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ICMAB INSTITUT DE CIÈNCIA DE MATERIALS DE BARCELONA EXCELENCIA SEVERO OCHOA

CSIC CONSEJO SUPERIOR DE INVESTIGACIONES CIENTÍFICAS

Progress and Opportunities of the Transient Liquid Assisted Growth (TLAG) method

Teresa Puig[1]

X. Obradors[1], R. Vlad[1], E. Pach[1,4], C. Pop[1], K. Gupta[1], C. Torres[1], O. Mola[1], A. Kethamkuzhi[1], E. Ghiara[1], V. Bertini[1], M. Voulhoux[1], D. Sanchez[2], S. Rasi[1], L. Salterilli[1], D. Garcia[1,3], S. Ricart[1], M. Tristany[1], R. Yanez[3], J. Farjas[2], E. Solano[4], L. Simonelli[4], J. Gutierrez[1]

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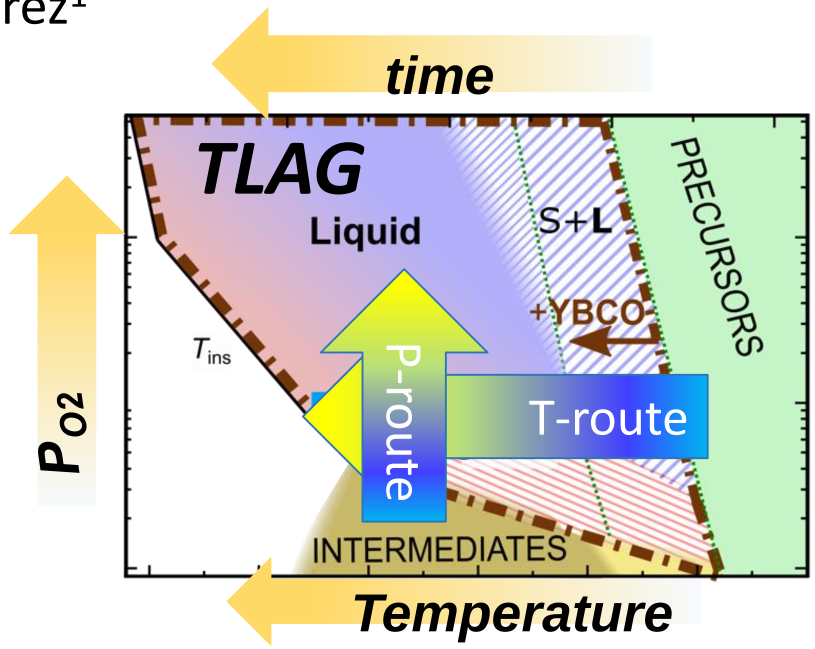

European Research Council Executive Agency

Work is supported by cost EUROPEAN COOPERATION IN SCIENCE & TECHNOLOGY

MINISTERIO DE CIENCIA E INNOVACIÓN

Plan de Recuperación, Transformación y Resiliencia

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AIM: Design a robust, high throughput and flexible growth method for the manufacturing of CC at competitive cost/performance ratio

Opportunities of TLAG

-High growth rates > 1000 nm/s
-Compatible with low-cost chemical solution deposition (CSD) and others (i.e Low Temperature PLD deposition, LTPLD)
-Wide processing window
-Compatible with different Rare Earch (RE) and nanocomposite growth
-Well-suited for large area fabrication
-Easily scalable manufacturing equipment

Need of R&D in advanced CC materials

-Generate new ideas and understanding
-Develop breakthroughs that enable high-volume CC production at lower cost/performance
-Fast screening methodologies and AI approaches to guide predictions
-Contribute to making CC production a sustainable and transformative technology

Outline

-Progess in process understanding and processing windows
-Growth rates
-High Throughput Experimentation approach
-Vortex pinning and overdoping
-Towards large area Coated Conductors

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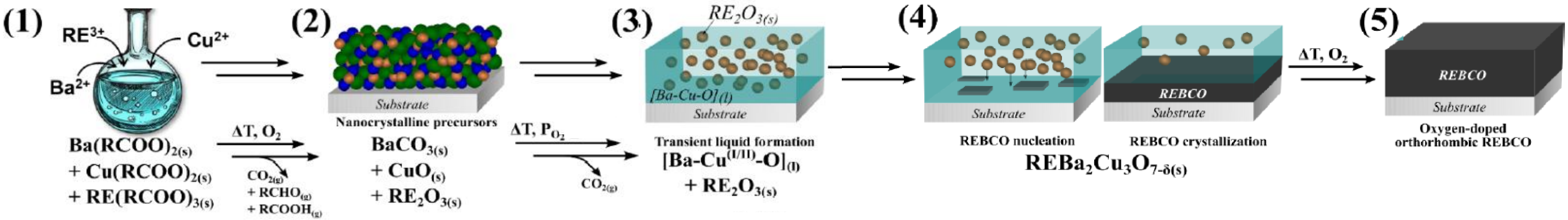
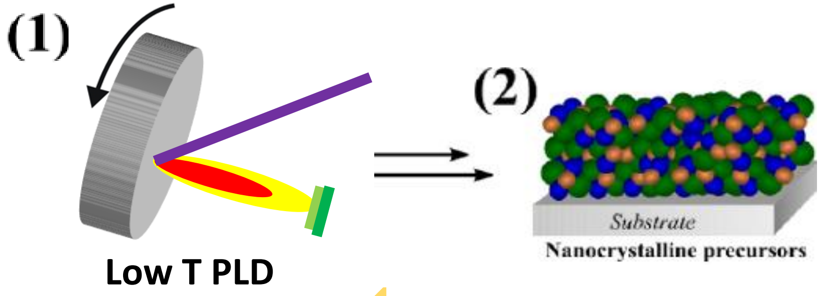
❶ TLAG-CSD process



TLAG-PLD process

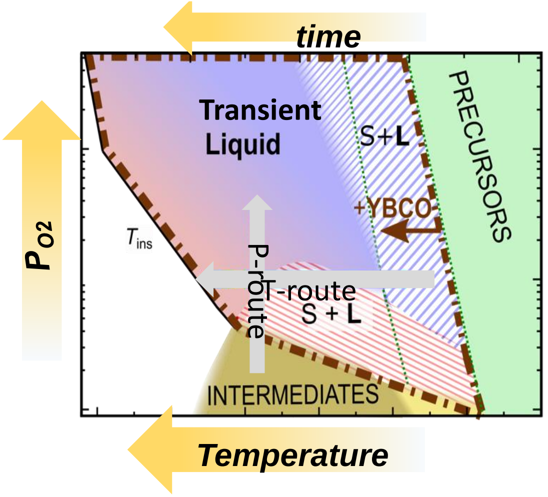


Kinetically governed by RE liquid supersaturation $\sigma = (C_\delta - C_e) / C_e$

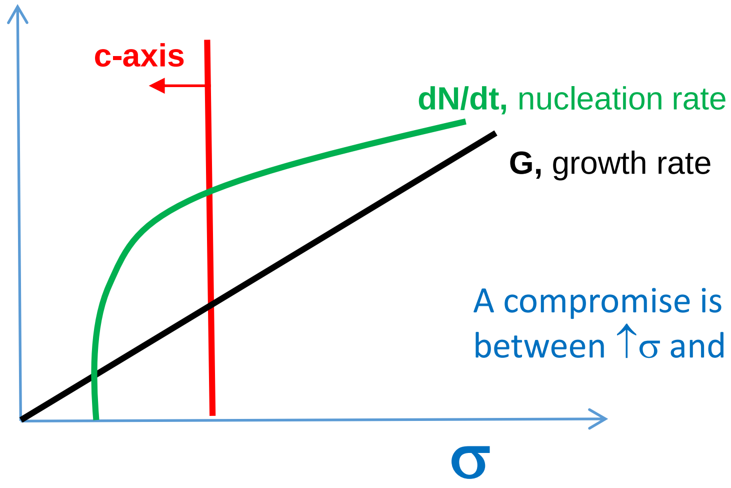
C_e: **thermody. param.** (T, P_{O2}, Ba/Cu liq. composition, RE ion)

C_δ : **kinetic param.** (heating ramp, pressure ramp, …)

if 1/T, P_{O2}, Ba/Cu or r_{RE} ↑ → σ ↑



Fast advanced tools have been developed to understand the process



A compromise is needed between ↑σ and epitaxy

L. Soler et al., Nat Comm (2020), S. Rasi, et al, Advance Science (2022)
A. Queralto et al, SUST (2023)

Inks, pyrolysis and multidepositions in TLAG-CSD

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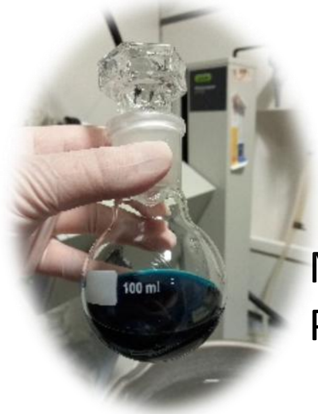
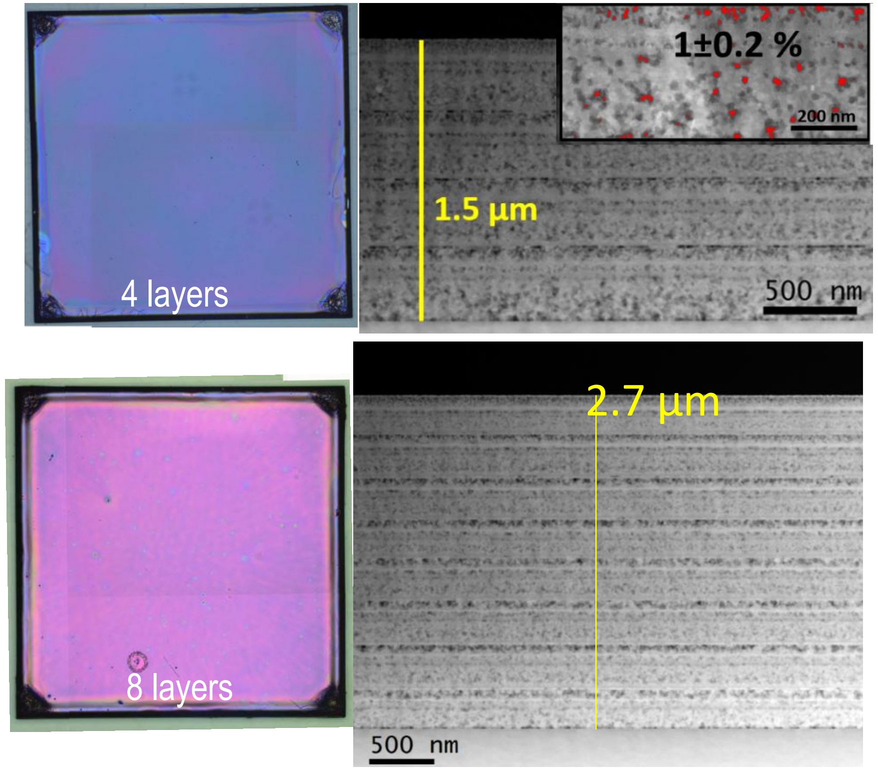
The increase in Cu content facilitates the decrease of supersaturation

