Impact of the High Growth Rates on microstructure and vortex pinning of Transient Liquid Asssisted Growth (TLAG) coated conductors

Teresa Puig[1]

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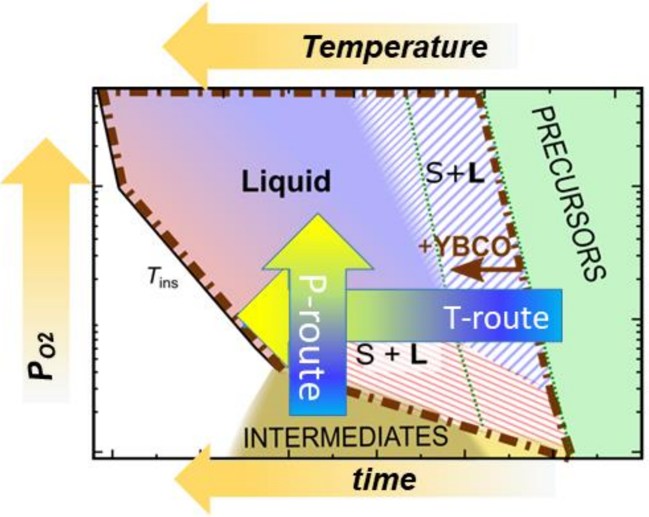
[3] GRMT, Department of Physics, University of Girona, Spain
[4] Diffabs beamline, Soleil Synchrotron, Paris, France
[5] NCD-Sweet beamline, ALBA Synchrotron, Barcelona, Spain



Work is supported by

IEEE-CSC, ESAS and CSSJ SUPERCONDUCTIVITY NEWS FORUM (global edition), Issue No. 57, Oct 2024. Presentation given at ASC 2024, Sept 2024, Salt Lake City, Utah, USA.

Vortex Pinning Centers in REBCO films

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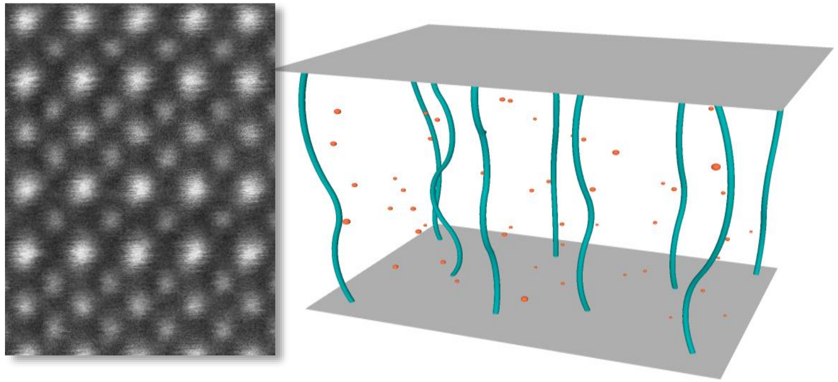
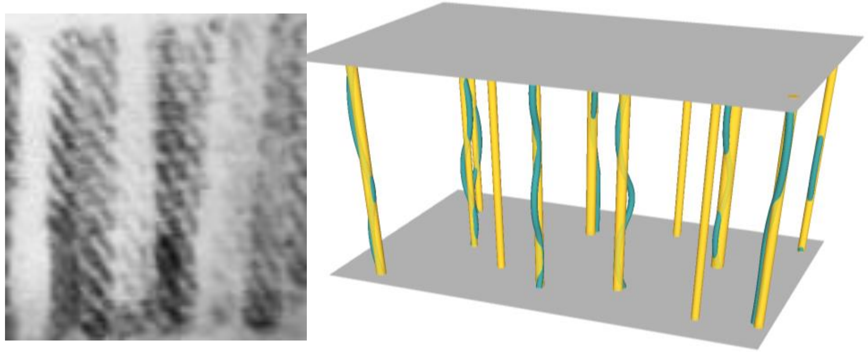
Great variety of nanometric pinning centers has facilitated nanoengineering as a path towards improving vortex pinning

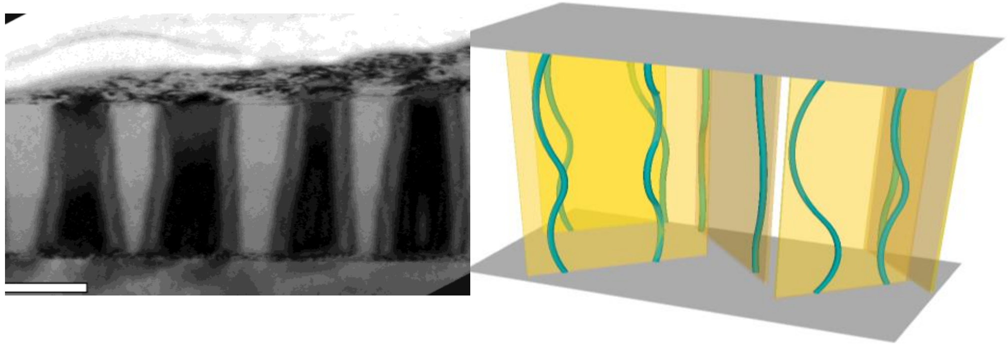
0D-PC: Oxygen vacancies, element substitutions, **point defects**



