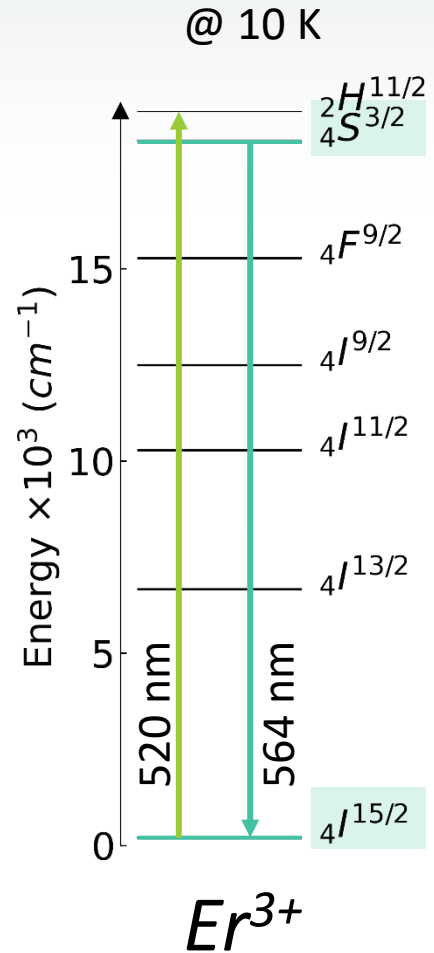
Results

Optical properties: Decays



Results

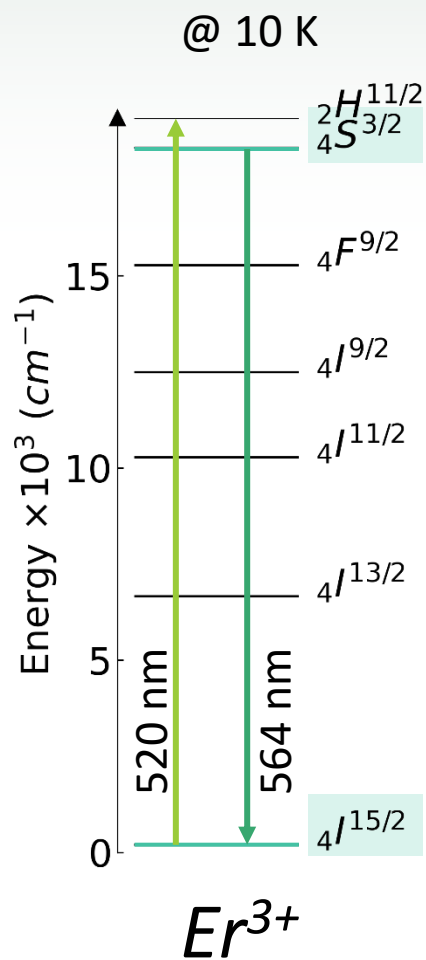
Optical properties: Decays



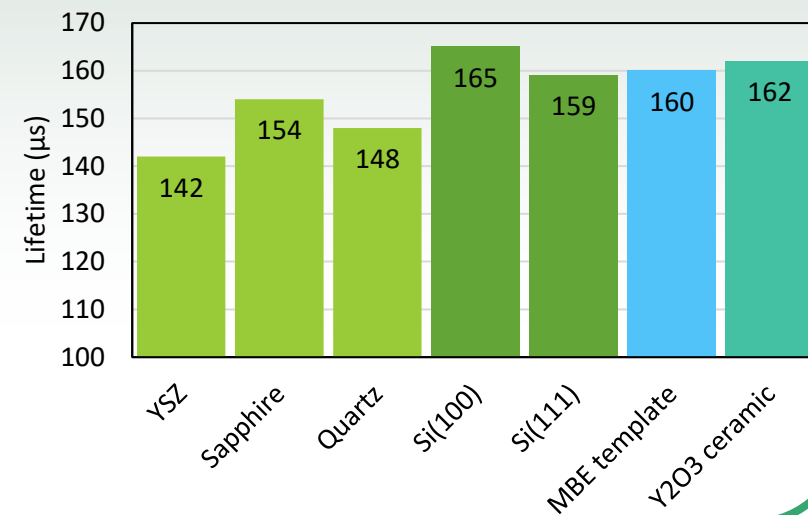
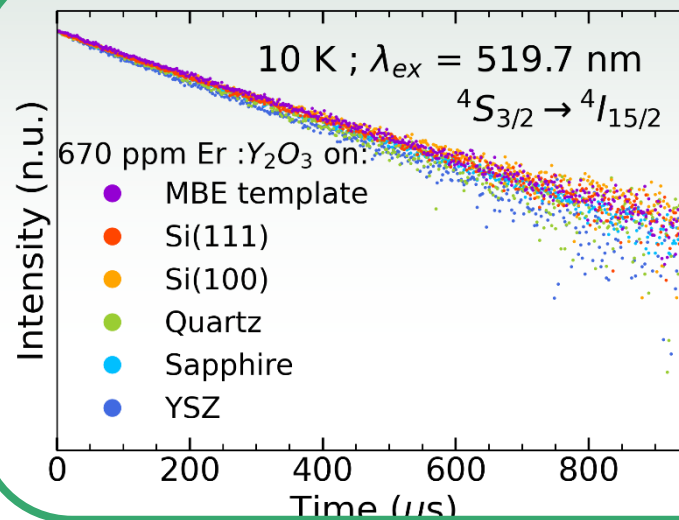
Analysis of decay **shape** and **time** gives information on RE environment