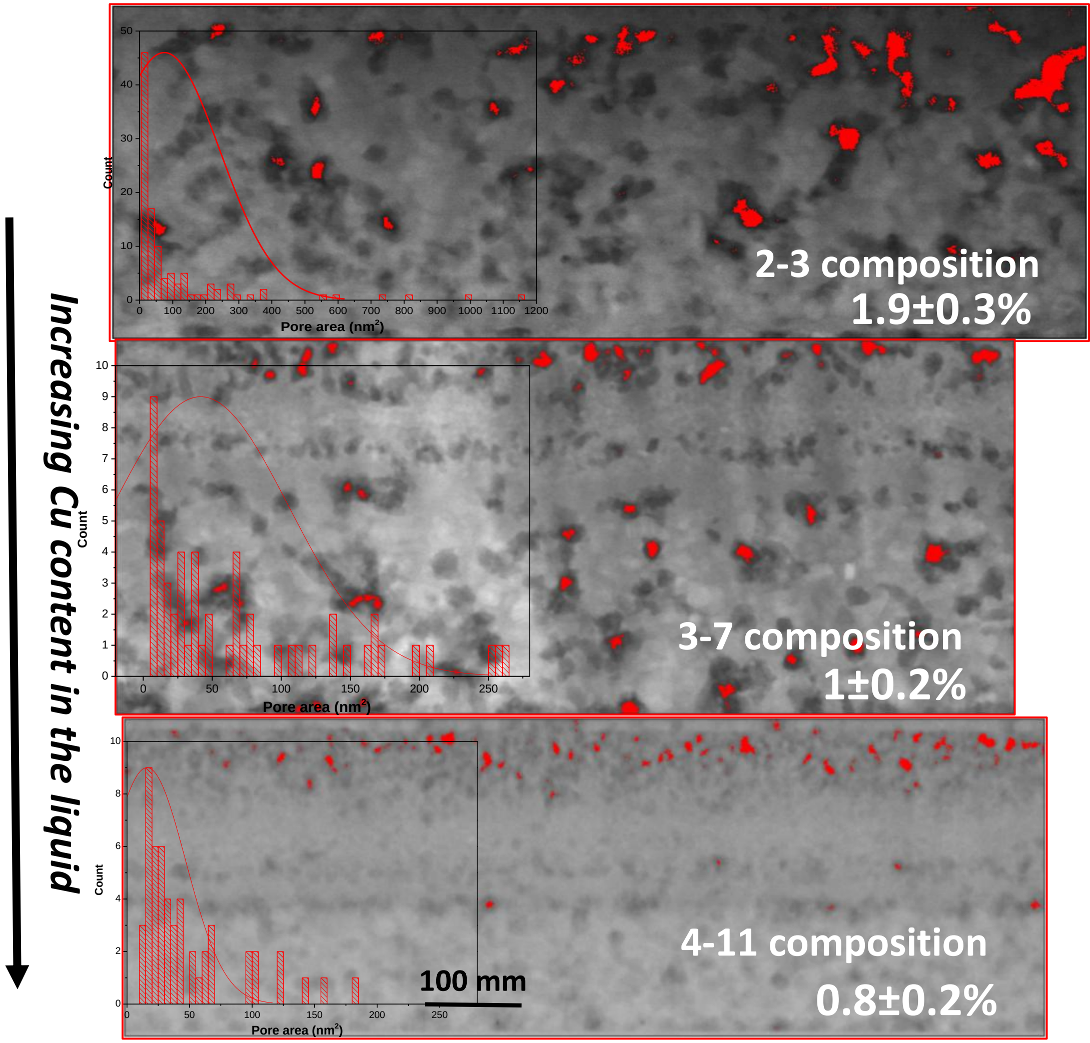


Multifunctional ink
Patent EP22382741

Adapted to multideposition with no loss in homogeneity



Also demonstrates for REBCO inks with RE= Y, Er, Gd, Sm, Yb



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

❷ Understanding TLAG growth mechanisms through in-situ 2D X-Ray Diffraction (XRD)

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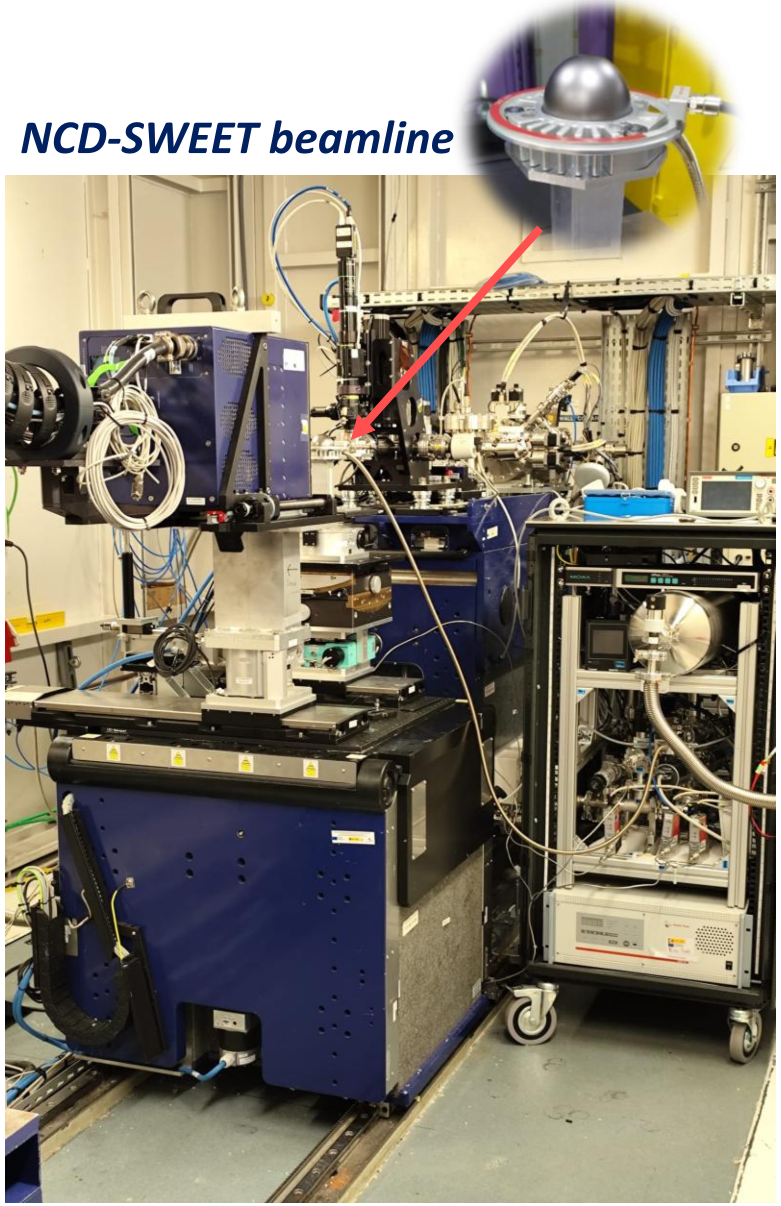
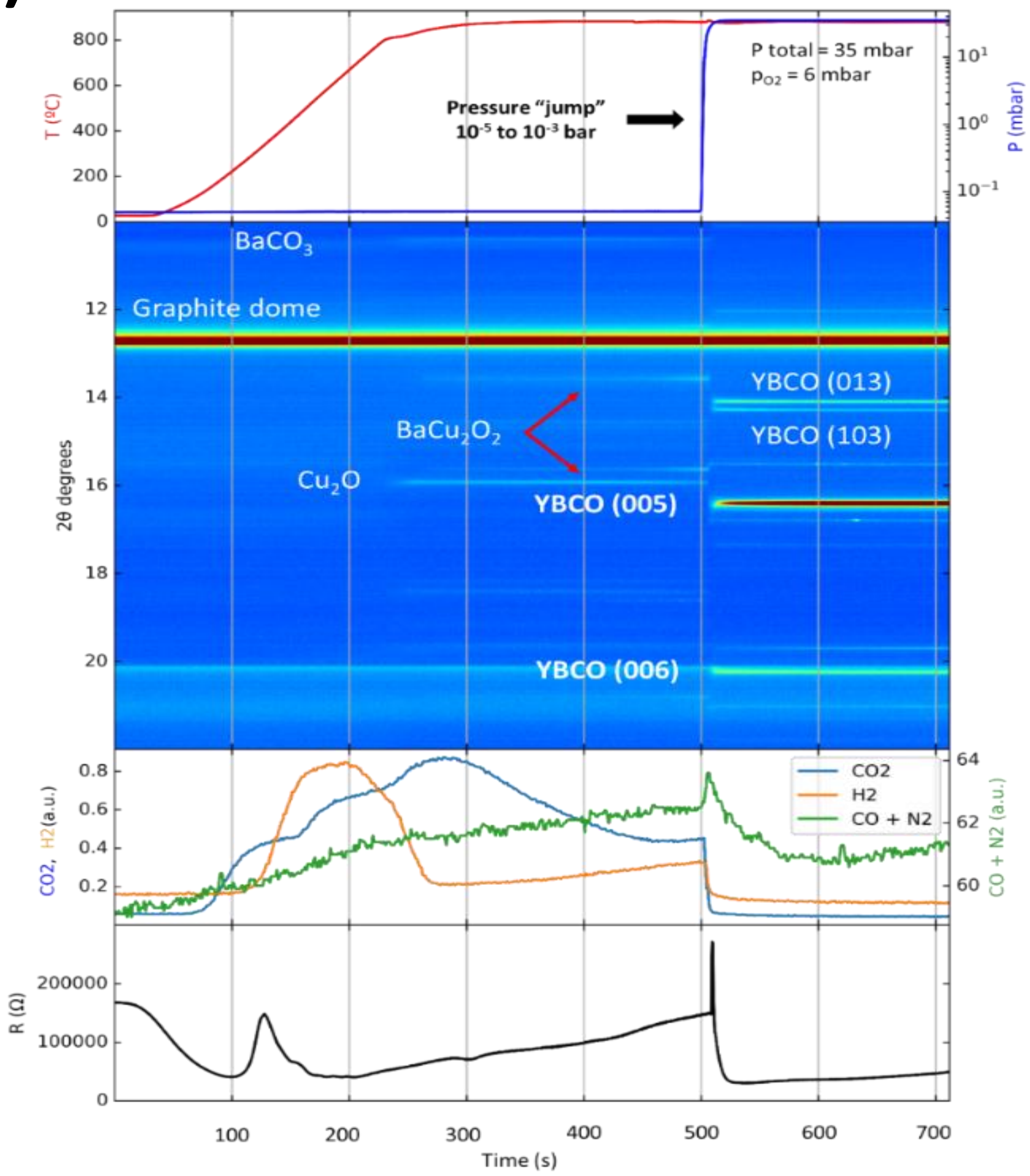
NCD-SWEET beamline





