

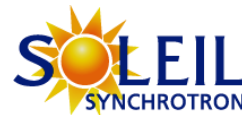
# Transient Liquid Assisted Growth (TLAG)

A method to increase CC throughput and meet applications requirements

**Teresa Puig**, L. Saltarelli, D. Garcia, K. Gupta, S. Rasi, R. Vlad, A. Kethamkuzhi, E. Pach, J. Banchewski, C. Pop, P. Gallego, A. Queralto, J. Gutierrez, X. Obradors  
*Institut de Ciència de Materials de Barcelona, ICMAB-CSIC, Spain*

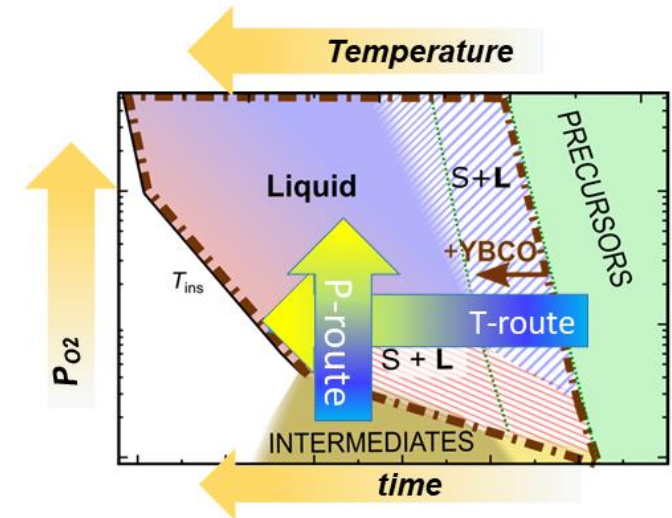
C. Mocuta

*Diffabs beamline, Soleil Synchrotron, Paris, France*



E. Solano

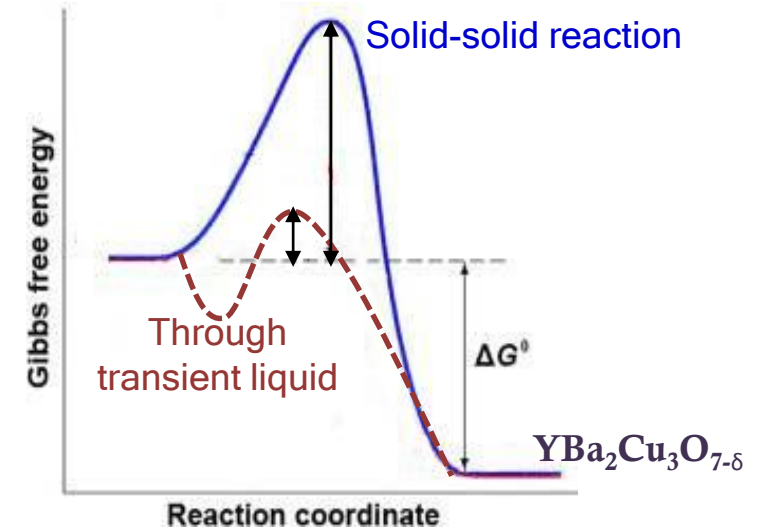
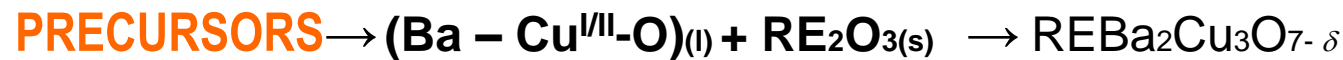
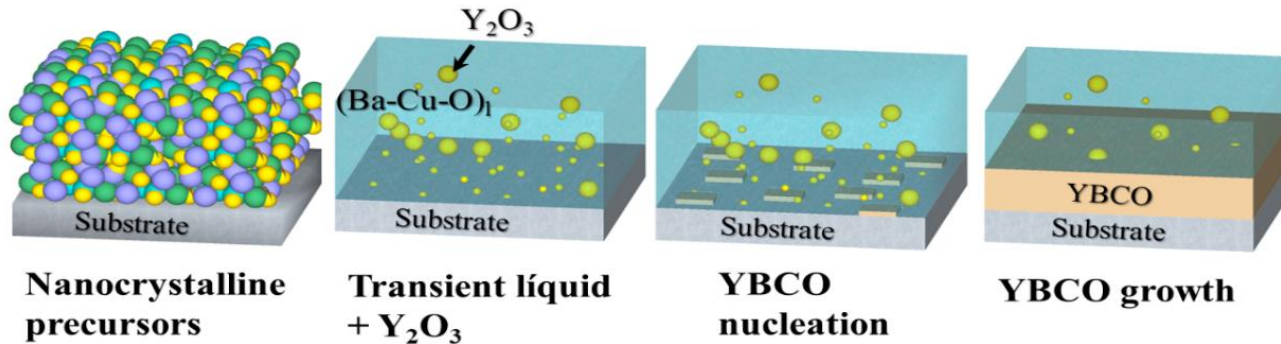
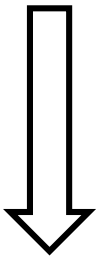
*NCD-Sweet beamline, ALBA Synchrotron, Barcelona, Spain*



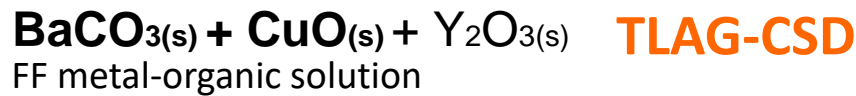
# TRANSIENT LIQUID ASSISTED GROWTH: TLAG

A high throughput non-equilibrium kinetically controlled growth process

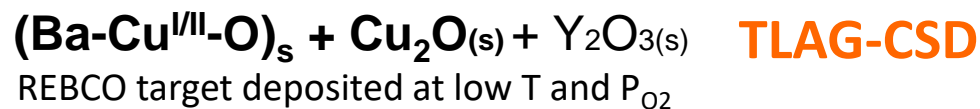
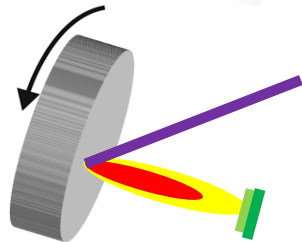
DEPOSITION METHOD



*L. Soler et al., Nat Comm (2020)*  
*S. Rasi, et al, Advance Science (2022)*



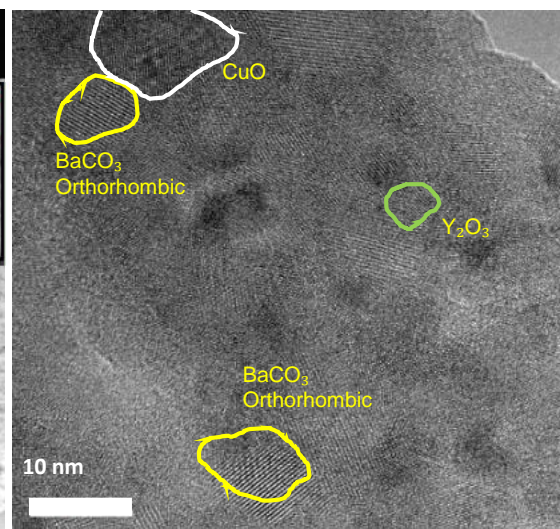
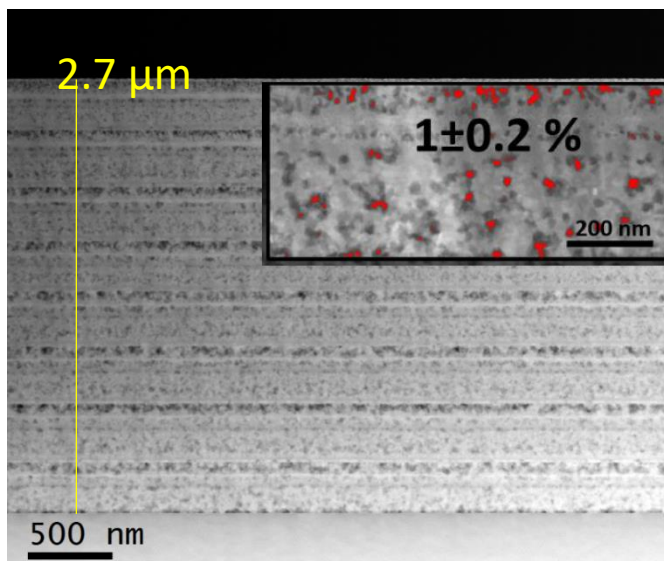
L. Saltarelli et al, ACS Appl. Mat. & Interf. (2022)



A. Quetalto et al, SUST (2023)

- Fast atomic diffusion
- Ultrafast growth rate, G, up to 2000 nm/s demonstrated
- Large area deposition
- Simple reactor
- High throughput
- Low cost/performance ratio

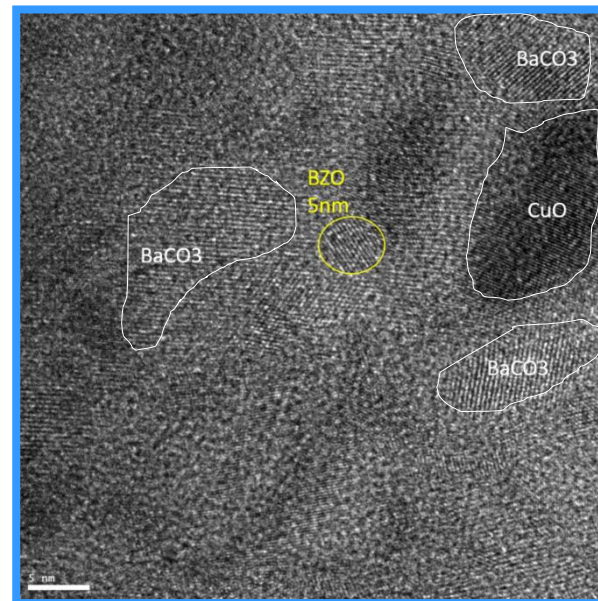
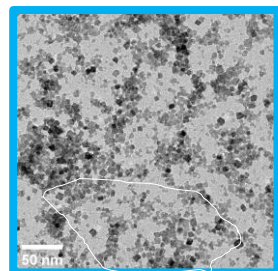
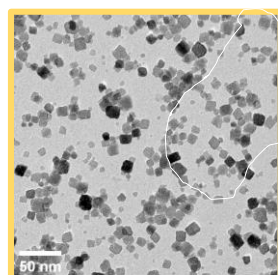
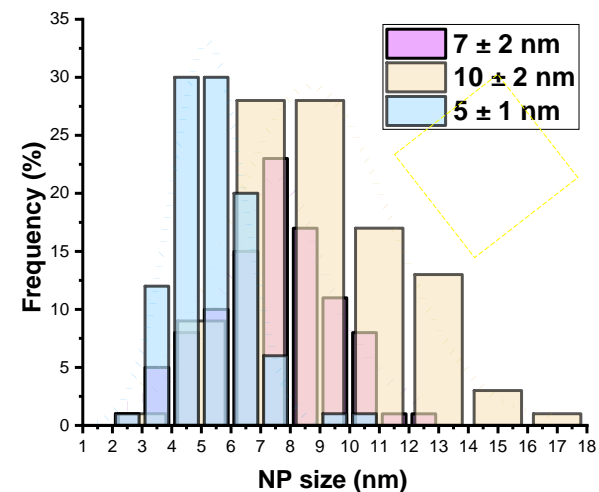
# CSD INKS AND PYROLYZED MULTIDEPOSITED FILMS