Results

Optical properties: Decays

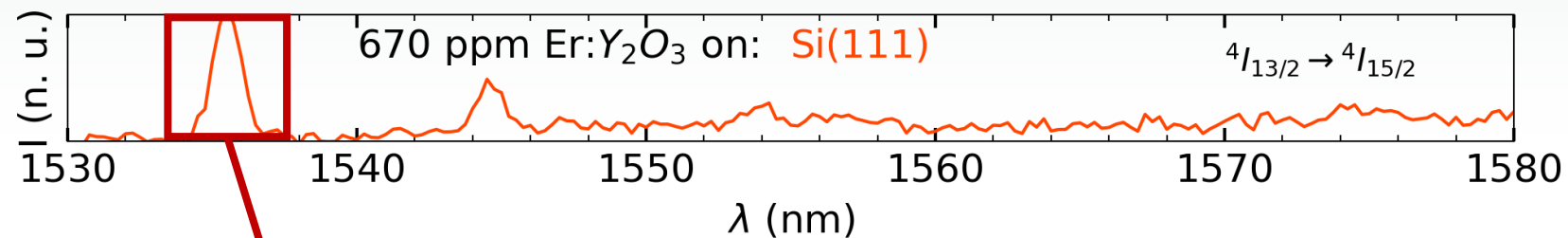
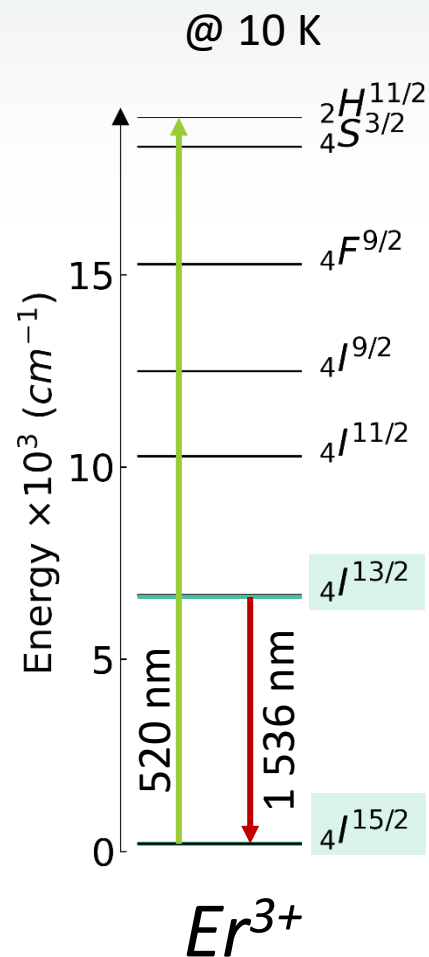