Experiment conditions

ALBA

ETJL Joint laboratory

FARADAY FACTORY JAPAN

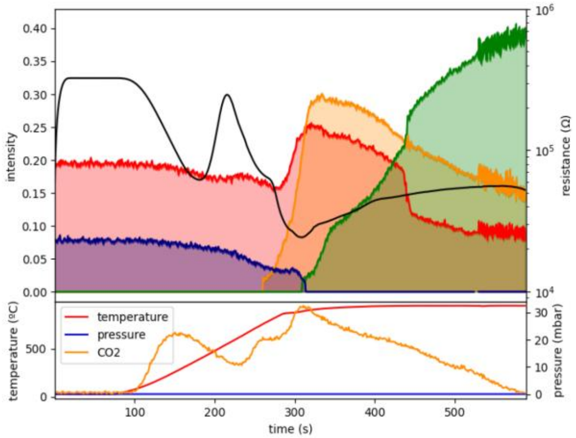
TLAG-LTPLD experiments March 2025

Synchrotron XRD

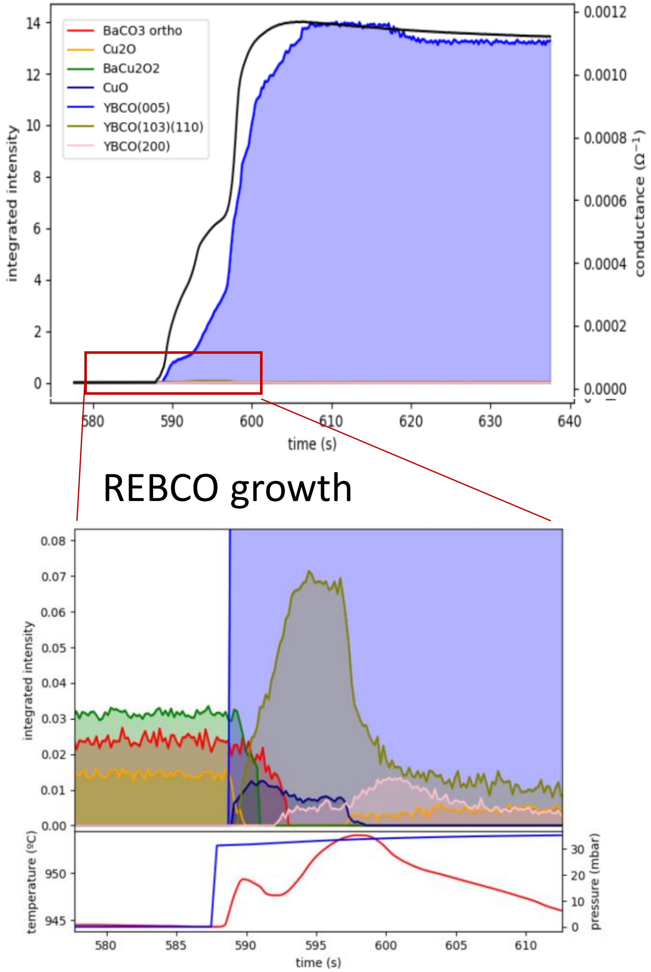
Mass spectroscopy

Resistivity

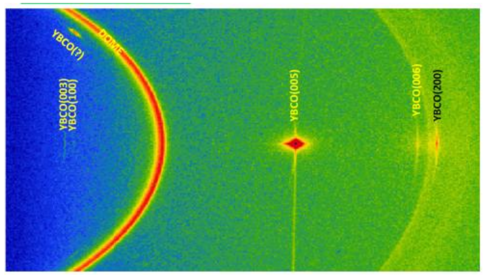
TLAG kinetic phase diagram from evolution of phases



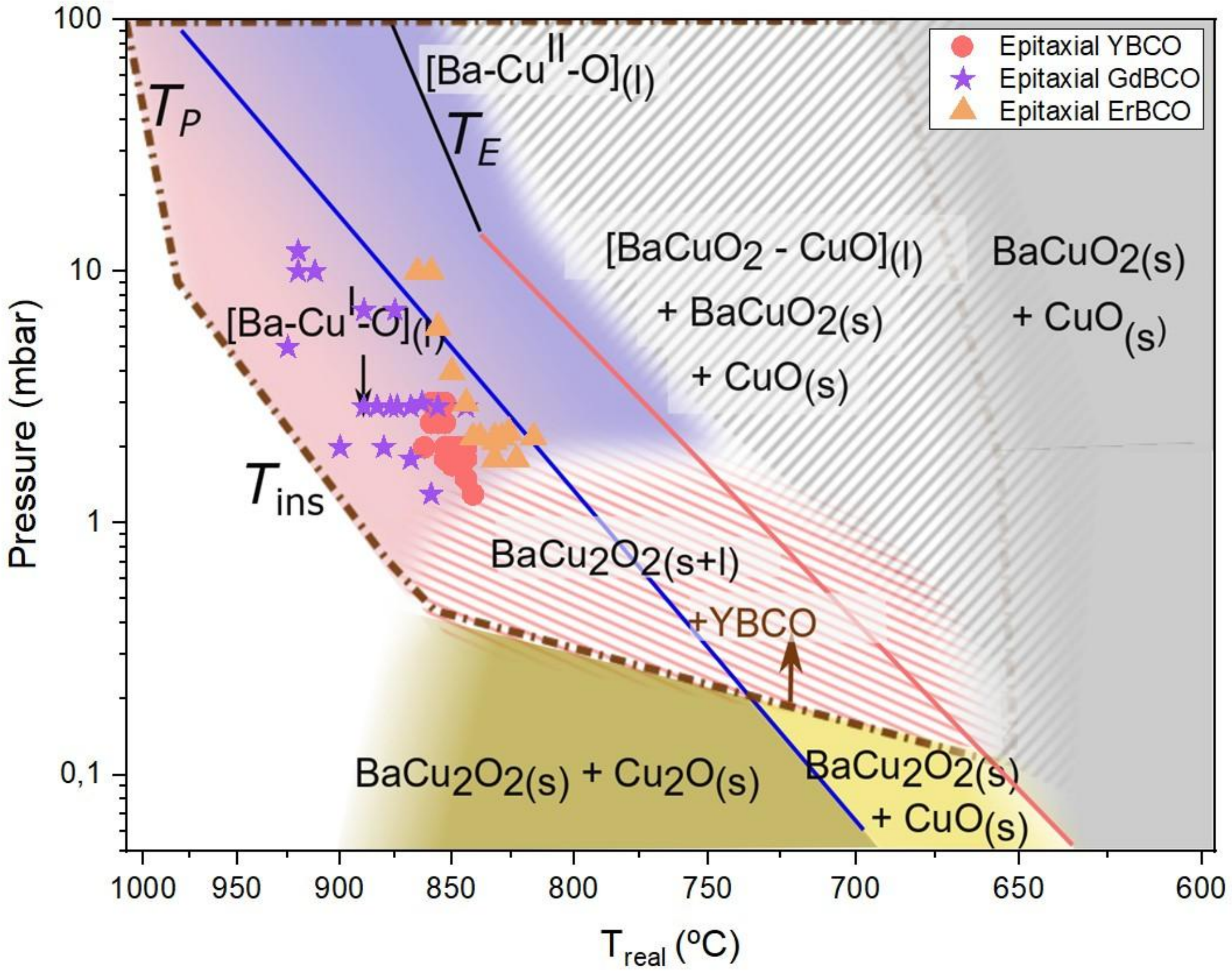
$BaCO_3$ elimination and intermediate phases



REBCO growth



Epitaxial growth



Wide processing window further controlled by RE ion

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Understanding TLAG growth mechanisms through in-situ X-Ray Absorption Spectroscopy (XAS)

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ALBA

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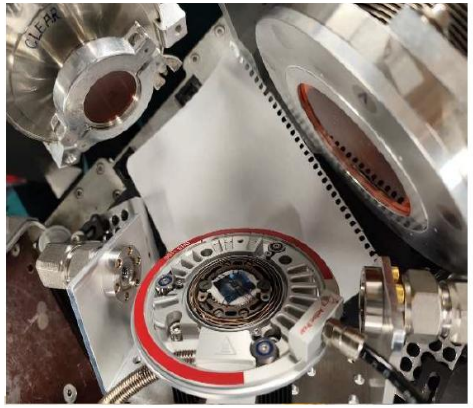
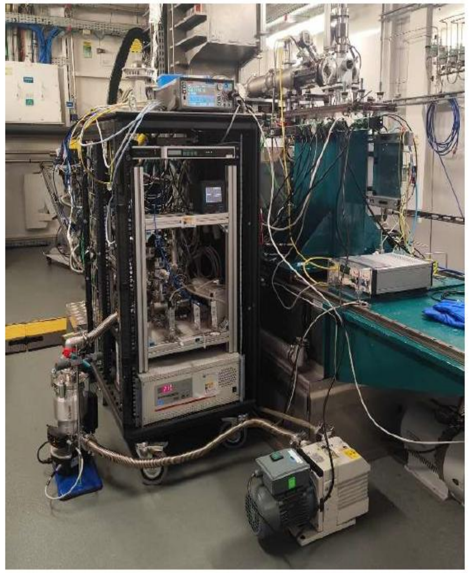
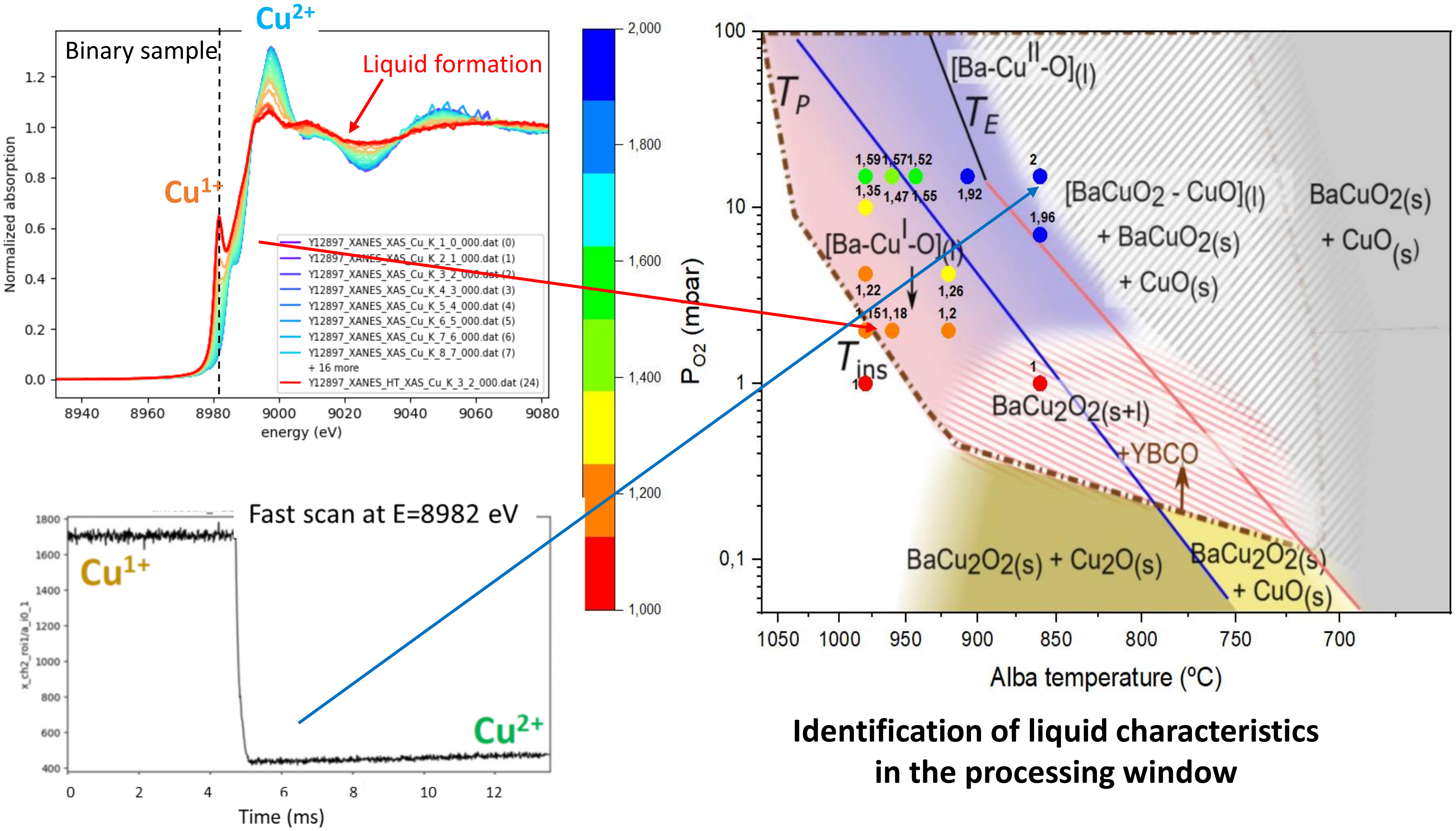