## Multifunctional ink formulation

- Uniform pyrolyzed layers
- Low porosity
- Nanocrystalline and homogeneous
- Thickness beyond 3 μm tested
- BaCO<sub>3</sub> eliminated in TLAG

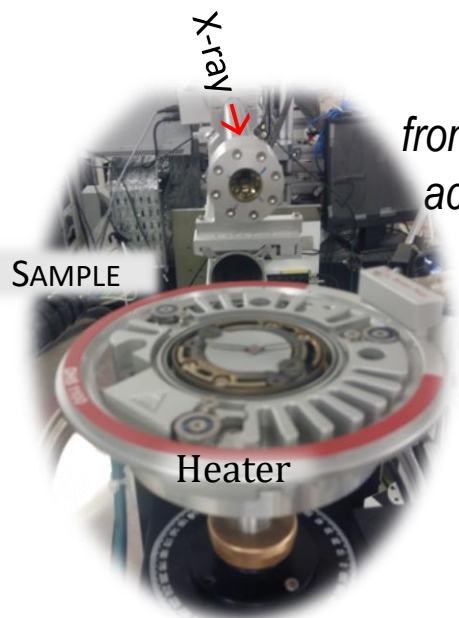
Patent EP22382741



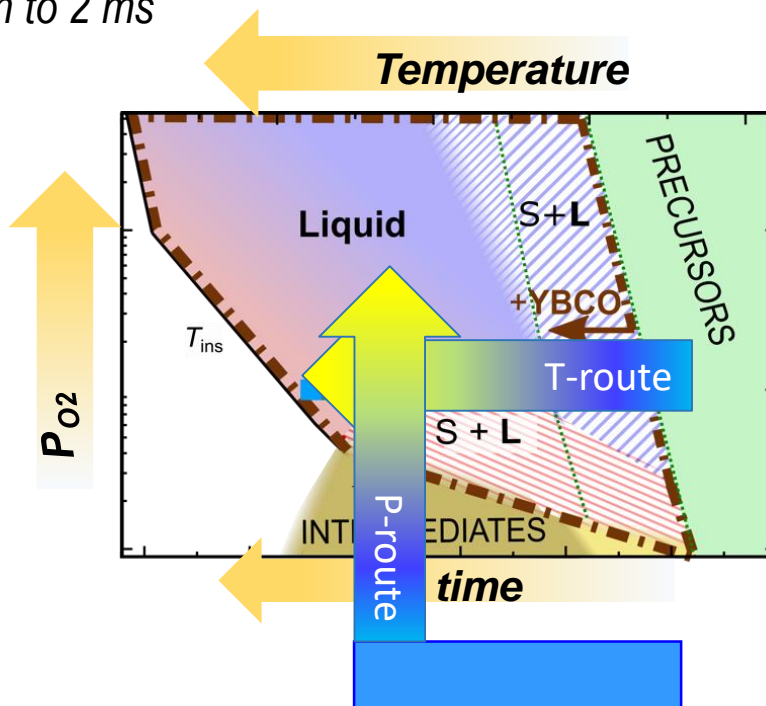
## Multifunctional colloidal ink

- Hybrid Hydrolytic-Solvothermal Synthetic Process (H<sub>2</sub>S<sub>2</sub>)
- BaZrO<sub>3</sub>, BaHfO<sub>3</sub> Np
- Similar performance
- Crystalline BZO NPs retain their shape and small size in the pyrolysis

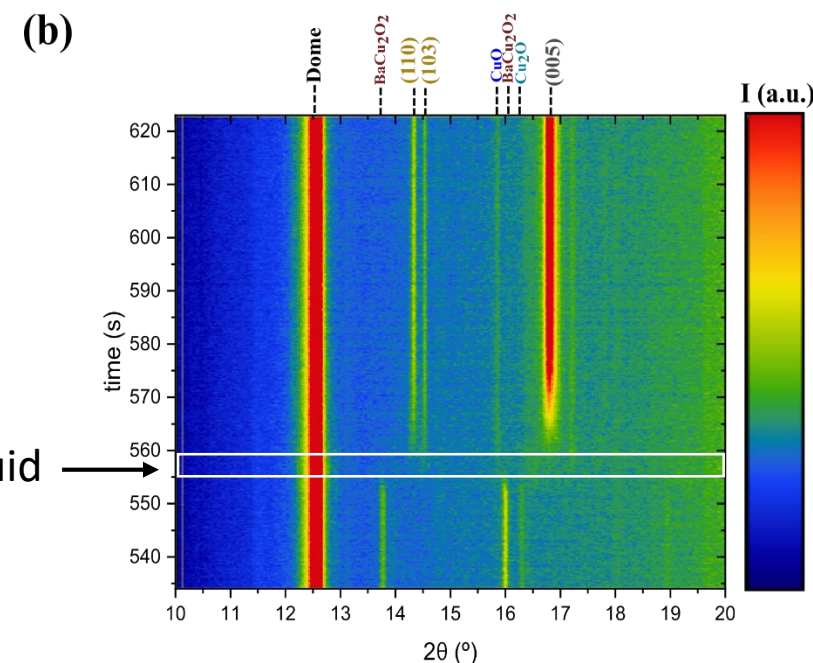
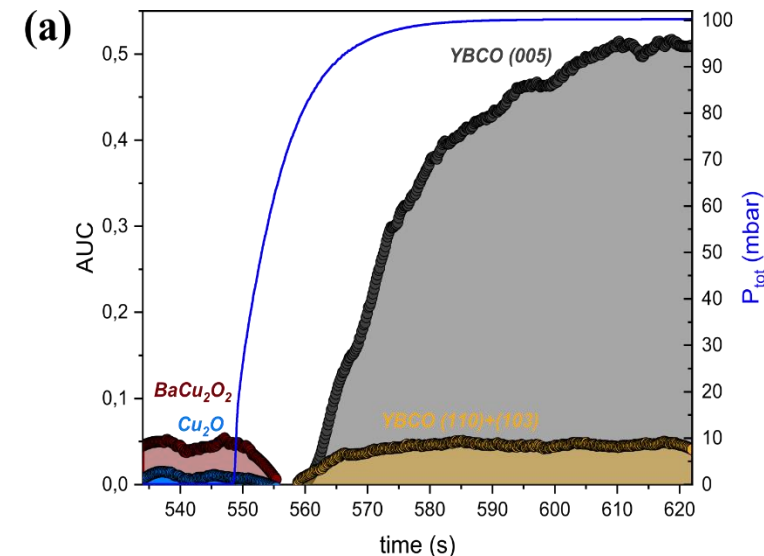
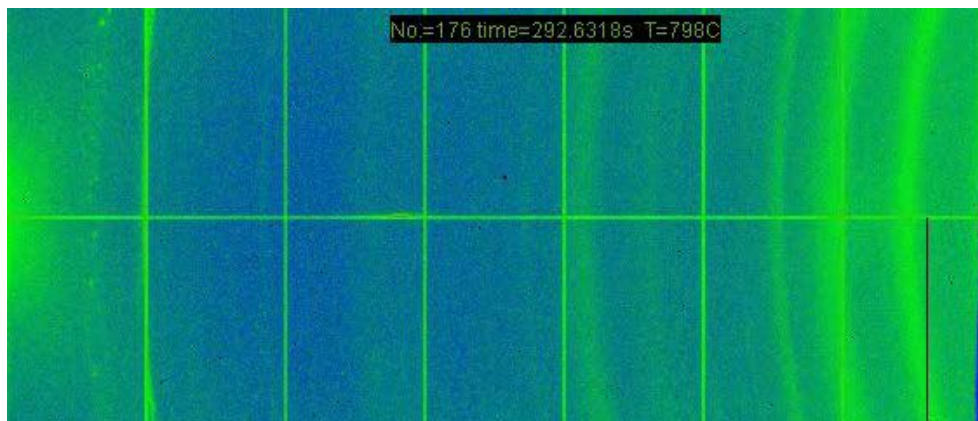
# TLAG in-situ growth evaluation