Visible

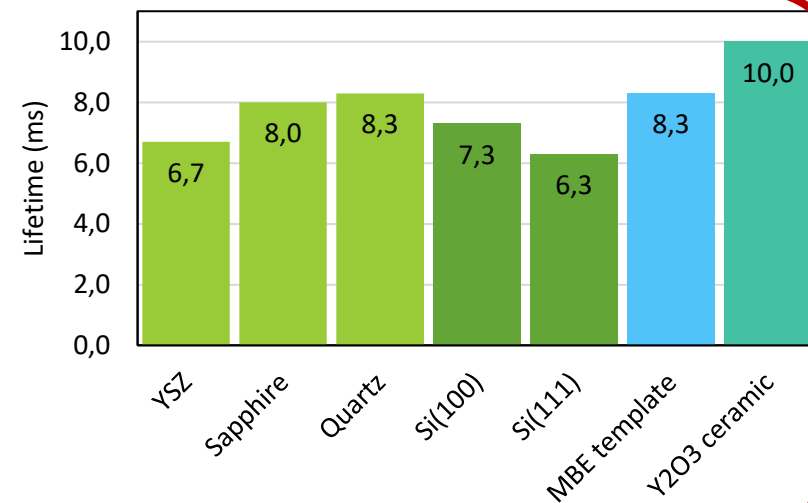
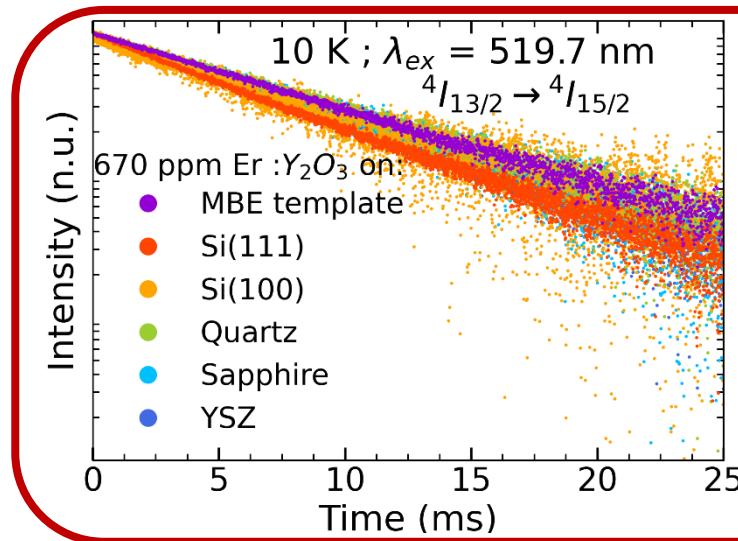


Results

Optical properties: Decays

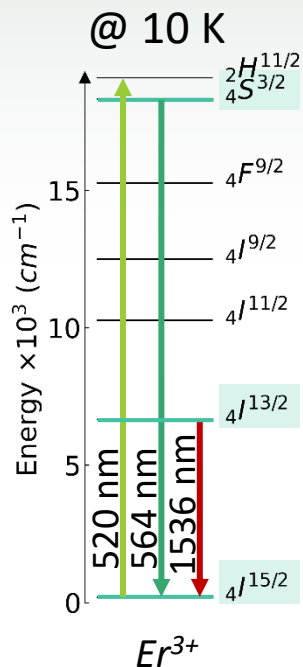


IR

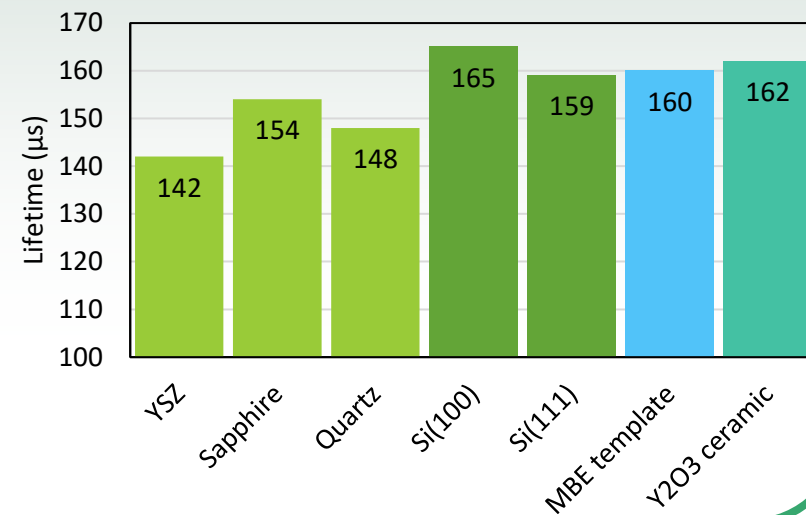
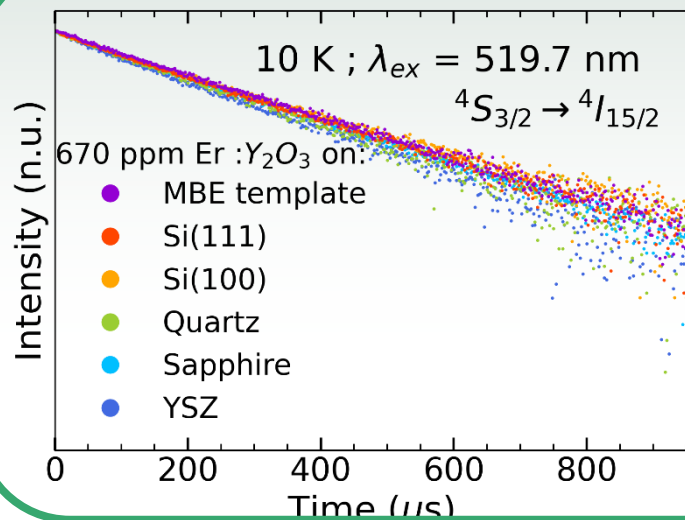