CLAESS beamline

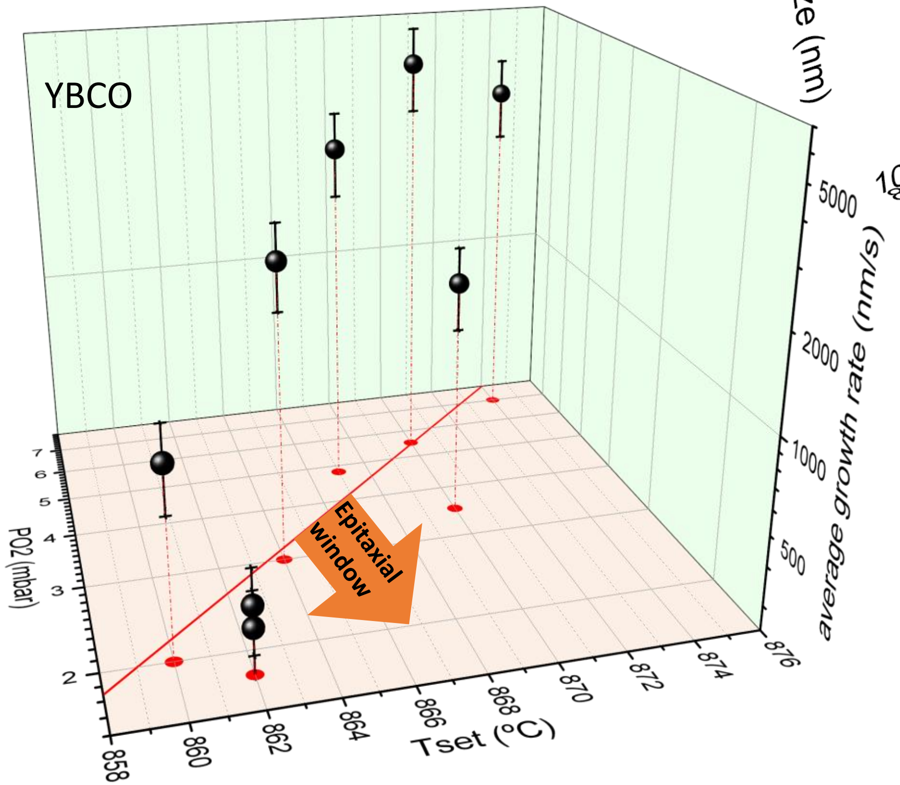




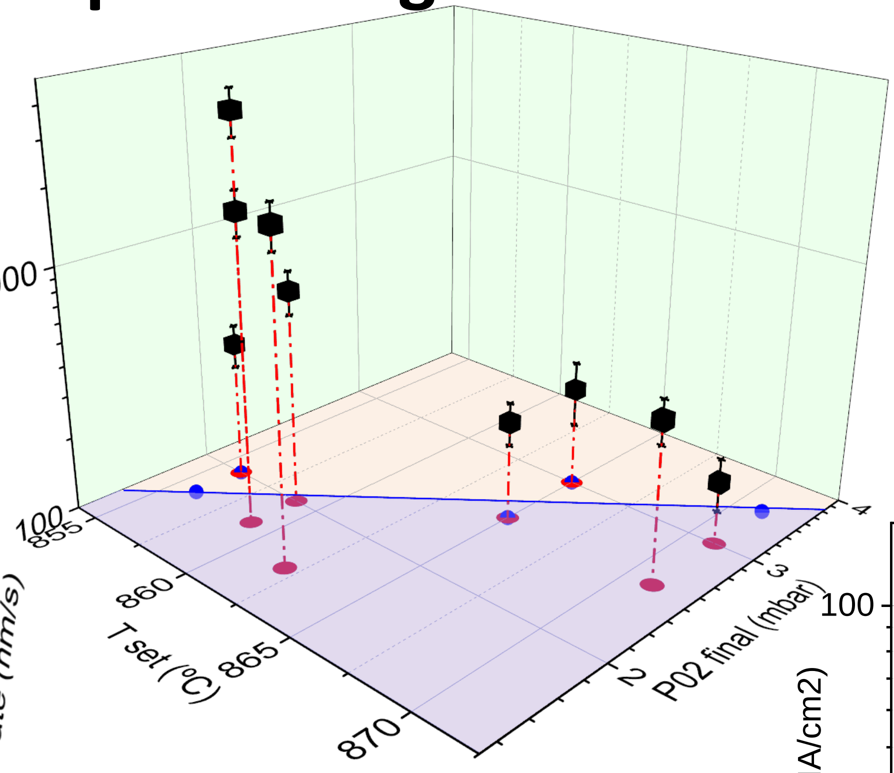
Identification of liquid characteristics in the processing window

❸ TLAG growth rates in the processing window

By in-situ resistivity

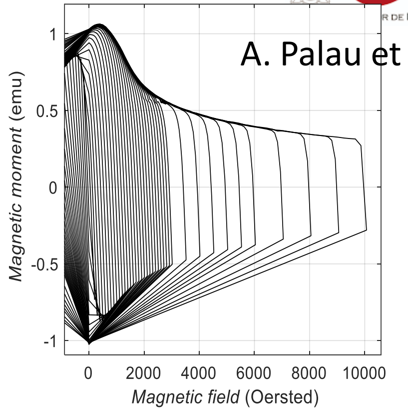


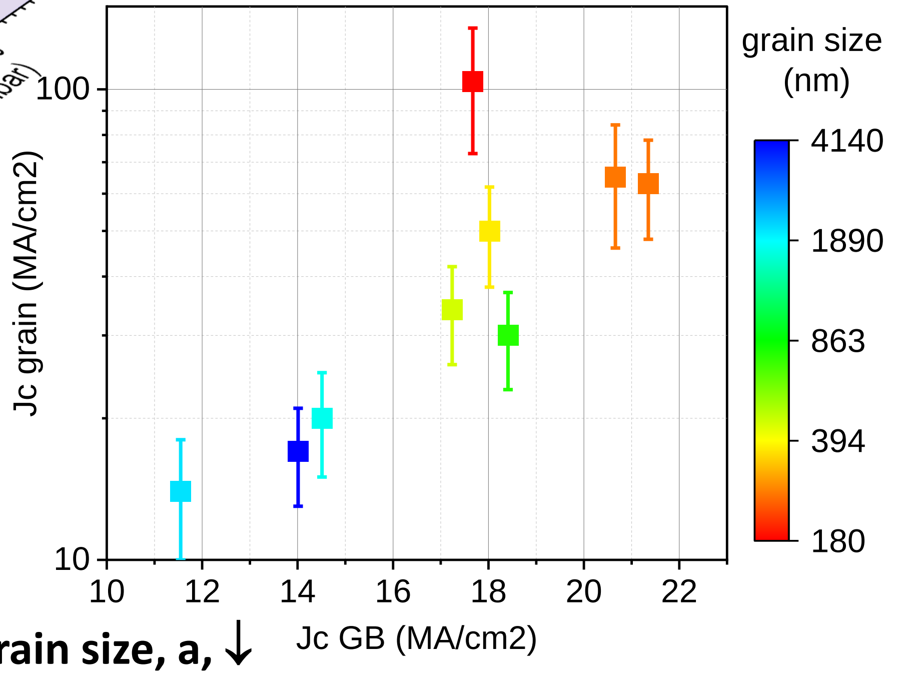
Growth rate, G, ↑ if P_{O2} (and T) ↑



Grain size, a, ↓ if P_{O2} (and T) ↑
(in agreement with ↑ nucleation density and ↑ supersaturation)

Going to high G is beneficial also for J_c





J_c ↑ if grain size, a, ↓
(in agreement with other CSD methods)

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❹ High Throughput Experimentation using Combinatorial RE Compositional gradients

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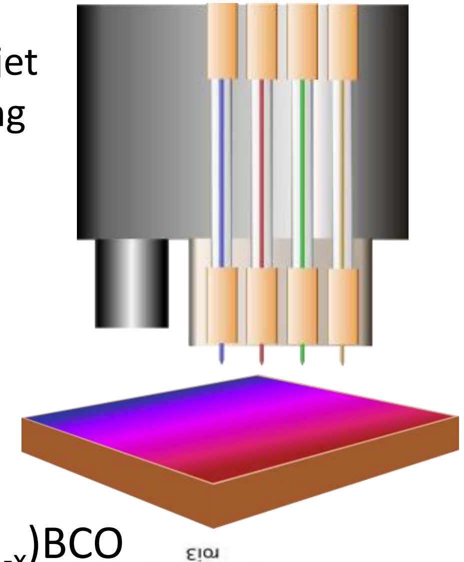
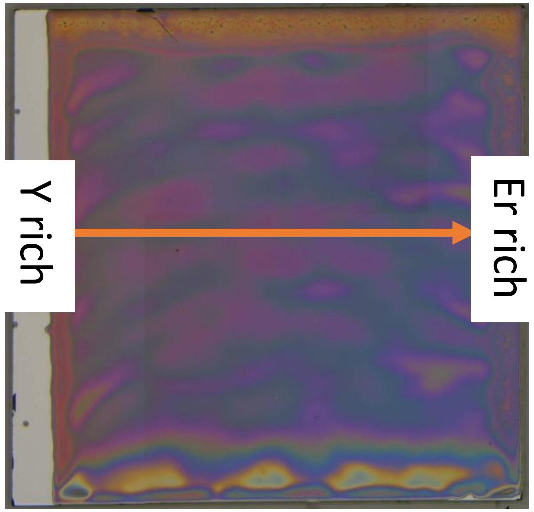
Collab. Z. Wu, T. Kiss — 九州大学 KYUSHU UNIVERSITY