from 100 ms down to 2 ms acquisition time

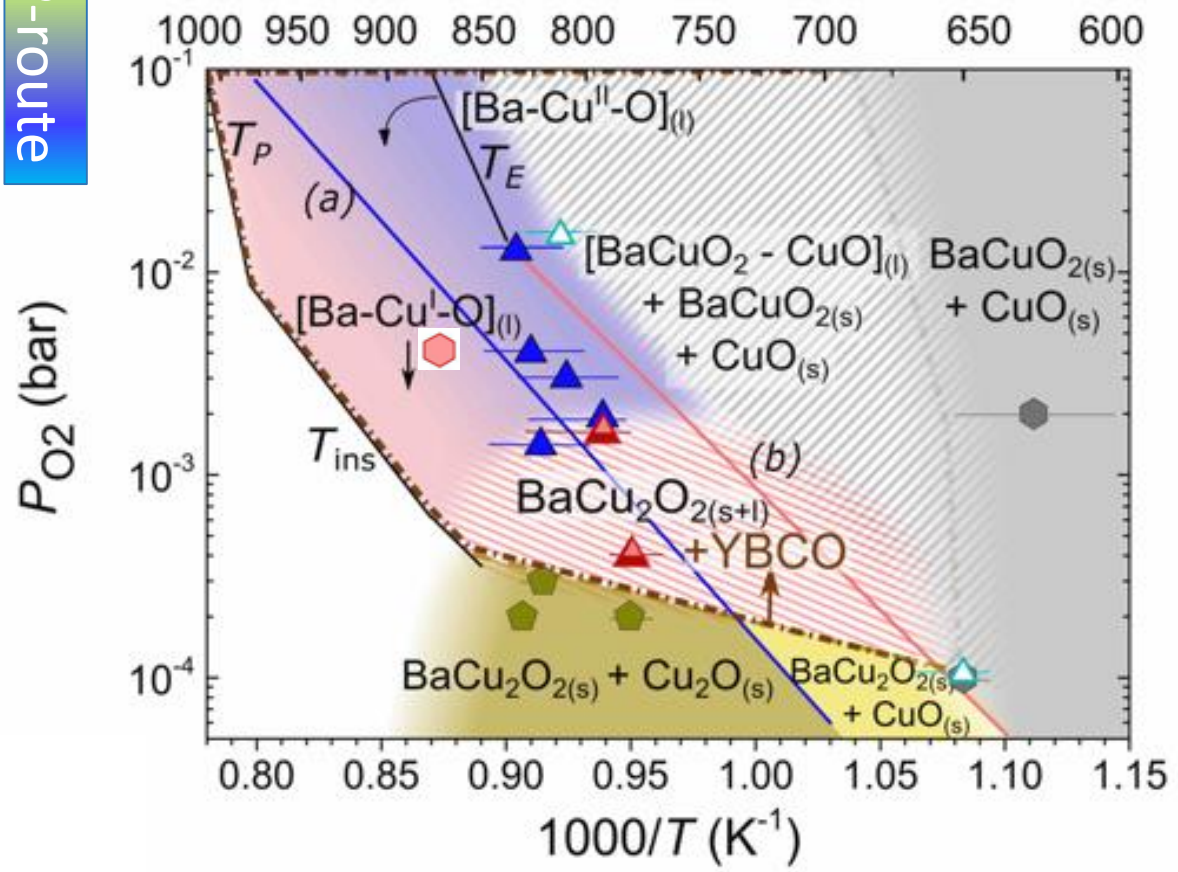


In-situ growth XRD synchrotron experiments

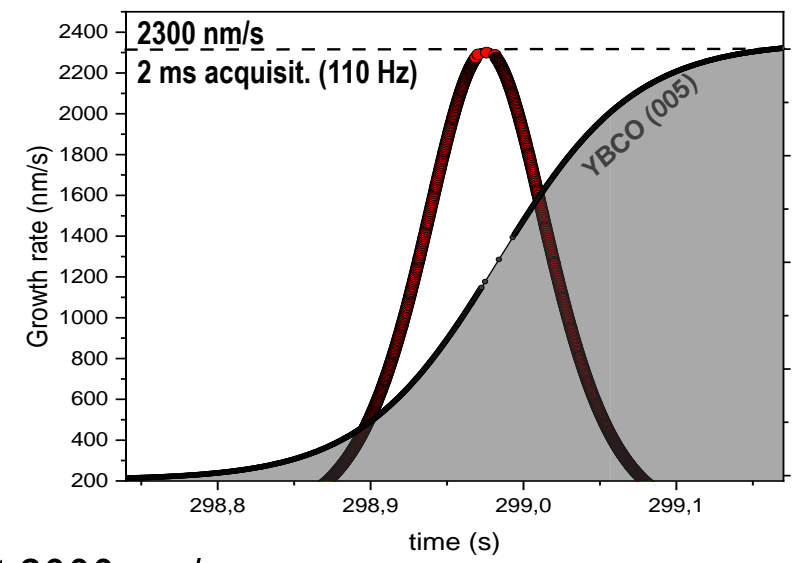
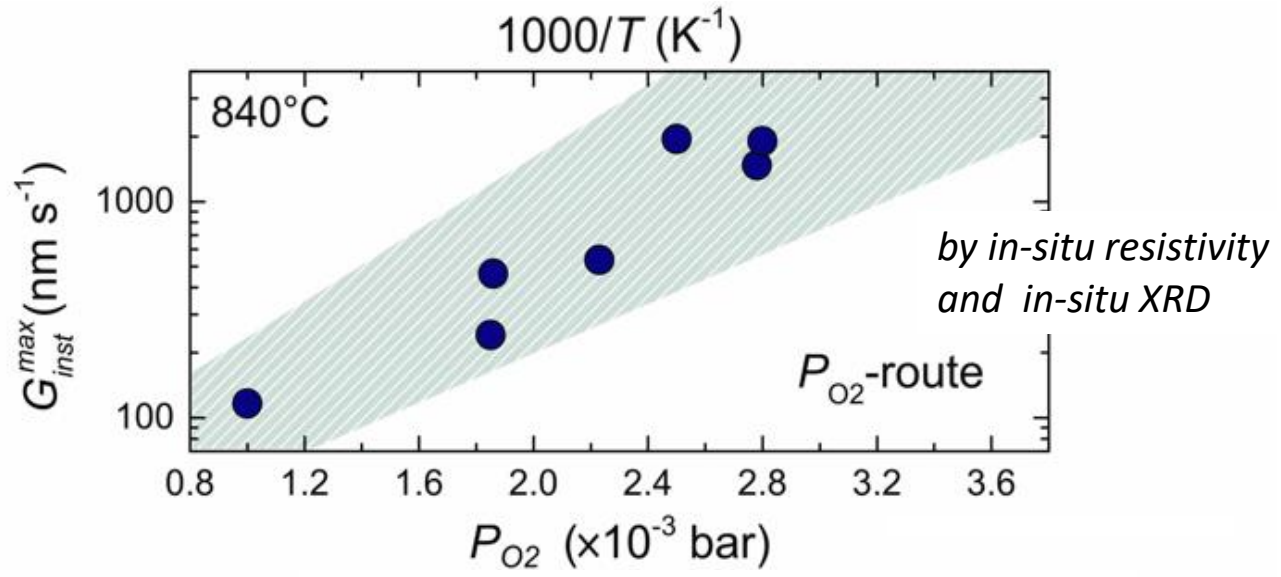


# TLAG Kinetic phase diagrams

P-route ↑

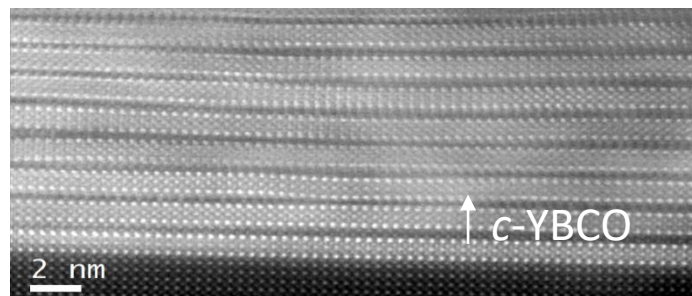
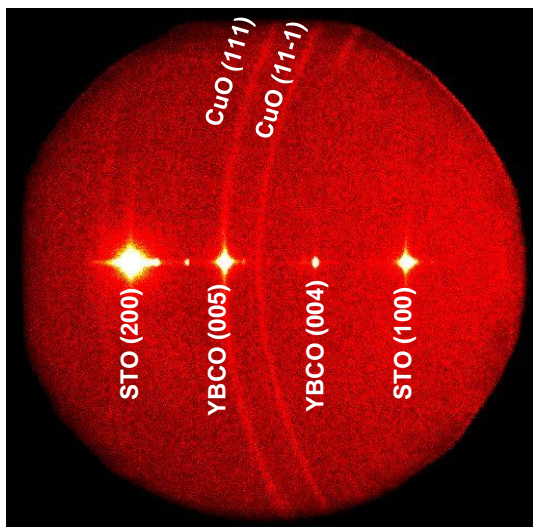


- △  $BaCuO_{2(s)} + YBCO + CuO_{(s)}$
  - ▲  $YBCO + Cu_xO_{(s)}$
  - ▲  $BaCu_2O_{2(s)} + YBCO + Cu_xO_{(s)}$
  - ◻ YBCO
- $T_E$  —  $BaCuO_{2(s)} + CuO_{(s)} \rightarrow [BaCuO_2 + CuO]_{(l)}$
  - $T_m$  —  $BaCu_2O_{2(s)} \rightarrow [BaCu_2O_2]_{(l)}$
  - (a) —  $CuO_{(s)} \rightarrow Cu_2O_{(s)}$
  - (b) —  $BaCuO_{2(s)} \rightarrow BaCu_2O_{2(s)}$

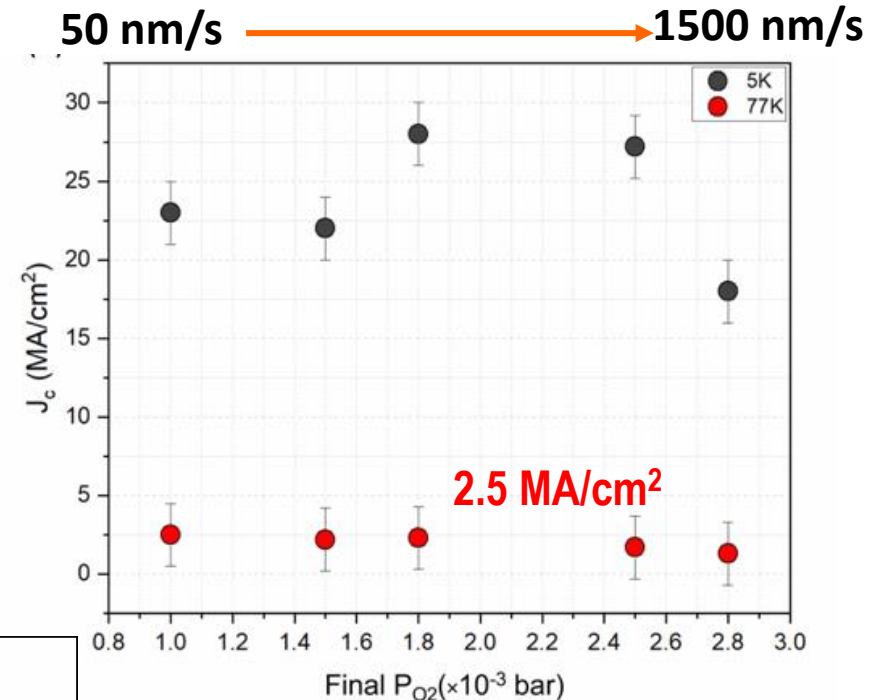
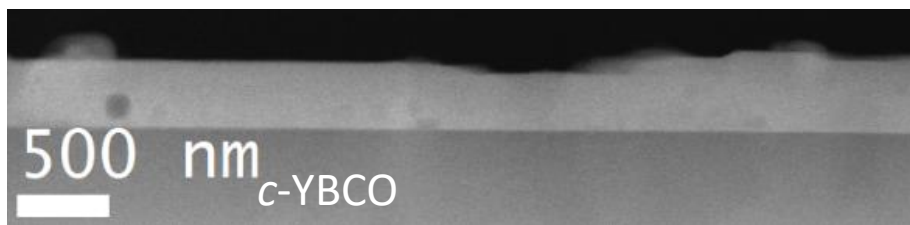