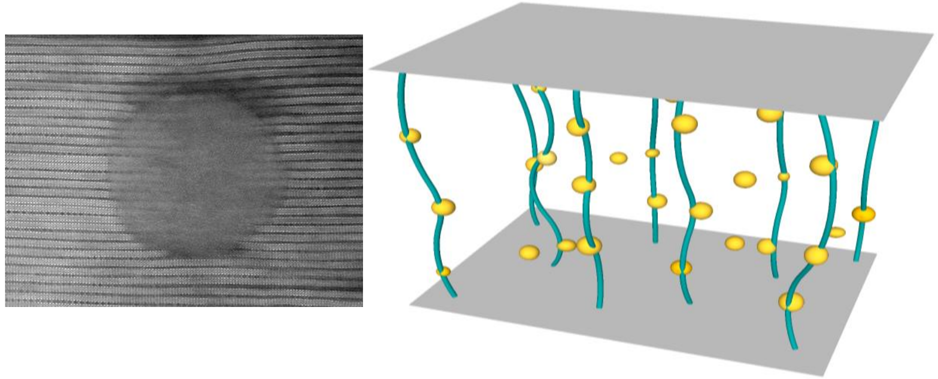
1D-PC: Dislocations, **nanorods**



2D-PC: twin boundaries, stacking faults, **planar defects**

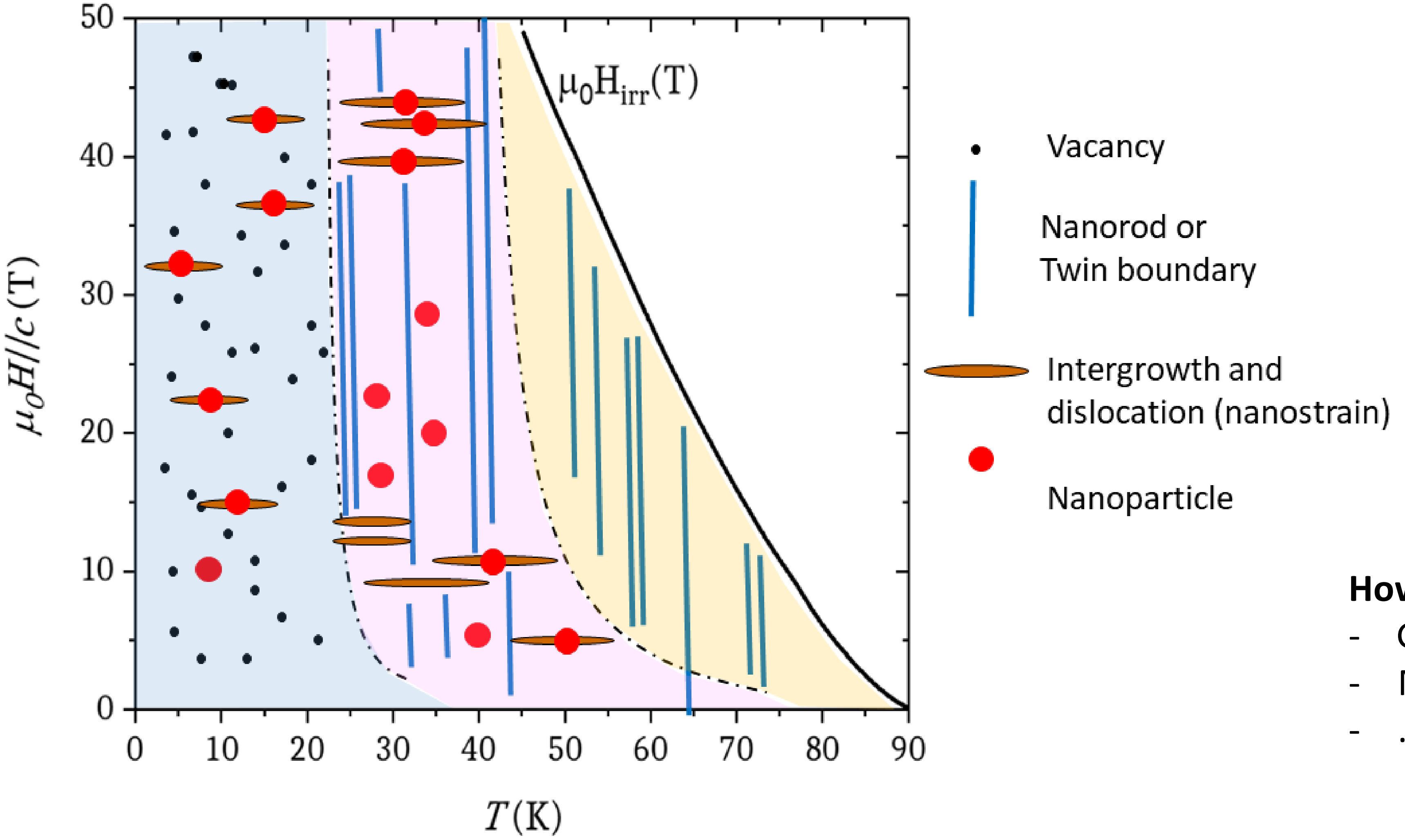


3D-PC: **Nanoparticles**, local strain



A. Llordes, Nat Mat (2012), A. Palau et al SUST (2018), F. Valles et al, Comm Mat (2022)

Correlation between microstructure and vortex pining



- 1D and 2D defects are more effective at high temperatures
- 0D (and 3D) are more effective at low (mid) temperatures

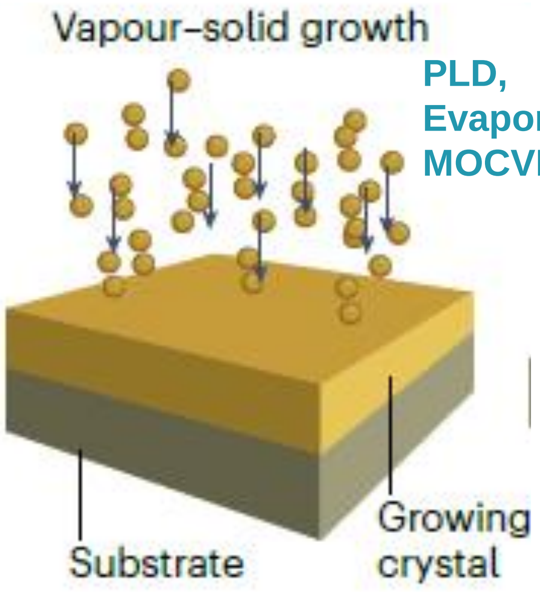
How can high growth rates affect?

- Generating disorder
- Misaligning vertical defects
- …

T. Puig et al, Nat Rev Phys (2024), F. Valles et al, Comm Mat (2022)

Epitaxial REBCO film Growth methods