Via inkjet printing

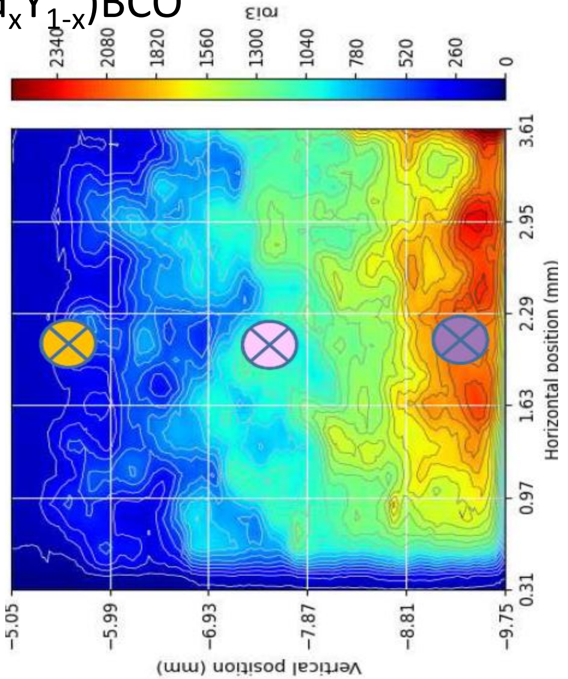


$(Er_xY_{1-x})BCO$



$(Gd_xY_{1-x})BCO$

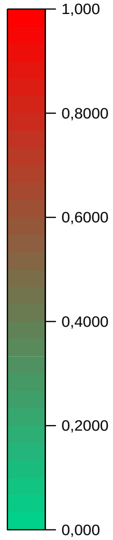
Gd elemental map from XAS



EDX Automated measurement

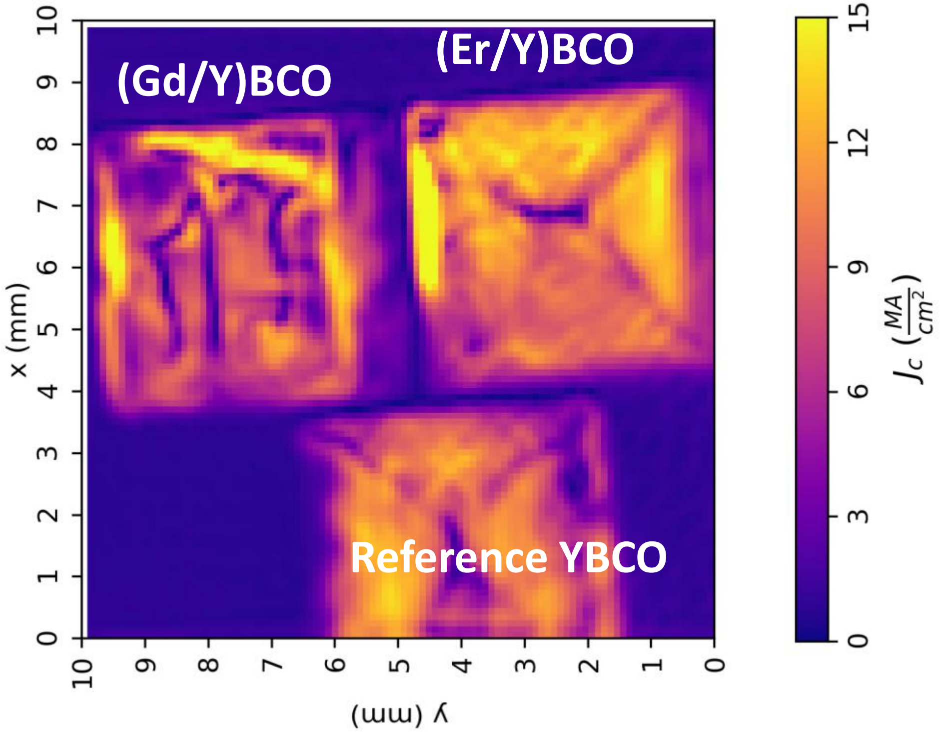
100% Y

y	-5,7	-5,19	-4,68	-4,17	-3,66	-3,15	-2,64	-2,13	-1,62	-1,11	-0,6
2.3	--	--	--	--	--	--	--	--	--	--	--
1.84	86%	79%	73%	78%	59%	55%	37%	39%	33%	35%	--
1.38	89%	84%	66%	65%	59%	41%	48%	35%	28%	22%	--
0.92	--	86%	74%	70%	58%	44%	39%	39%	18%	12%	--
0.46	92%	85%	84%	70%	56%	56%	48%	31%	25%	13%	--
0	--	88%	62%	67%	76%	46%	59%	44%	25%	17%	--
-0.46	--	86%	85%	70%	72%	53%	41%	38%	20%	8%	--
-0.92	--	89%	87%	64%	59%	45%	48%	23%	34%	19%	--
-1.38	--	85%	80%	57%	75%	57%	45%	28%	26%	15%	--
-1.84	--	87%	66%	80%	54%	54%	33%	32%	30%	13%	--
-2.3	--	88%	77%	64%	66%	25%	35%	27%	22%	14%	--



100% Er

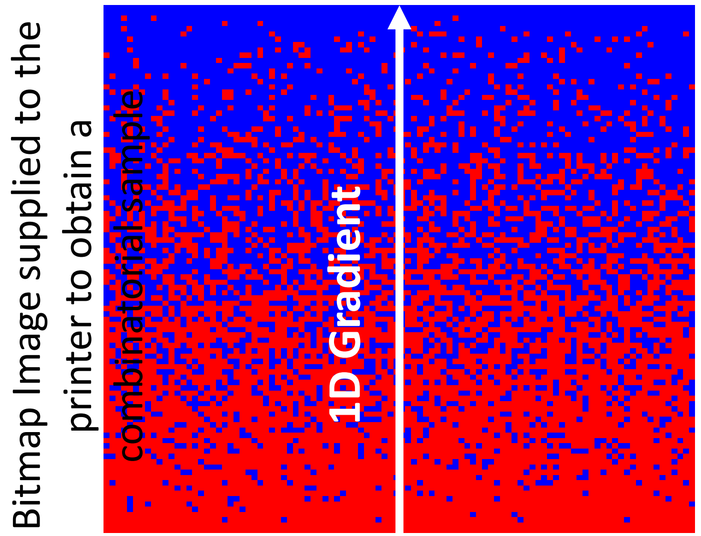
Scanning Hall Probe Microscopy

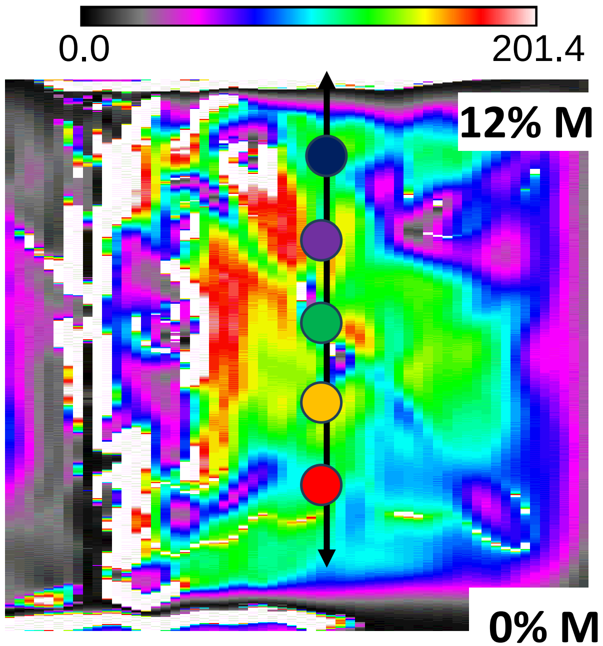


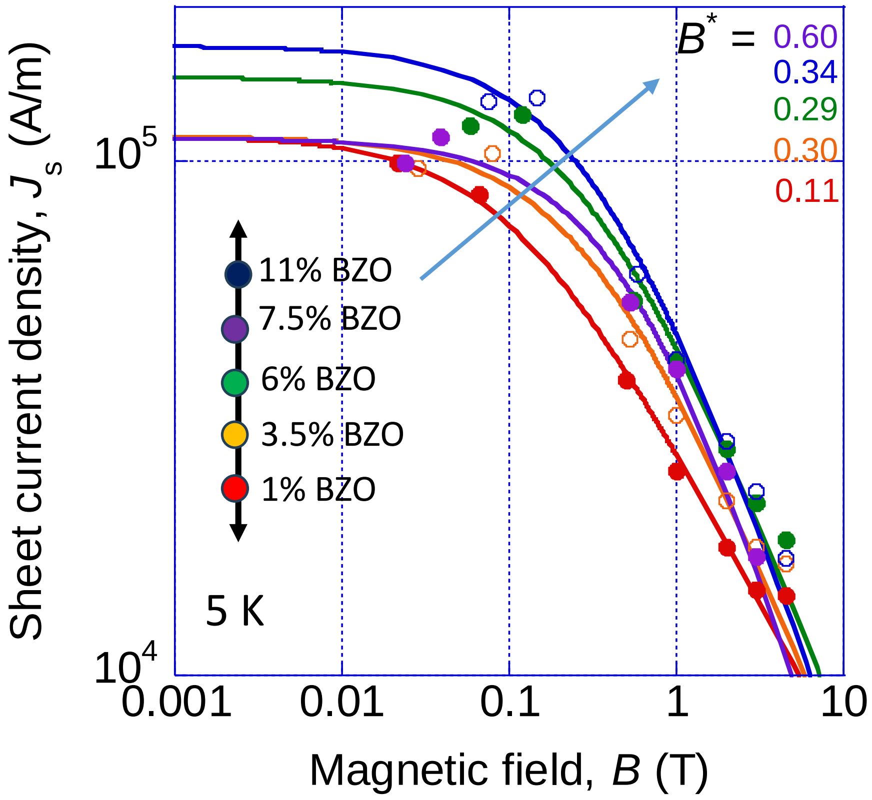
Towards Machine Learning strategies for J_c prediction

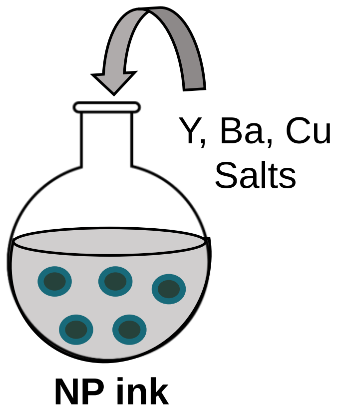
High Throughput Experimentation using Combinatorial Compositional gradients: Nanocomposites (work in progress)

Collab. Z. Wu, T. Kiss

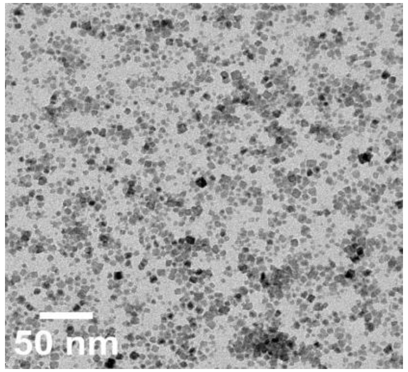








$BaMO_3$ (M= Zr, Hf)



(Patent EP22382741)

Open opportunities towards gradients with different nanoparticles composition and size