Epitaxial films and nanocomposites at 2000 nm/s

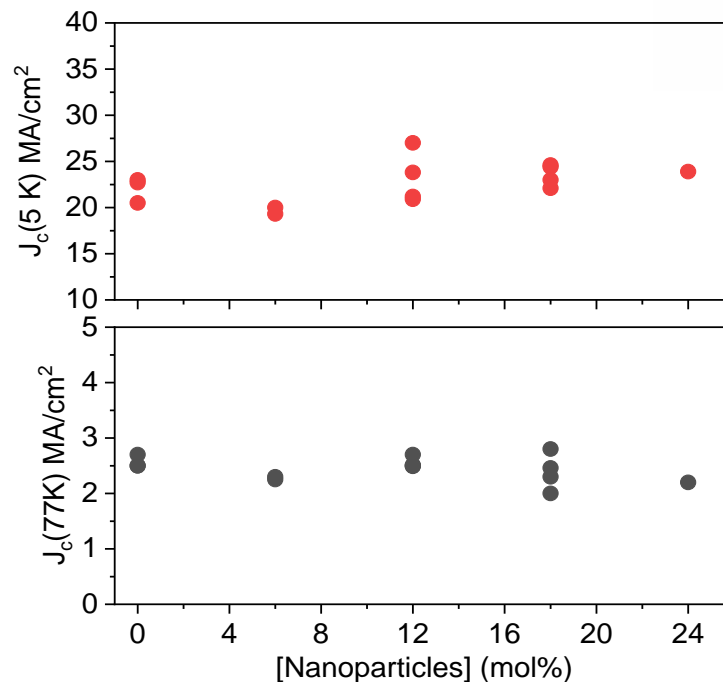
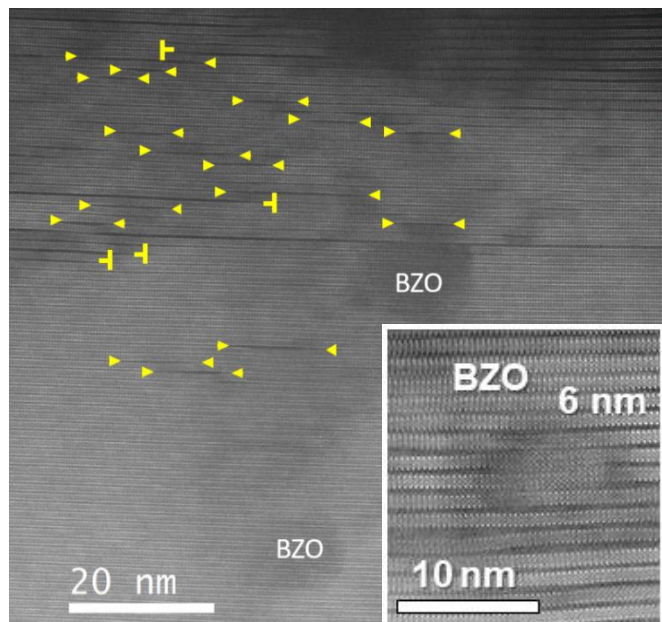
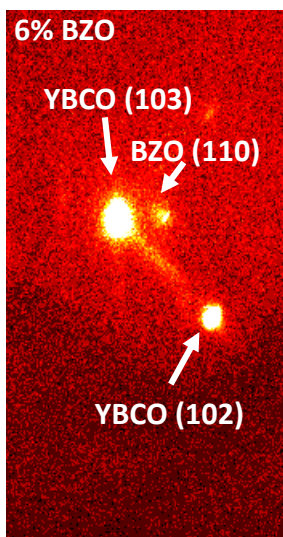
# YBCO TLAG-CSD FILMS



$\gamma_{\text{eff}} = 2.5-3.5$



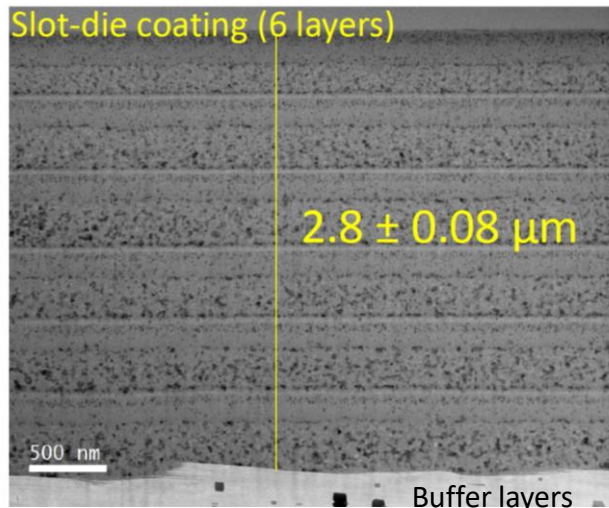
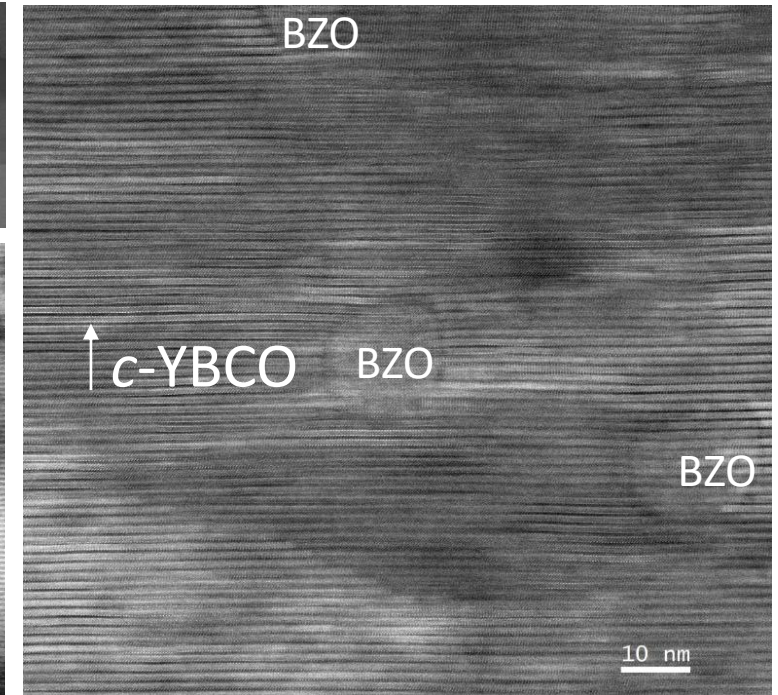
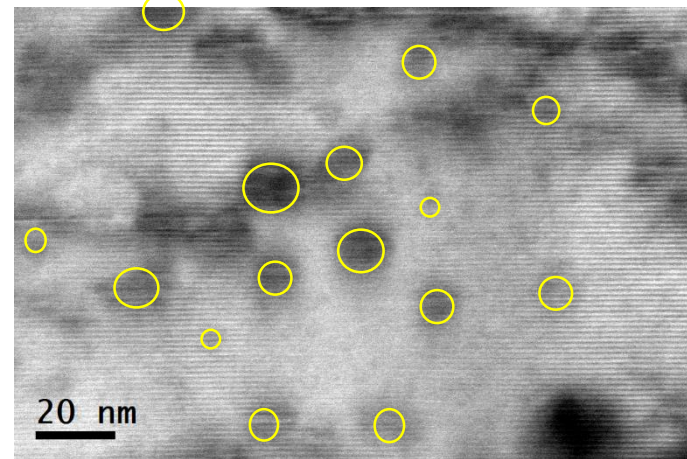
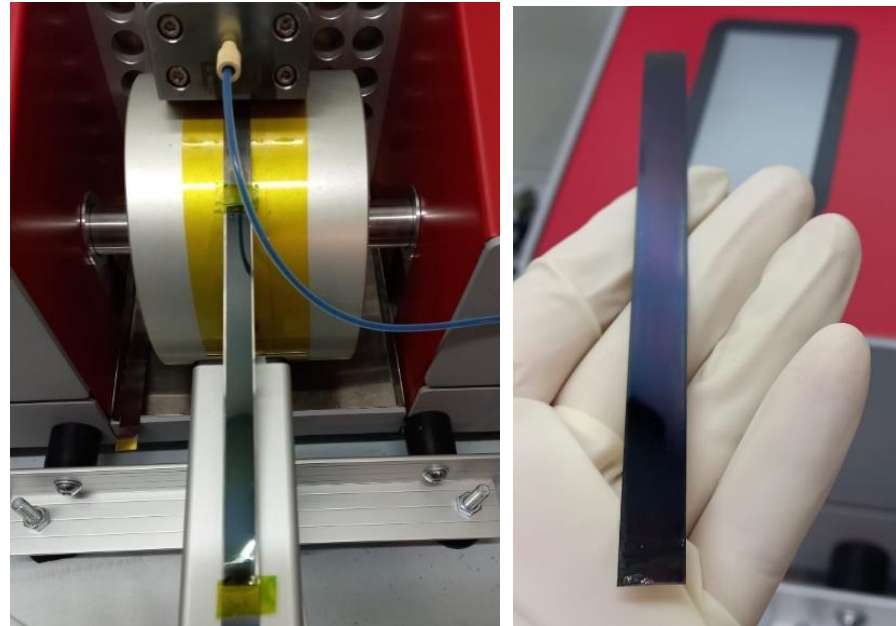
## Nanocomposites