Results

Optical properties: Decays

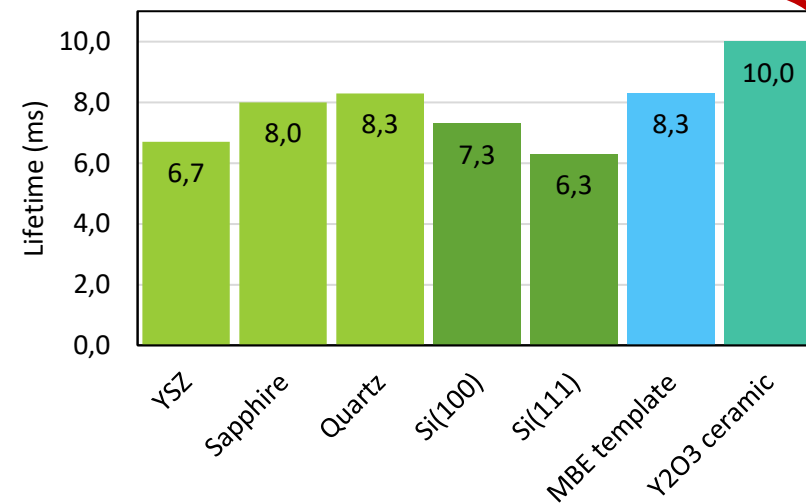
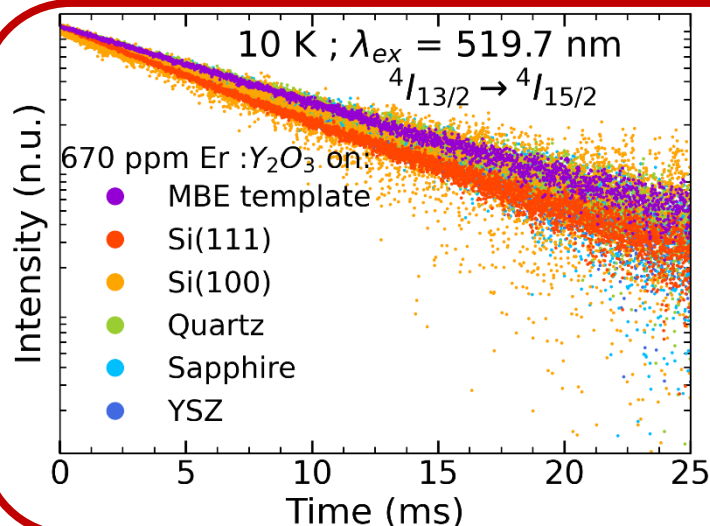