$$J_s(B,x,y) = J_{s0}(x,y) * \left(1 + \frac{B}{B^*(x,y)}\right)^a$$

❺ Vortex Physics in TLAG Nanocomposites

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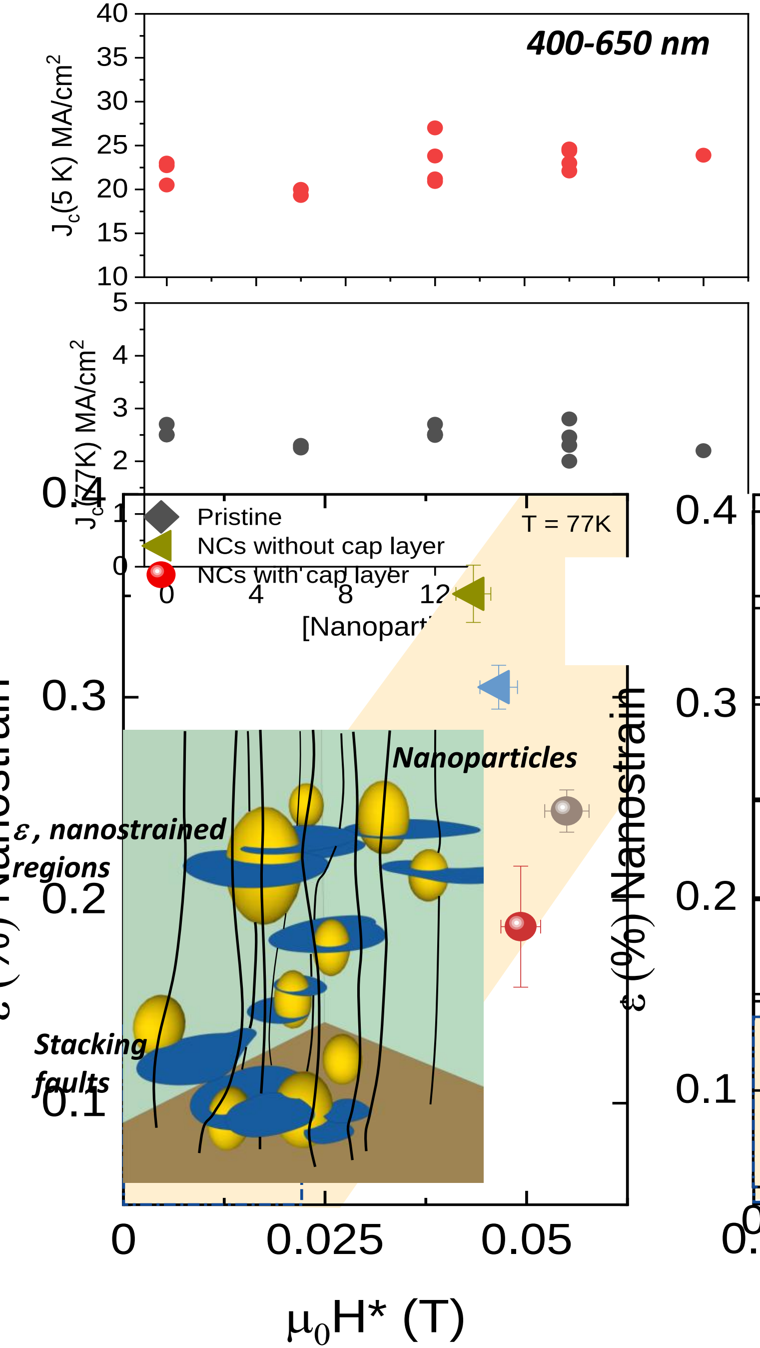
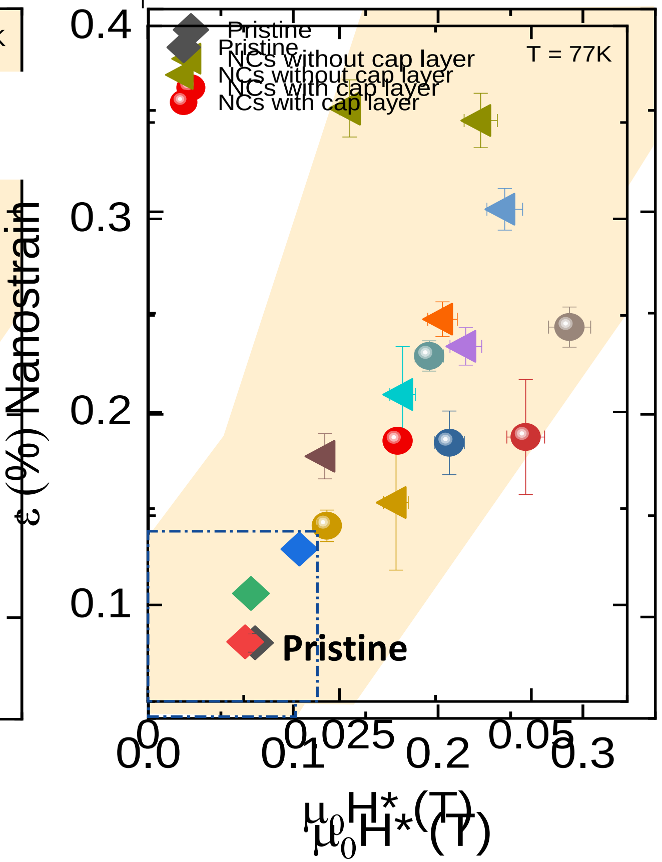
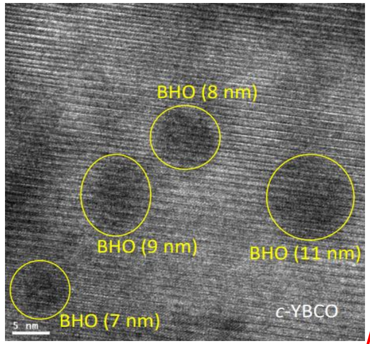
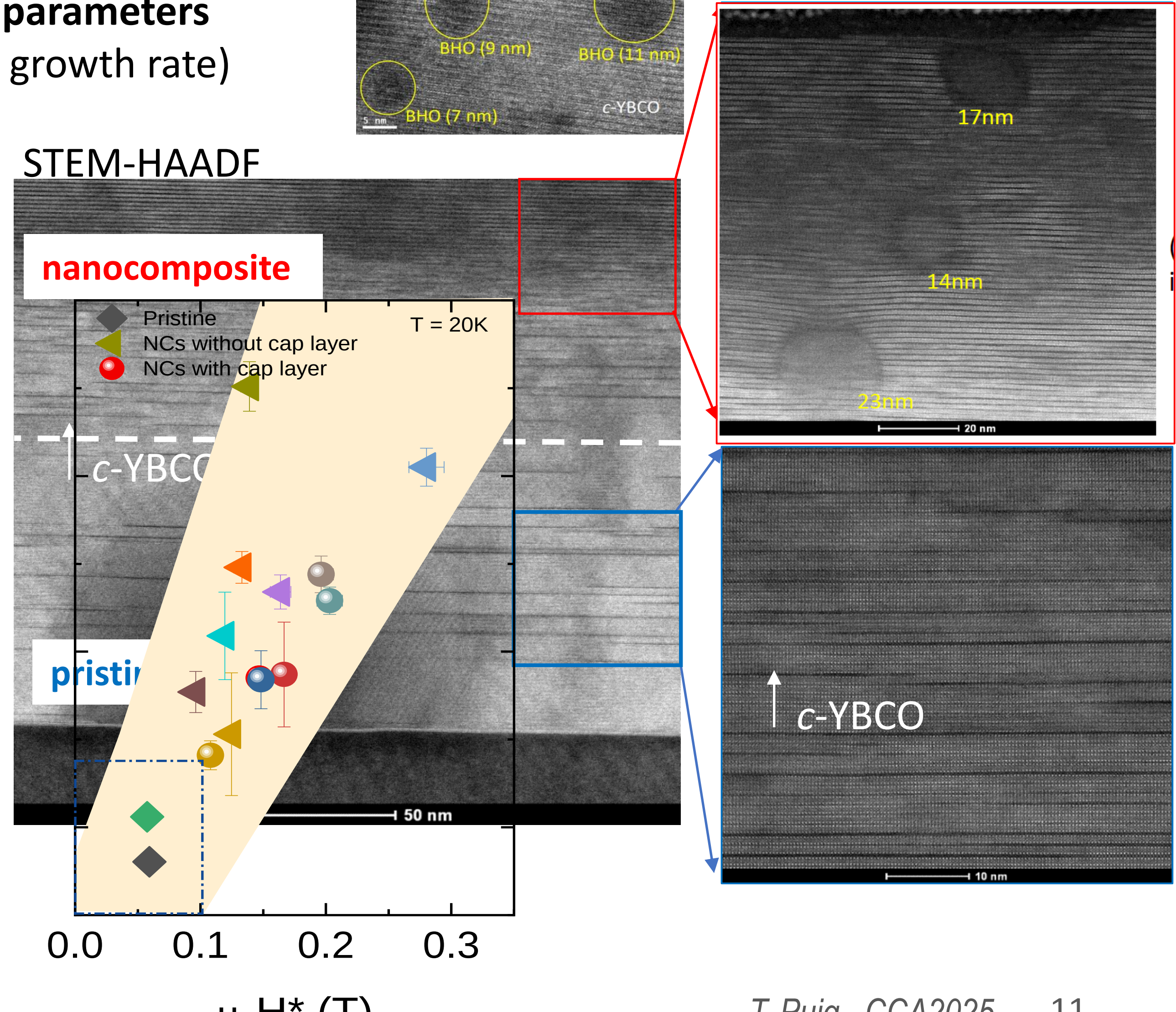
Tuning of vortex pinning by nanostrain and nanoparticles depend on process parameters
(i.e. supersaturation, growth rate)