$\gamma_{\text{eff}} = 2-2.5$

# YBCO TLAG-CSD NANOCOMPOSITE FILMS

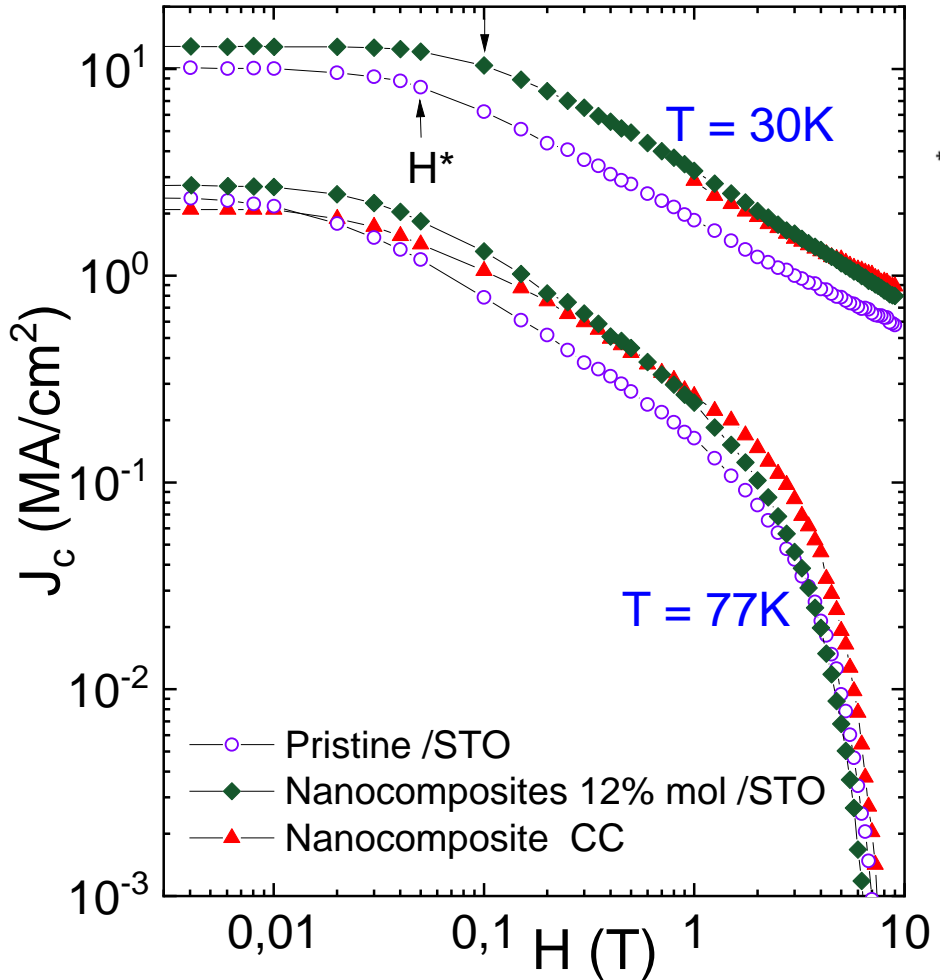
Extended to technical substrates  
in collaboration with



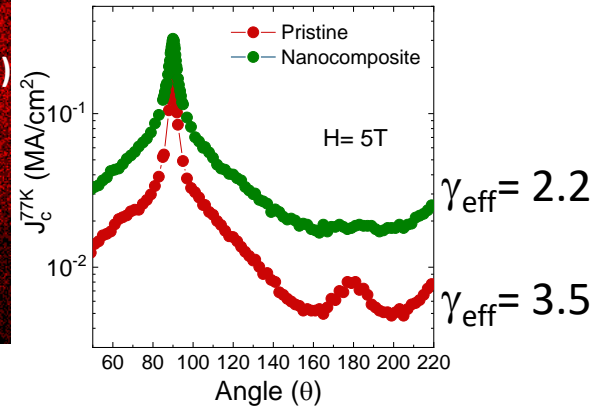
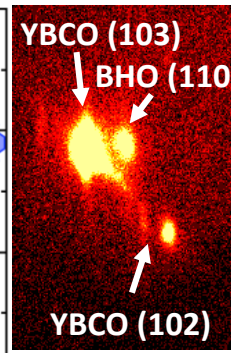
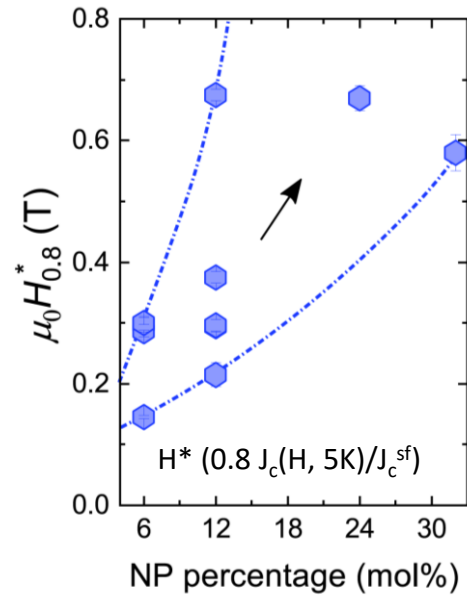
$$J_c(5K) = 24 \text{ MA/cm}^2 \quad J_c(77K) = 2 \text{ MA/cm}^2, \quad I_{c, 750 \text{ nm}}(77K) = 130 \text{ A-w}$$

High density of defects is present as well as embedded nanoparticles

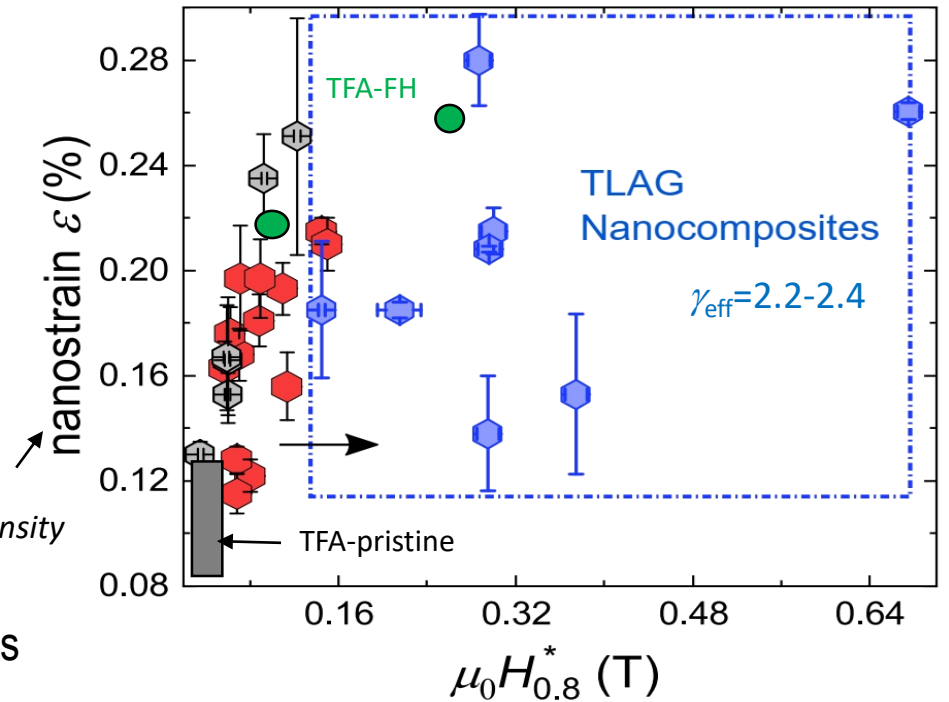
# VORTEX PINNING IN TLAG-CSD NANOCOMPOSITE FILMS



TLAG films respond to vortex pinning determined by high density of defects (SF, point vacancies) and nanoparticles