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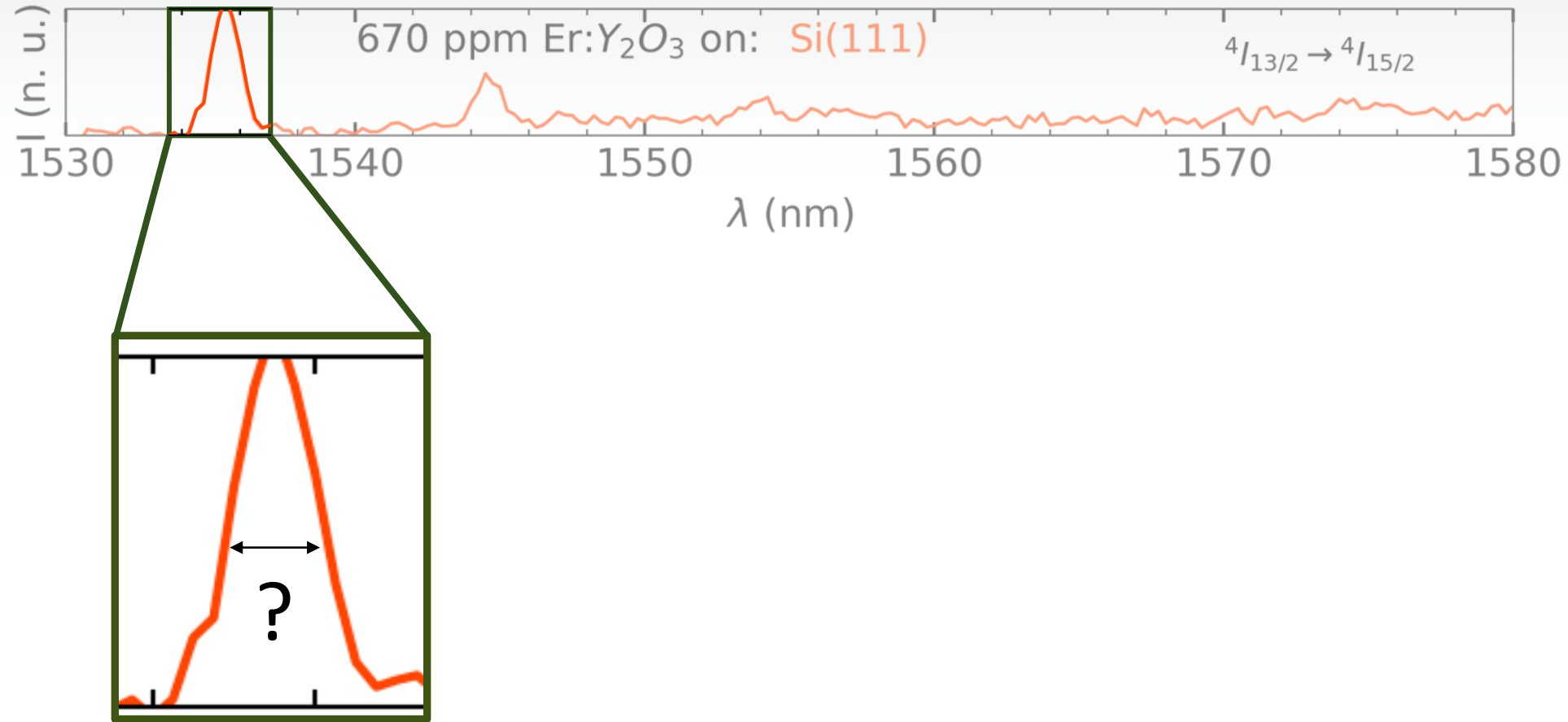
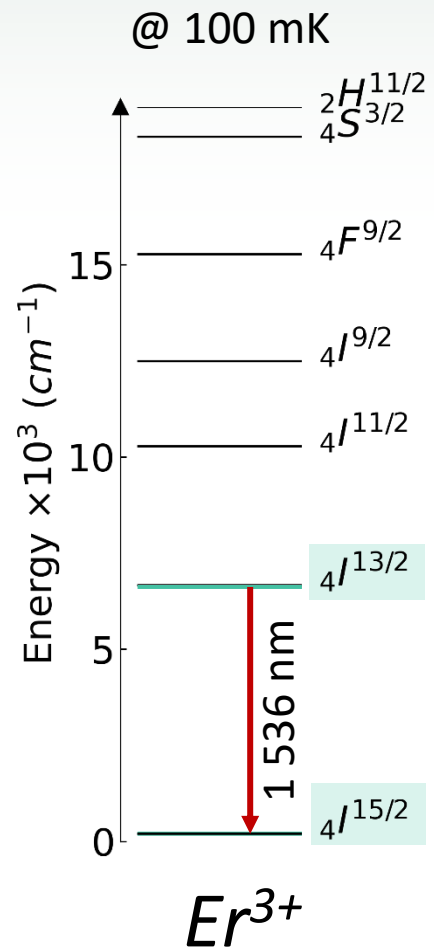
- Similar lifetimes for **all samples** and close from ceramic
- YSZ** : shorter lifetime \rightarrow smaller grains ?