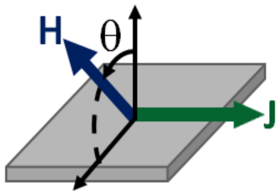


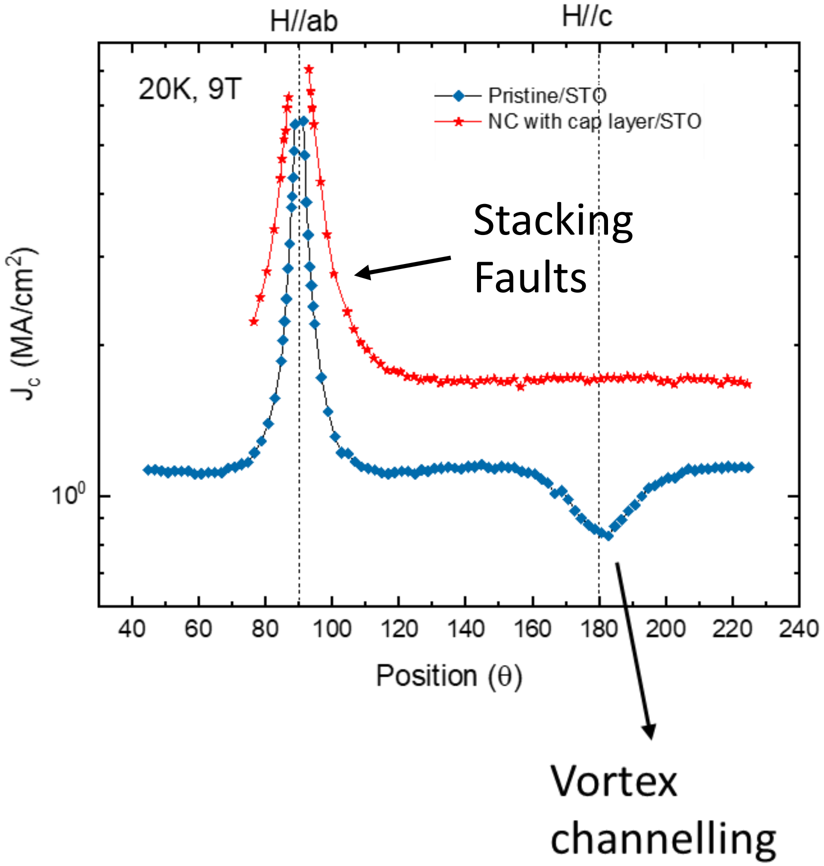


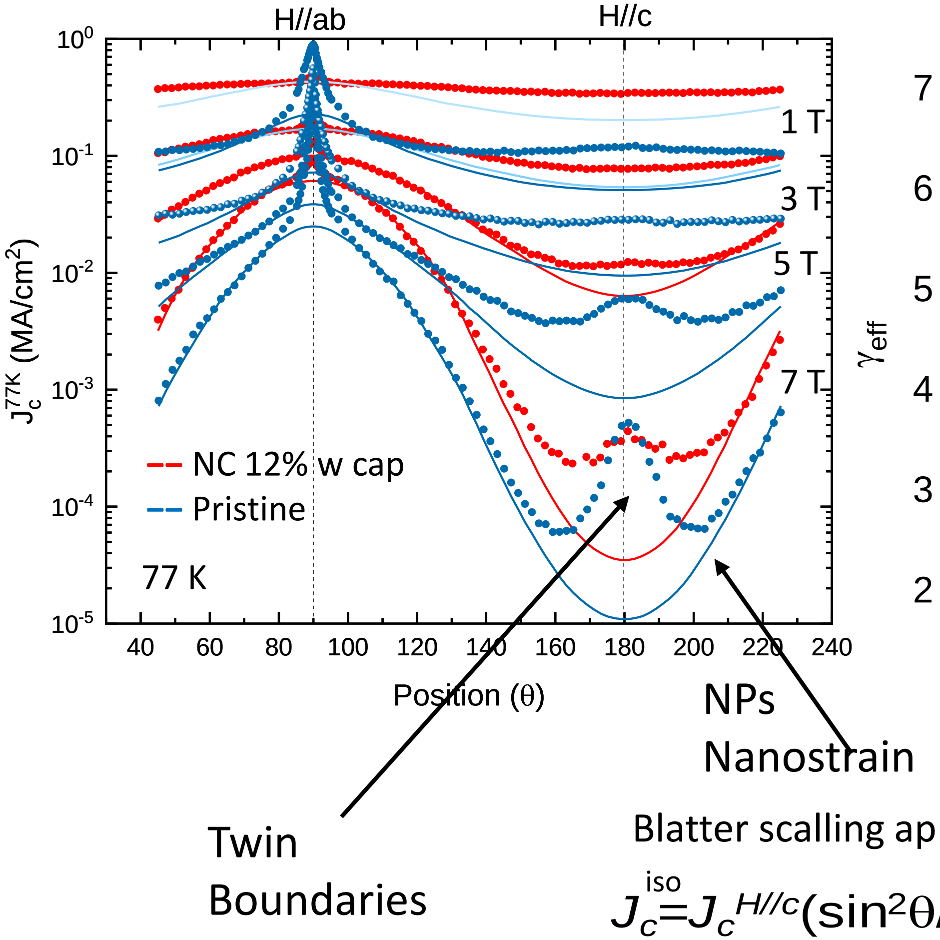


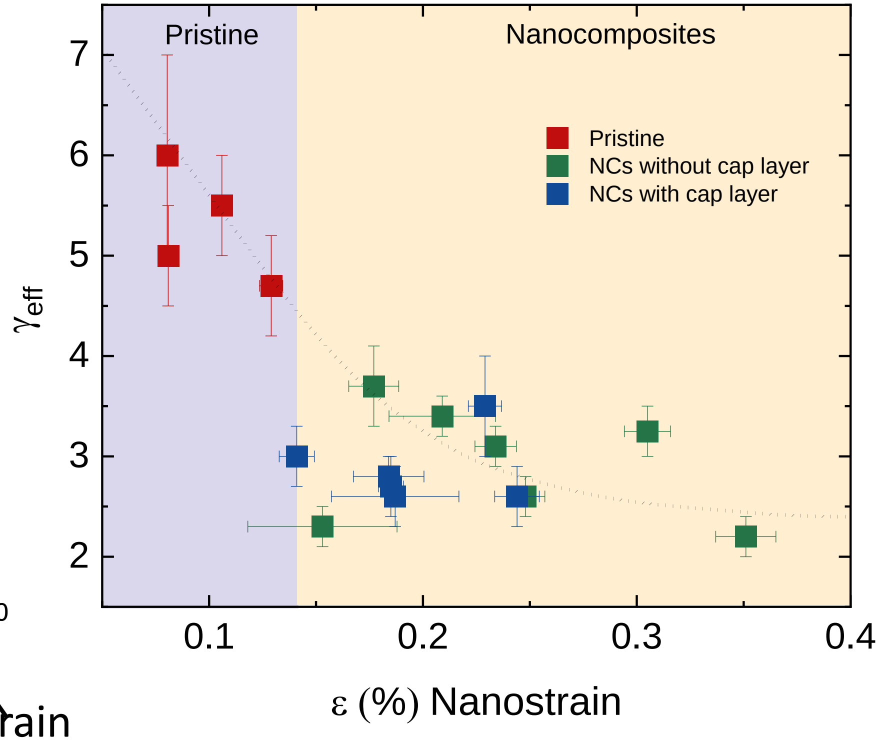
T. Puig et al, Nat Rev Phys (2024)

Vortex Pinning anisotropy in TLAG films









Blatter scalling approach

$$J_c^{iso} = J_c^{H//c}(\sin^2\theta/\gamma_{eff}^2 + \cos^2\theta)$$

Nanocomposites decrease vortex dissipation and induce less anisotropy

Vortex behaviour depends on T, H, θ and process parameters

F. Valles et al, Comm Mat 3 (2022)

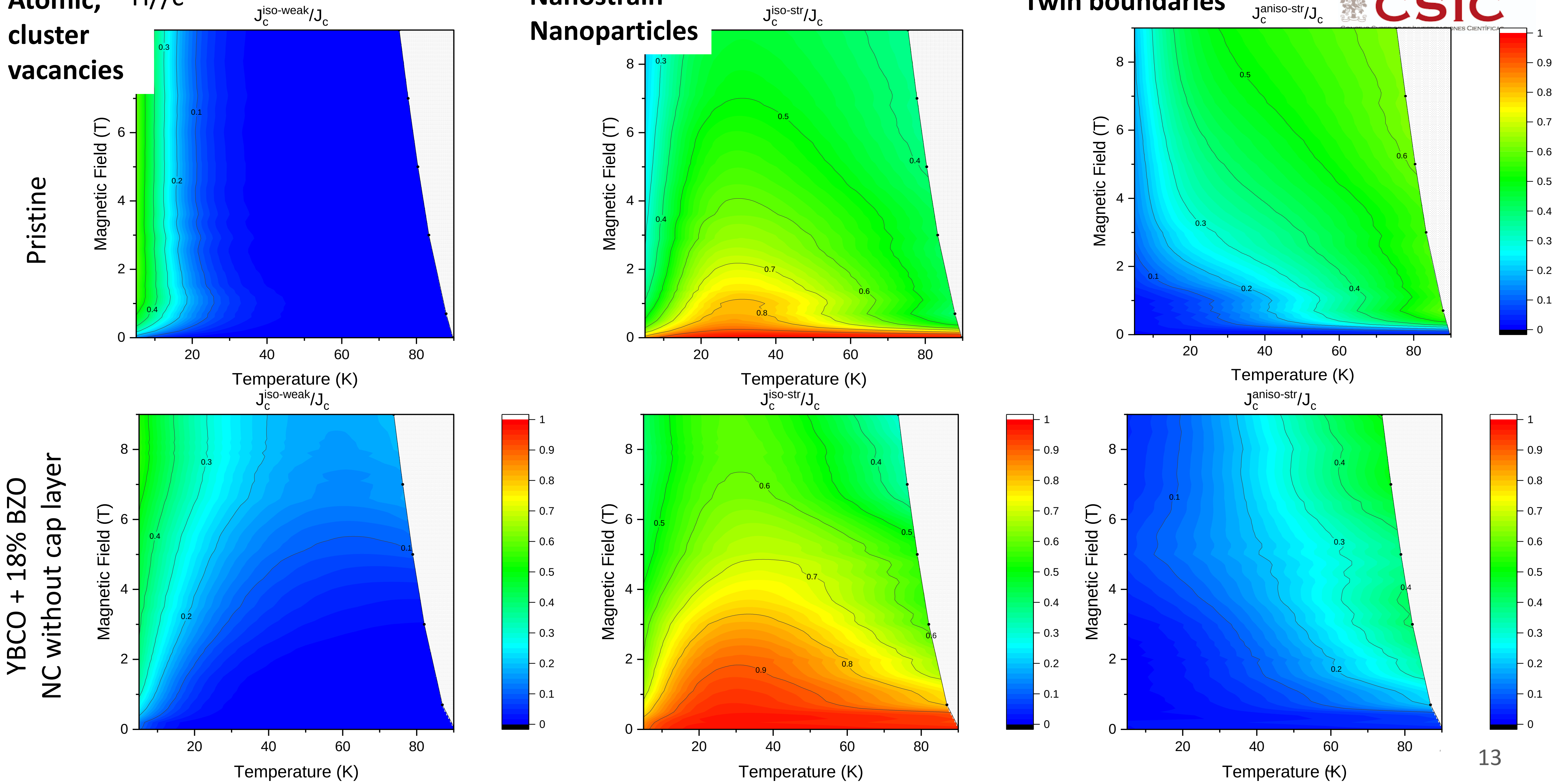
TLAG Vortex Pinning diagrams

$J_c(T) = J_c^{iso-wk}(T) + J_c^{iso-str}(T) + J_c^{aniso-str}(T)$

Atomic, cluster vacancies H//c

Nanostrain Nanoparticles

Twin boundaries



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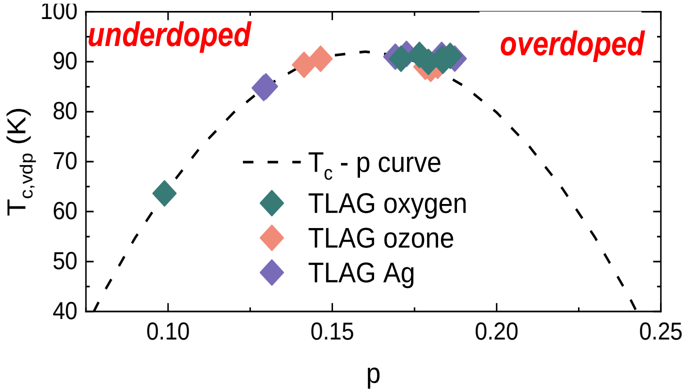
The overdoped state: Opportunity to increase pinning efficiency



Study of different oxygenation methods

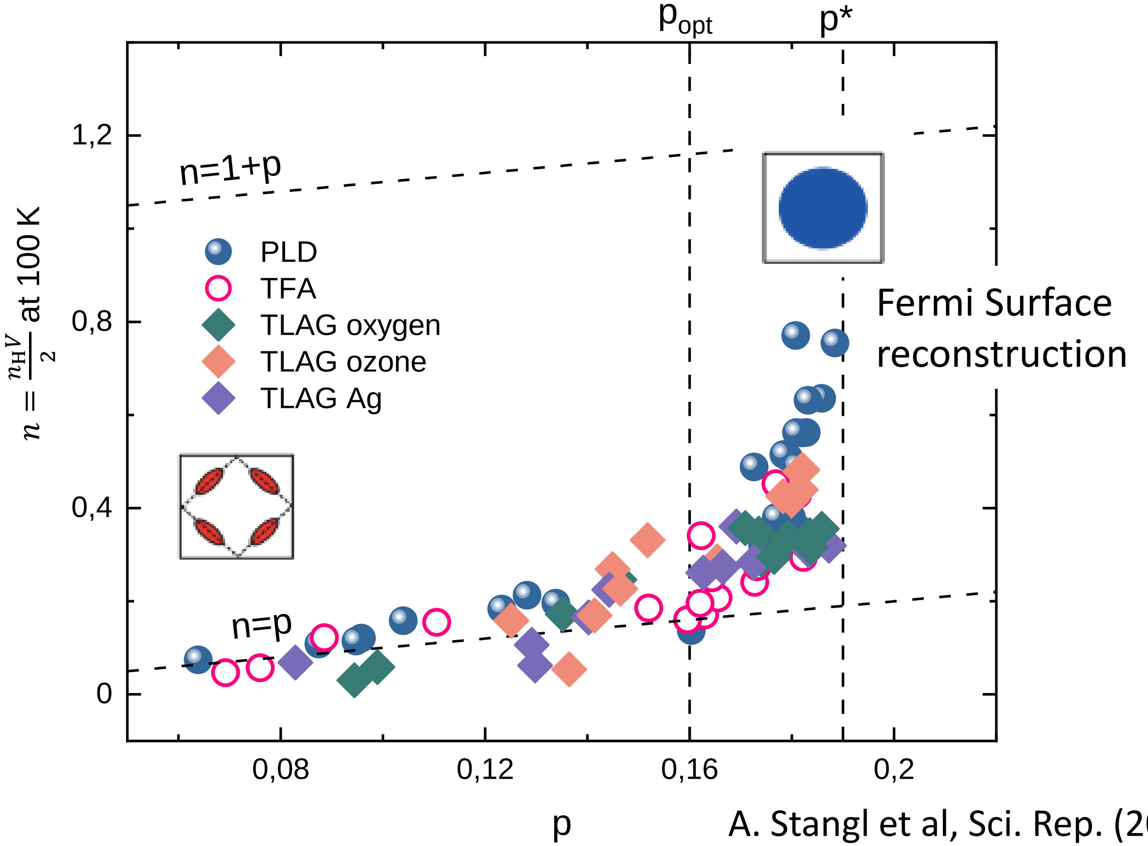
where charge carrier density and condensation energy increases