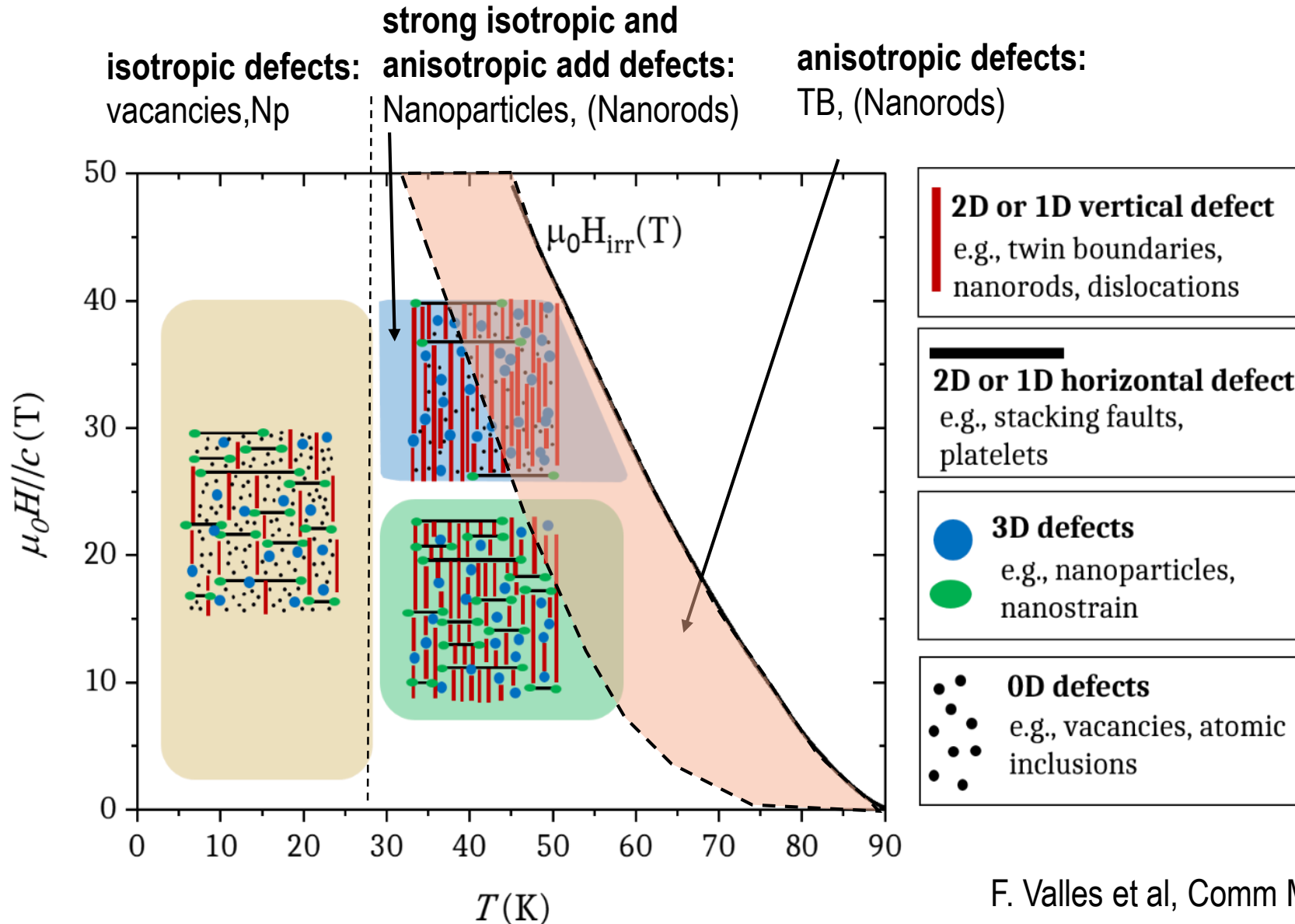


from XRD William son-Hall analysis associated to SF density



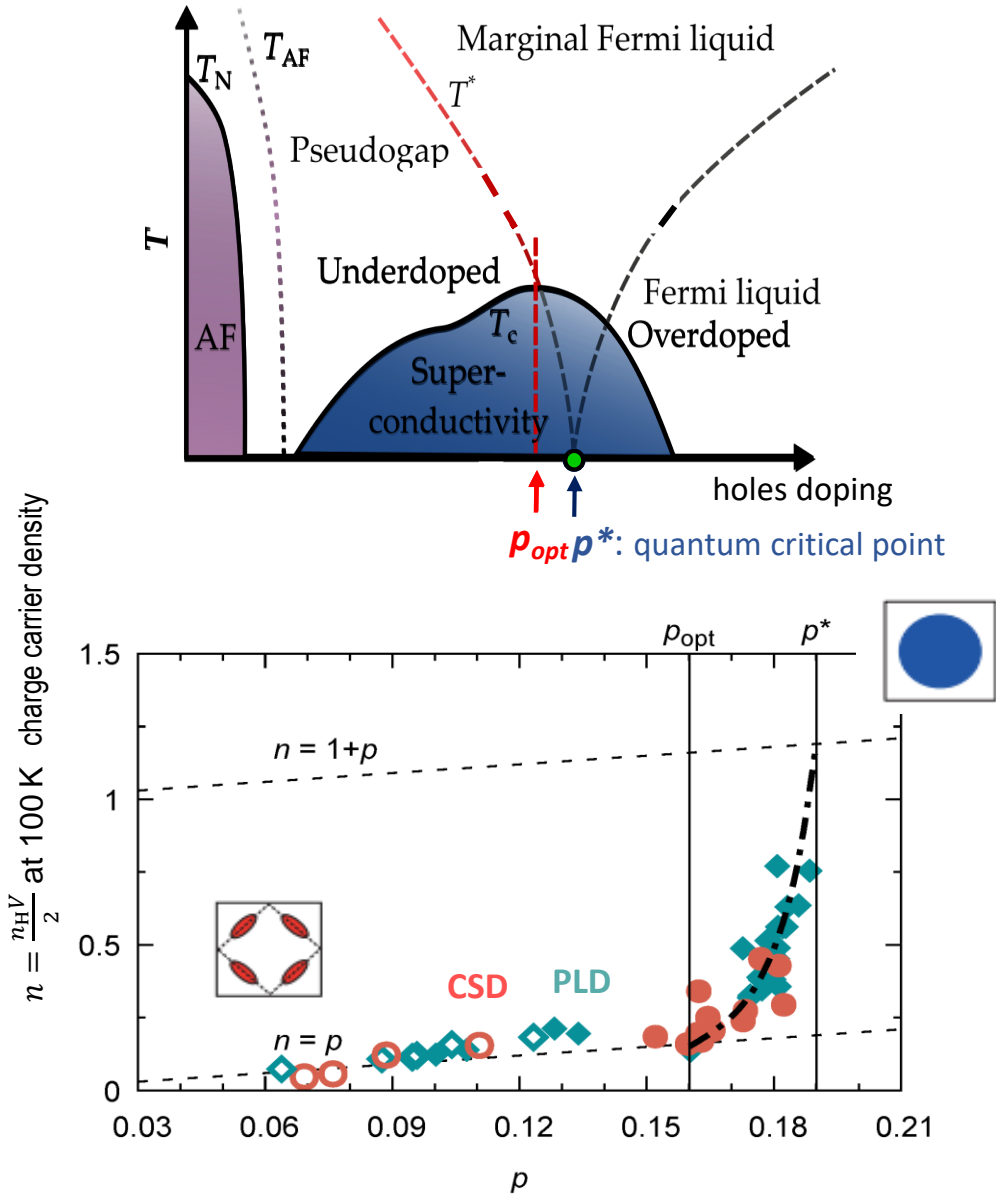


# OPTIMIZING PINNING LANDSCAPES



Different microstructure requirements for different regions

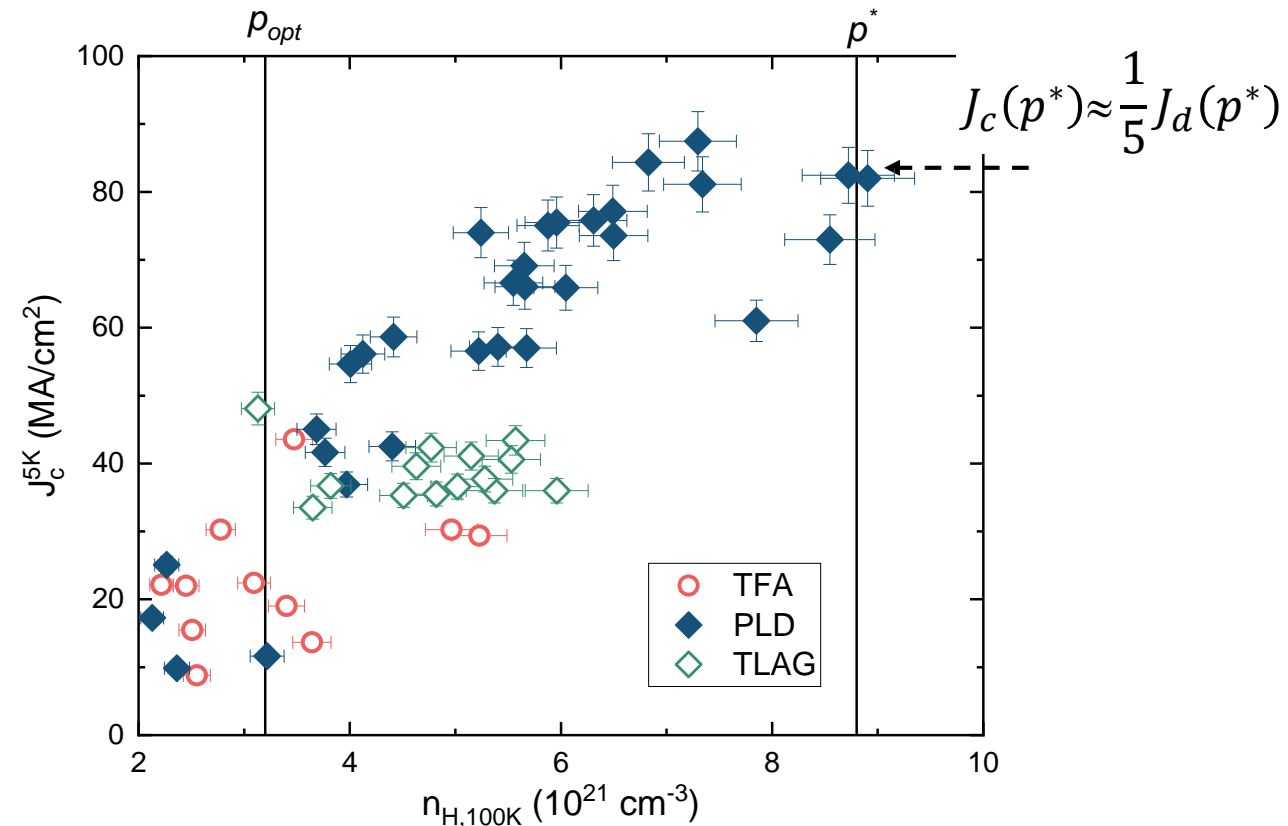
# OVERDOPING: A ROBUST METHOD FOR PINNING