IR



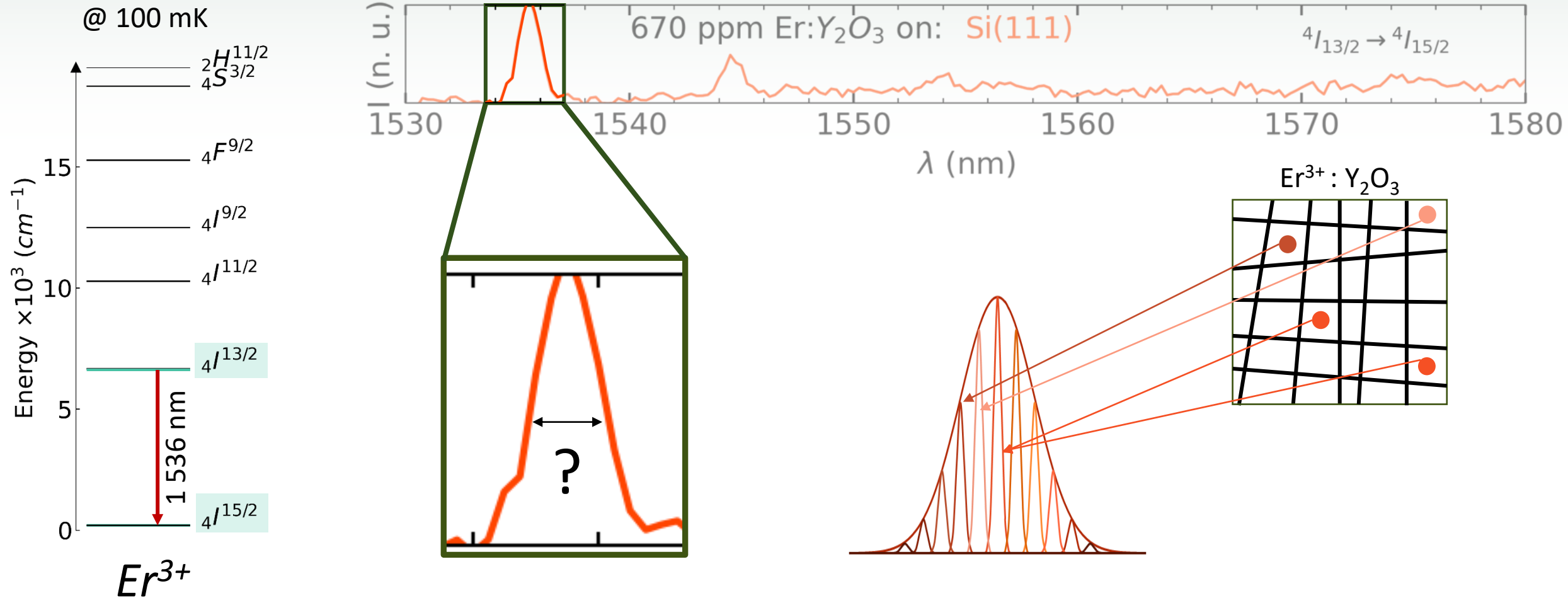
Results

Optical properties: Inhomogeneous linewidth



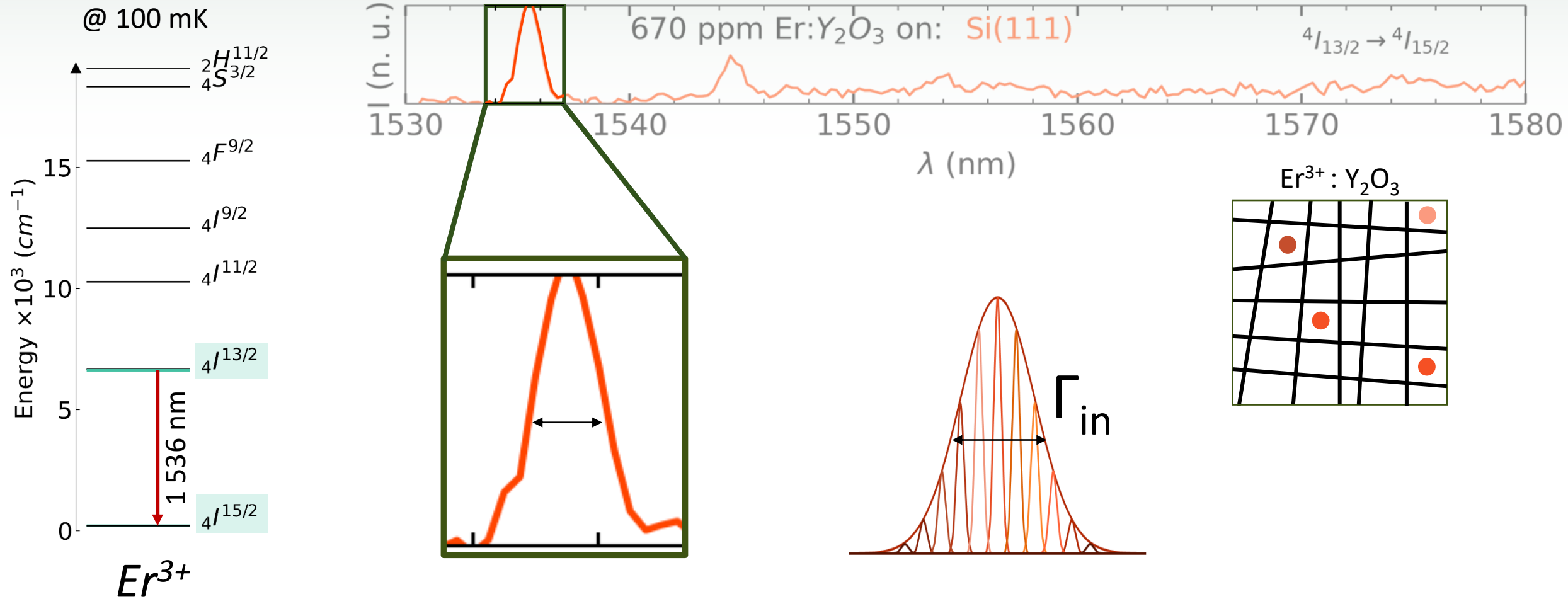
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Optical properties: Inhomogeneous linewidth