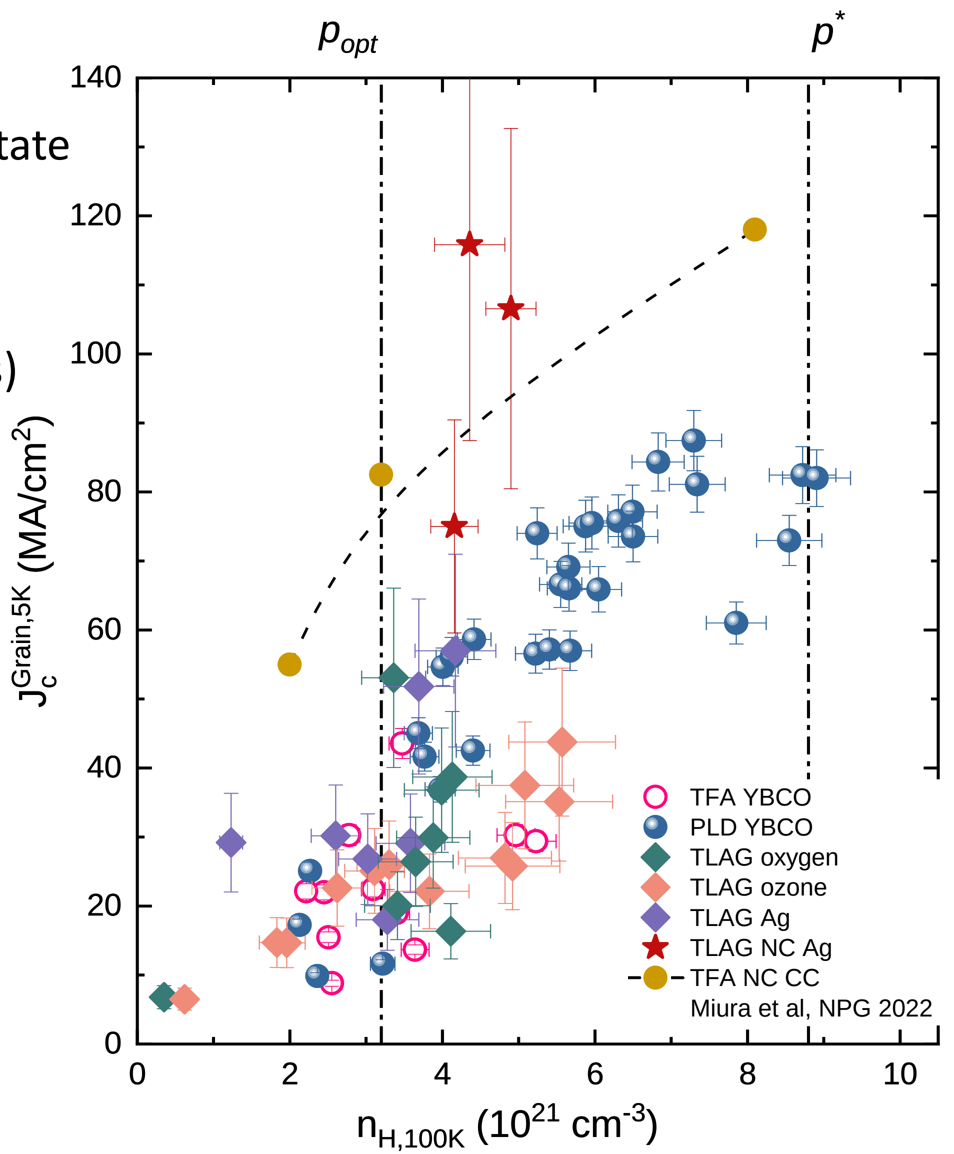
$$n_H \propto E_c \propto f_p$$

Strong increase of J_c in overdoped state

$$\lambda\downarrow, \xi\downarrow \rightarrow E_c \uparrow$$

(Higher efficiency of pinning defects also for nanocomposites)





A. Stangl et al, Sci. Rep. (2021), X. Obradors et al, Nature Mater. N&V (2024)

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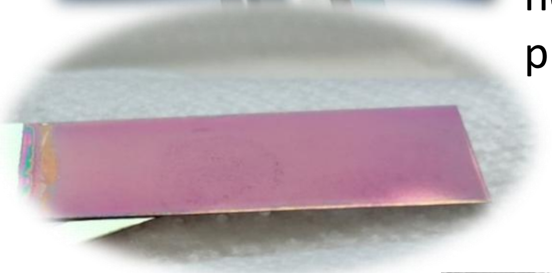
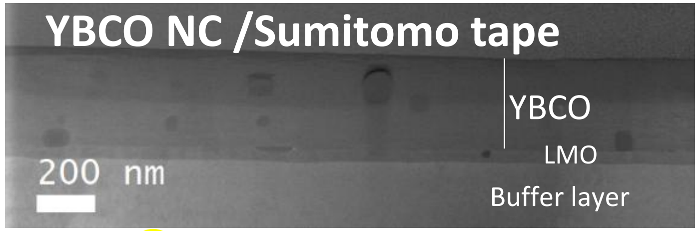
❻ Towards wide area TLAG Coated Conductors

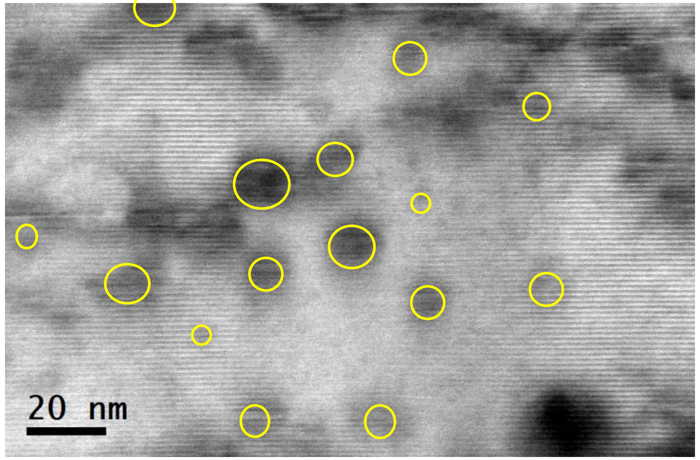
SUMITOMO ELECTRIC · ICMAB INSTITUT DE CIÈNCIA DE MATERIALS DE BARCELONA · EXCELENCIA SEVERO OCHOA · SuNAM Zero to Infinity · CSIC CONSEJO SUPERIOR DE INVESTIGACIONES CIENTÍFICAS

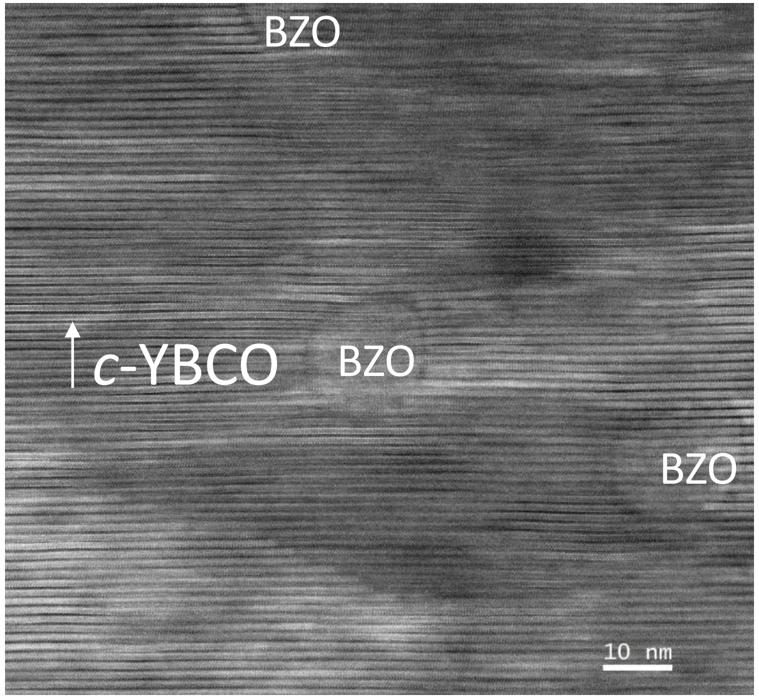


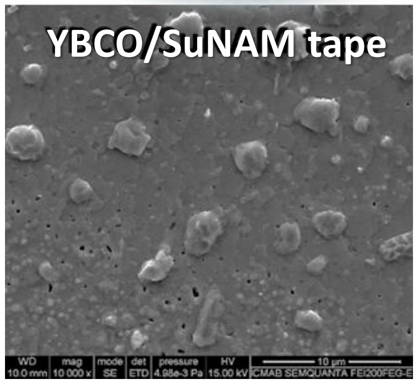
Colloidal Ink
scaled ~ liter

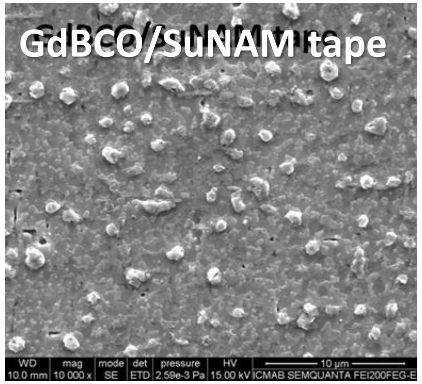
Slot die
now with 40 mm-width printhead capabilities

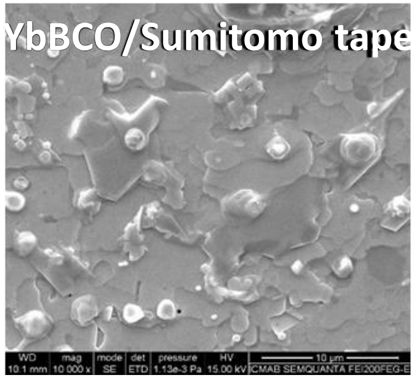












Microstructure is reproduced in 250-750 nm CC
High superconducting properties: J_c(77K) =1.7-2 MA/cm^2

Construction of a furnace for 40 mm-wide tape is on going

Conclusions and outlook

- TLAG is a high-throughput process, very versatile and with a large processing window that can support large volume CC fabrication while reducing cost/performance ratio
- In-situ Synchrotron techniques are ideal to underpin the TLAG mechanisms
- High Throughput Experimentation with combinatorial gradients and AI should accelerate the selection of compositions and conditions
- Vortex pinning in the overdoped state is the future to achieve higher efficiency due to the increase in condensation energy in a robust way
- TLAG should be a large area processing method that adapts to the needs of increased production as well as opens to new applications