$$J_c^2 \propto n_H E_c(n_H)$$

A. Stangl et al, Sci. Rep. (2021)

Condensation energy and charge carrier density increases in the overdoped state



A. Stangl et al, Sci. Rep. (2021)

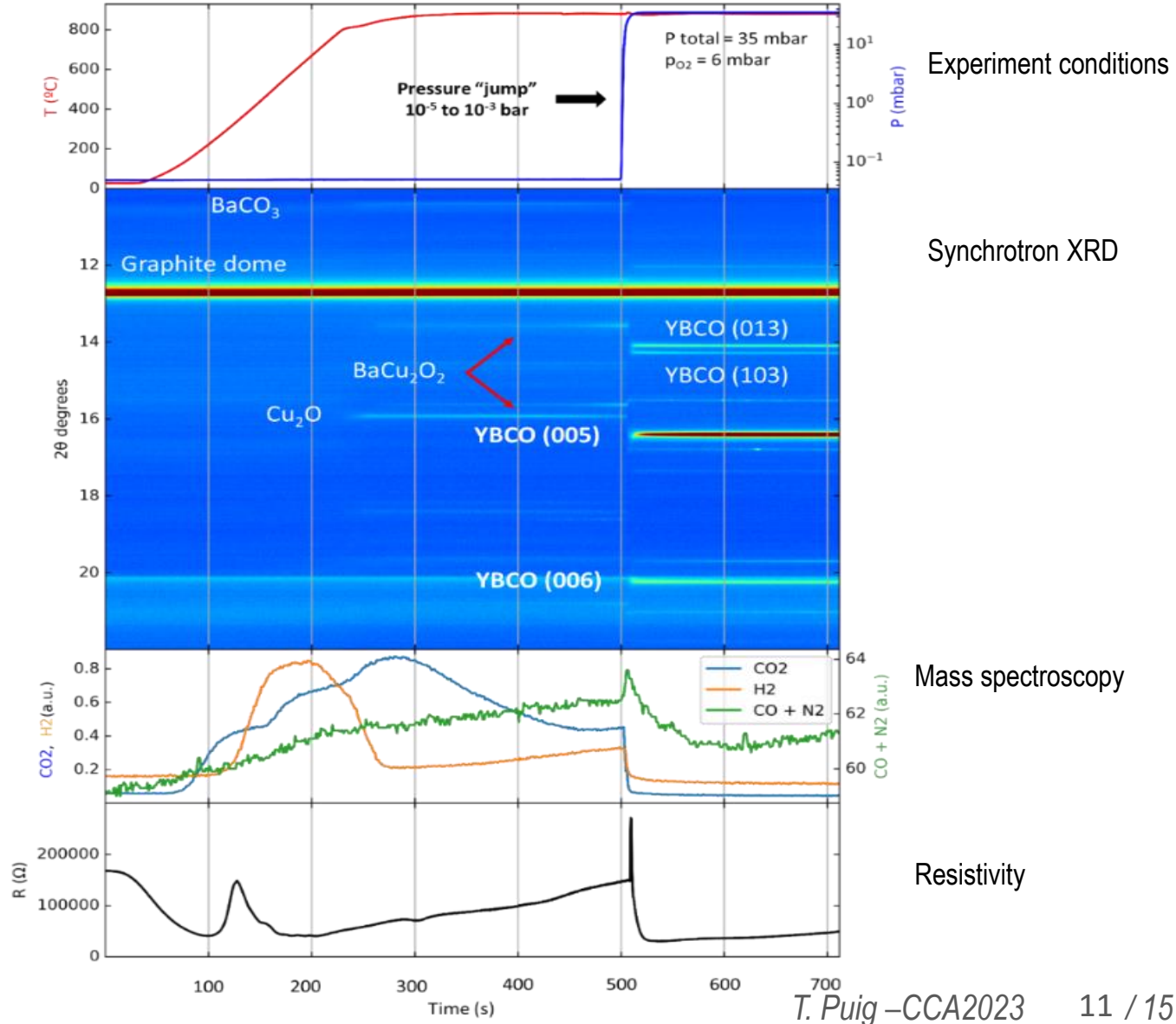
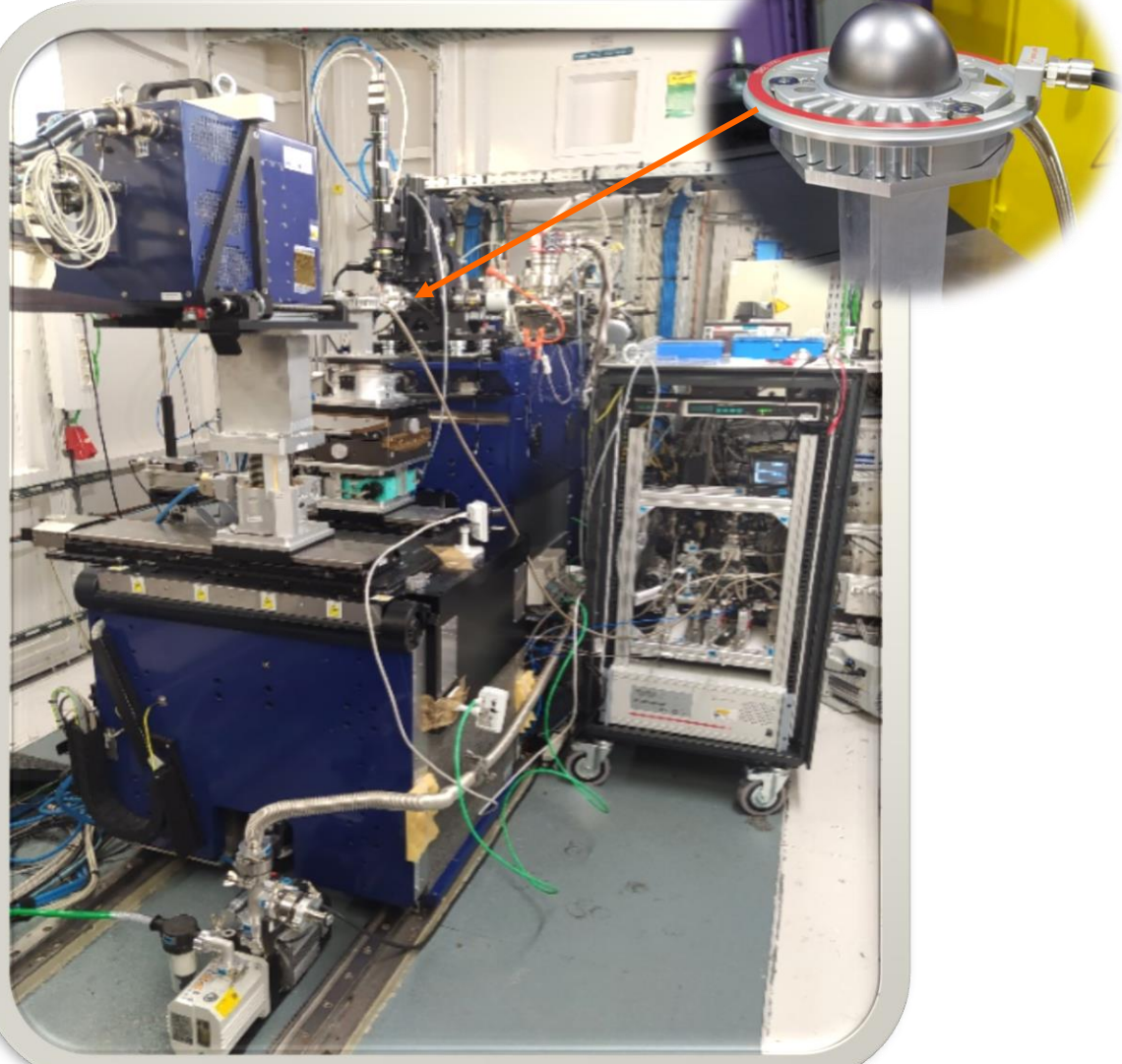
# HIGH THROUGHPUT EXPERIMENTATION



## In-Situ ALBA Synchrotron installation

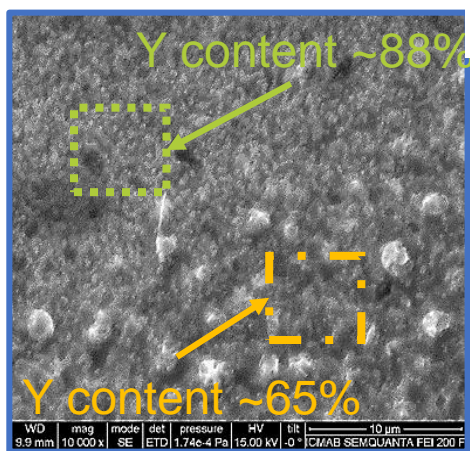
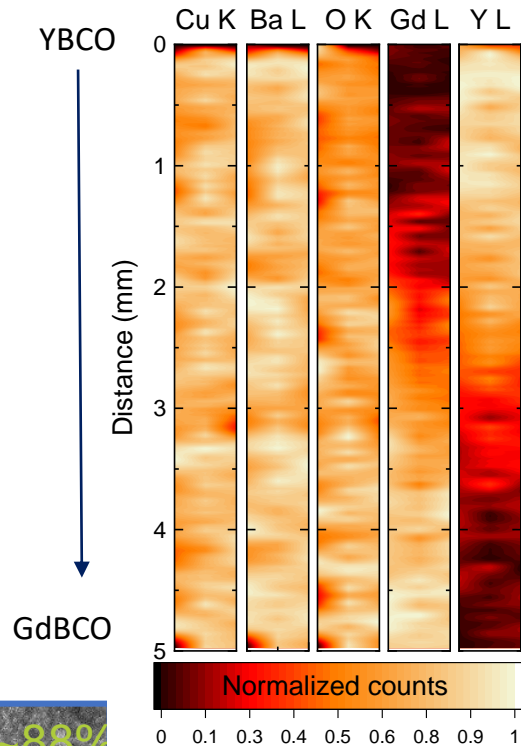
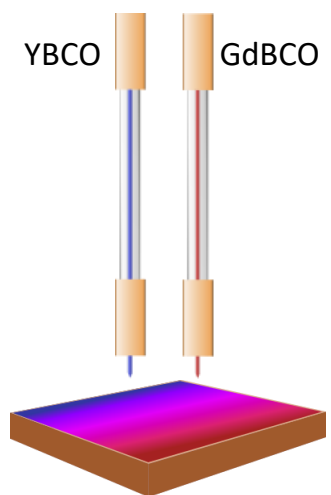
NCD-SWEET BEAMLINE  
GIWAXS detector