Results

Optical properties: Inhomogeneous linewidth

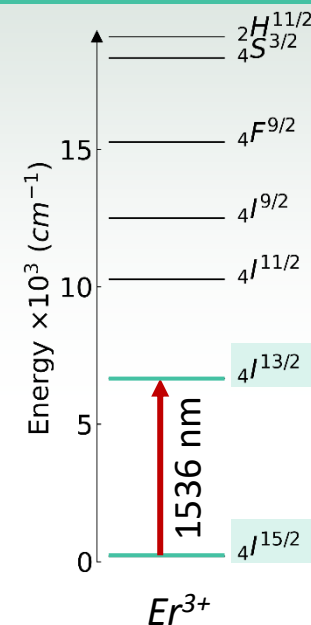
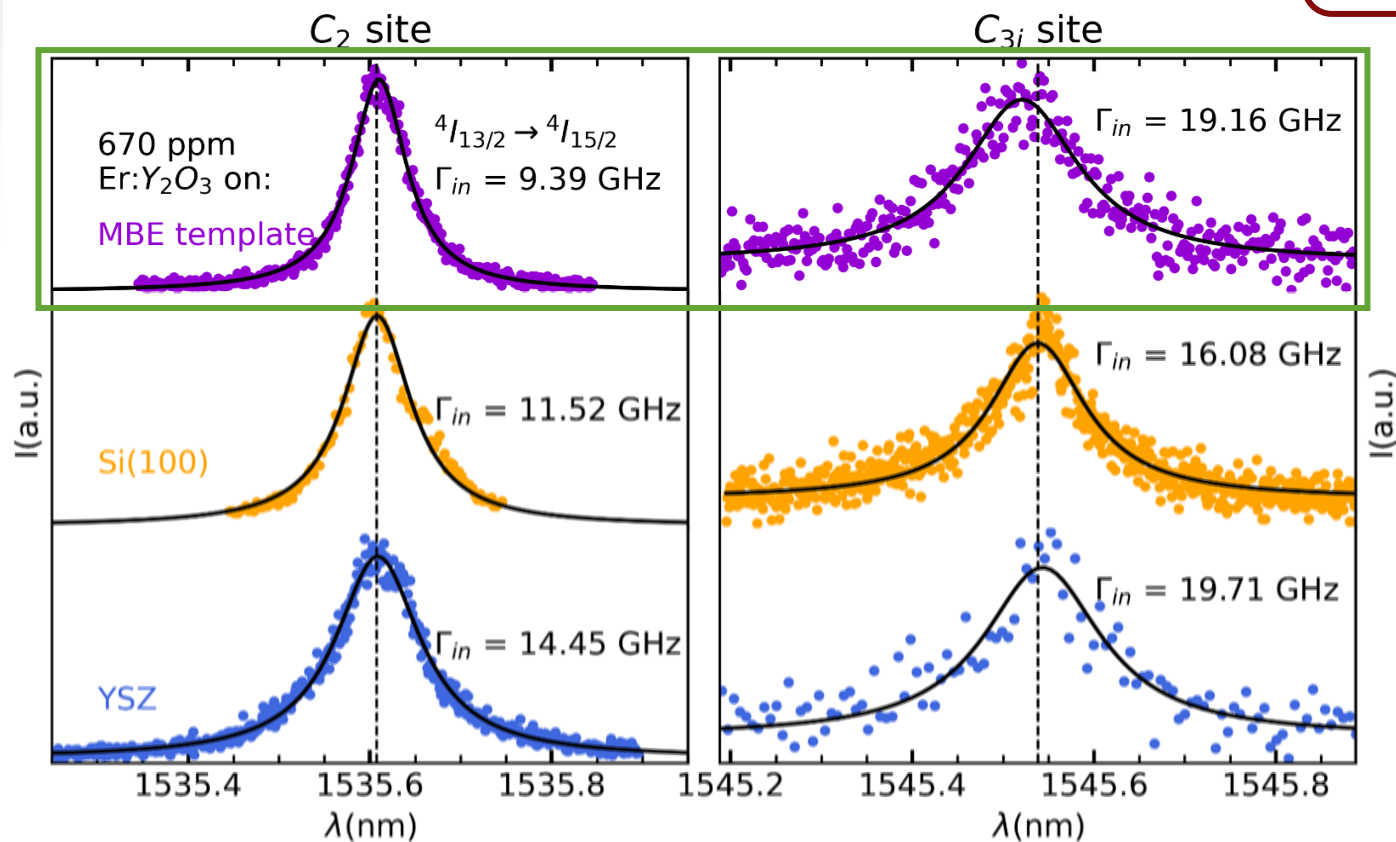