Heating stage

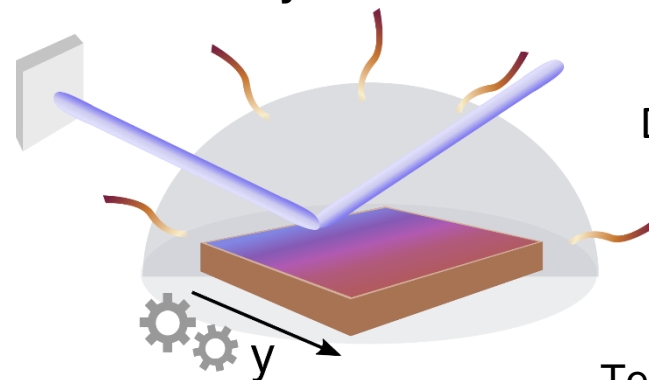


# Fast optimization using Compositional Gradients

## Combinatorial DoD Ink Jet Printing



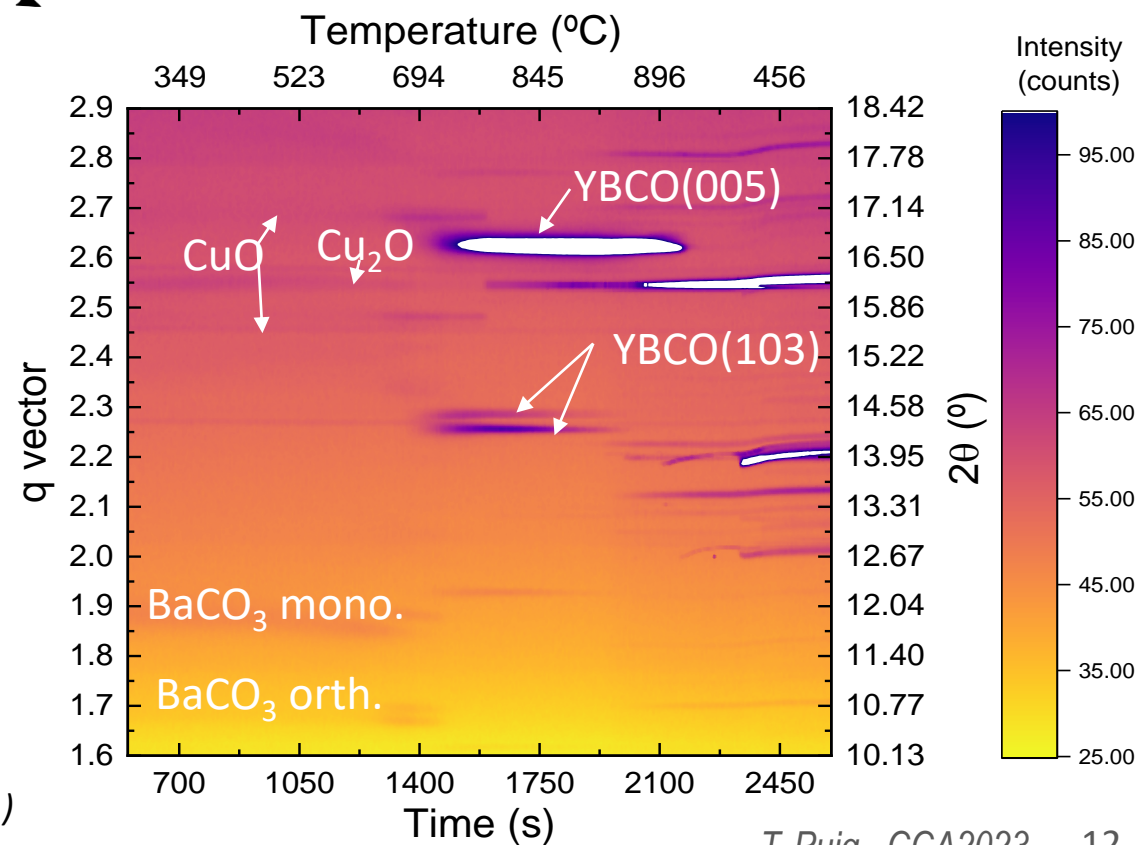
## in-situ synchrotron XRD



Data is segmented by positions for analysis

Case position 1

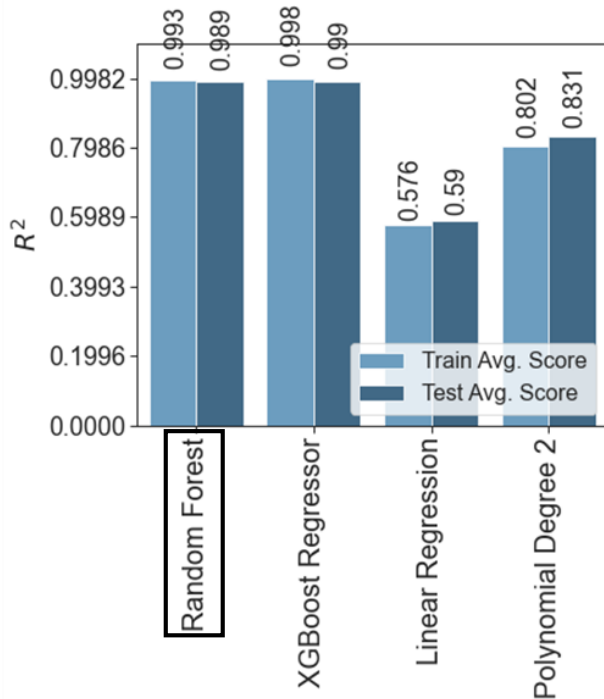
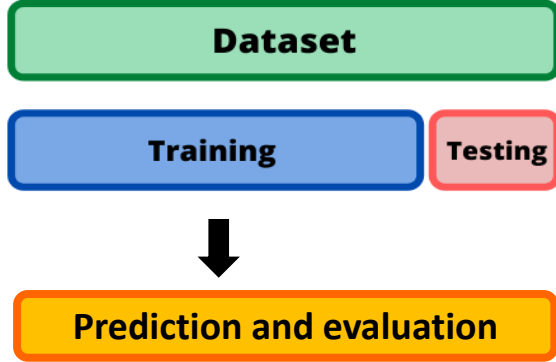
T-route



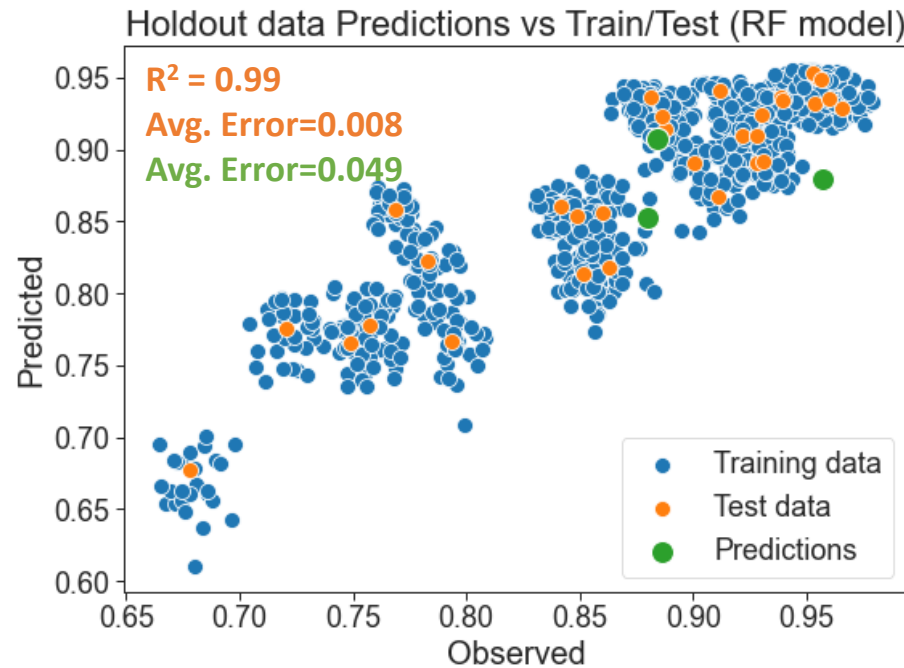
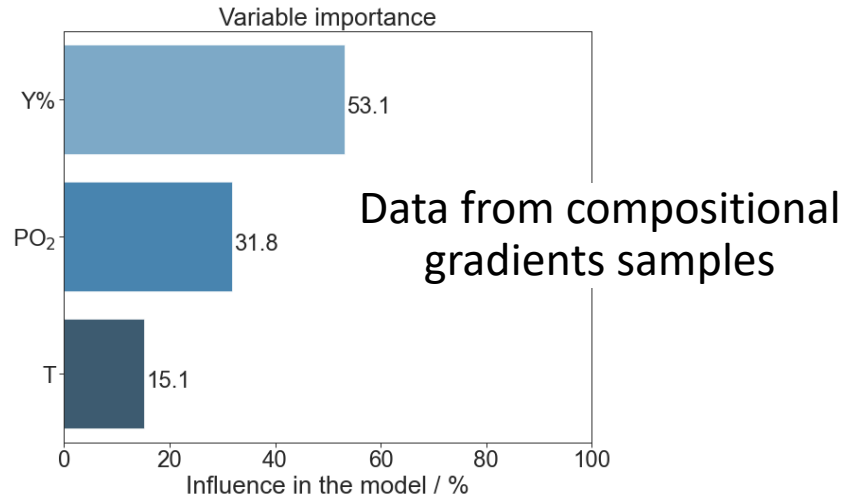
A. Queralto et al., ACS Appl. Mater. Interfaces (2021)



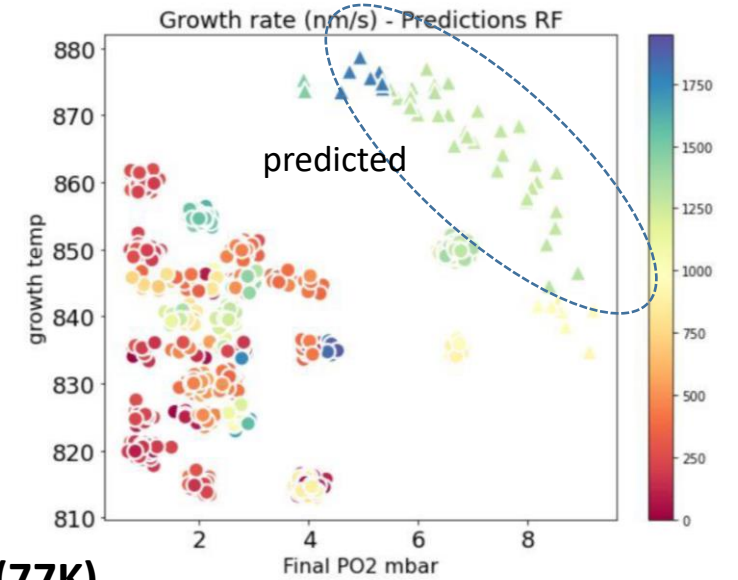
# Machine Learning for fast guideness



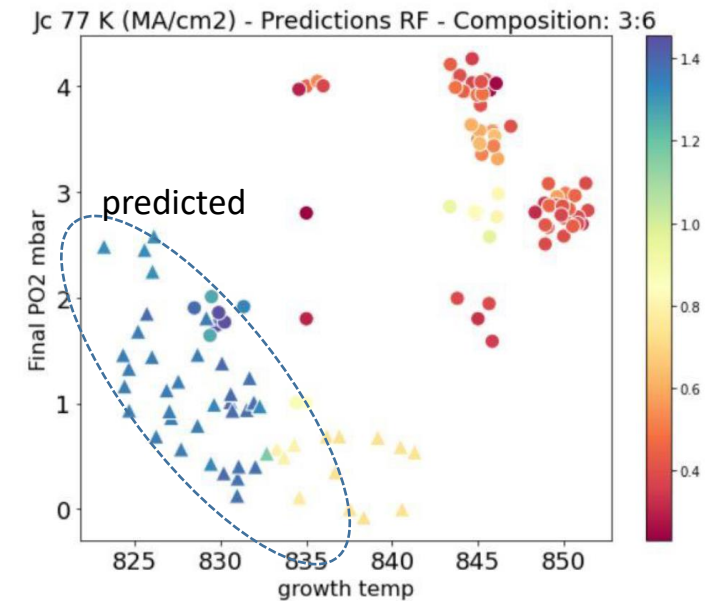
## Optimizing c-axis growth by ML



## Growth Rate



## $J_c(77K)$



# Conclusions

- TLAG is a non-equilibrium growth methodology with ultrafast growth rates achieving high performance Coated Conductors compatible with CSD
- In-situ synchrotron XRD experiments have been essential to understand the kinetic growth mechanisms and obtain epitaxial layers at ultrafast growth rates
- Pinning landscapes can be optimized at different regions depending on applications needs, overdoping being very appealing
- High Throughput Experimentation initiatives are foreseen to fasten optimization schemes
- Industrialization of TLAG should lead to a high throughput manufacturing process which uses simple and large area reactors for R2R CC production

# Contributions from CC Materials Research