Results

Inhomogeneous broadening @100 mK



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- Larger Γ_{in} than $Er:Y_2O_3$ bulk crystals (0.4 GHz) but similar to MBE $Er:Y_2O_3$ films (5 - 24 GHz)
- Narrowest Γ_{in} for **MBE template**

R. Fukumori et al., *Phys. Rev. B*, **101**, 214202 (2020)

M. K. Singh et al., *APL Mater.*, **8**, 031111 (2020)

Conclusion and outlook

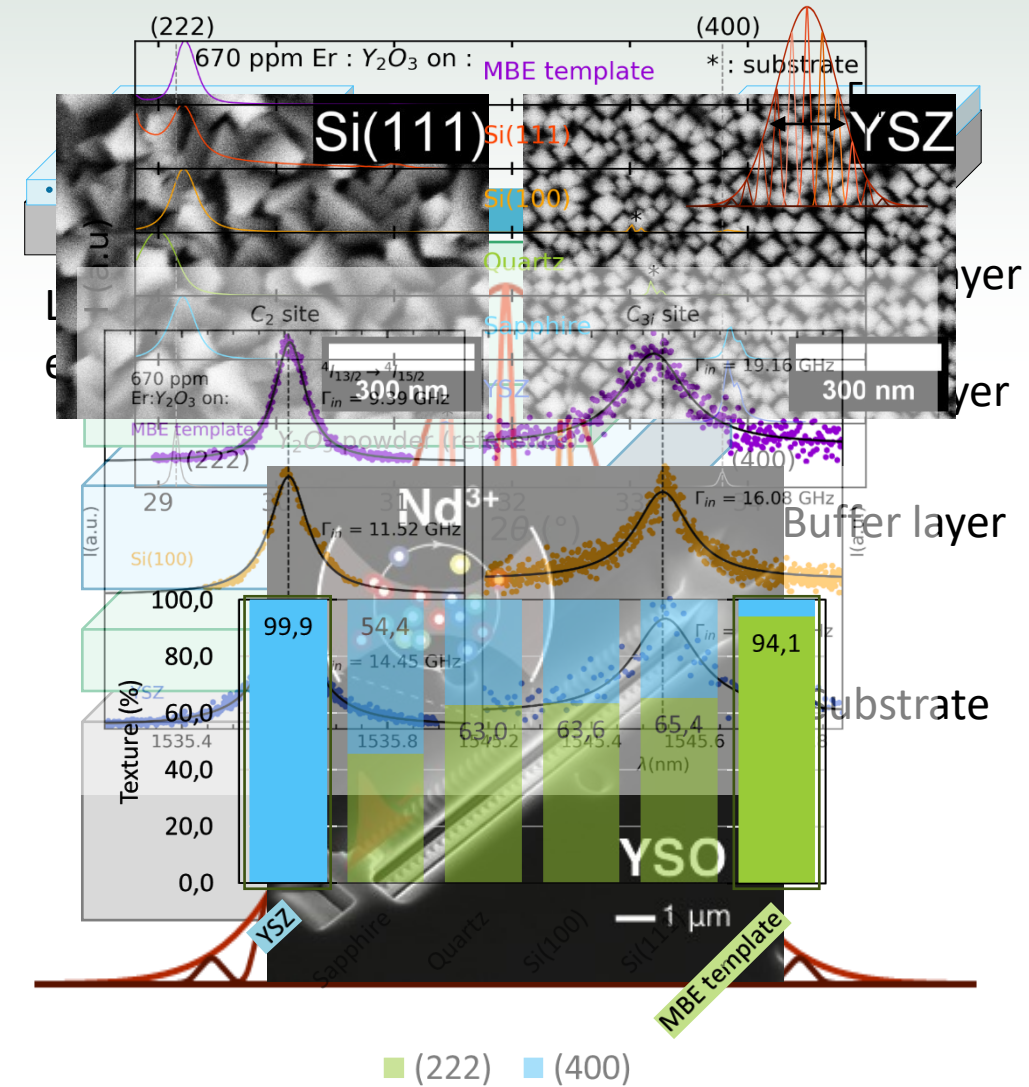
Conclusion and outlook

Conclusions

- Films well crystalized were grown on **all substrates**
- **YSZ** and **MBE template** especially seem promising regarding structural and morphological properties
- Film on the **MBE template** gives a slightly **narrower Γ_{in}**

Outlooks

- Measurements of the Γ_h of the most promising samples
- Develop strategies to improve crystalline quality
- **Nanostructuring** of the films to integrate them into resonators or waveguides



Thanks for your attention !