Supersaturation, σ, is the driving force for crystallization: **σ ∝ G** (growth rate)



PLD, Evaporation, MOCVD

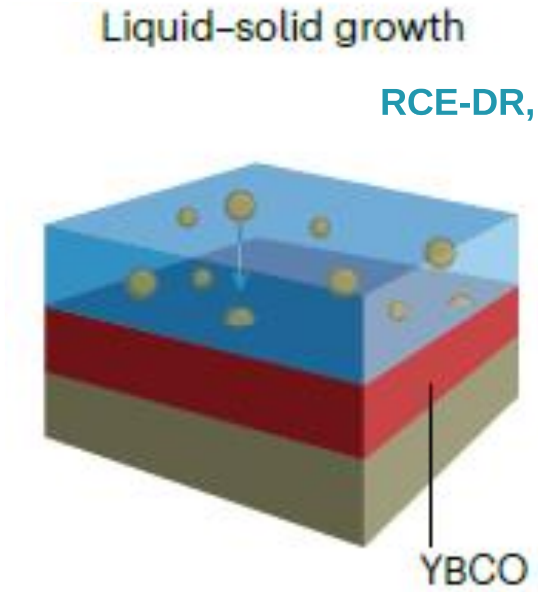
$$\sigma = (P_{ad} - P_{ad,e}) / P_{ad,e}$$

$P_{ad,e}$ = ad-atoms equilibrium pressure at surface growth front
P_{ad} = ad-atoms pressure at surface growth front

TFA-CSD

$$\sigma = f\,(\ln (P_{HF}^2 / P_{H2O}))$$

P_{HF} = HF partial pressure
P_{H2O} = water partial pressure



RCE-DR, TLAG-CSD

$$\sigma = (C_\delta - C_e) / C_e$$

C_e = RE equilibrium concentration in the liquid
C_δ = RE actual concentration

Growth rate: **G= 0.5-25 nm/s** | **G= 0.5-5 nm/s** | **G=10-1000 nm/s**

T. Puig et al, Nat Rev Phys (2024), J. Driscoll et al, Nat Rev Mat (2021)

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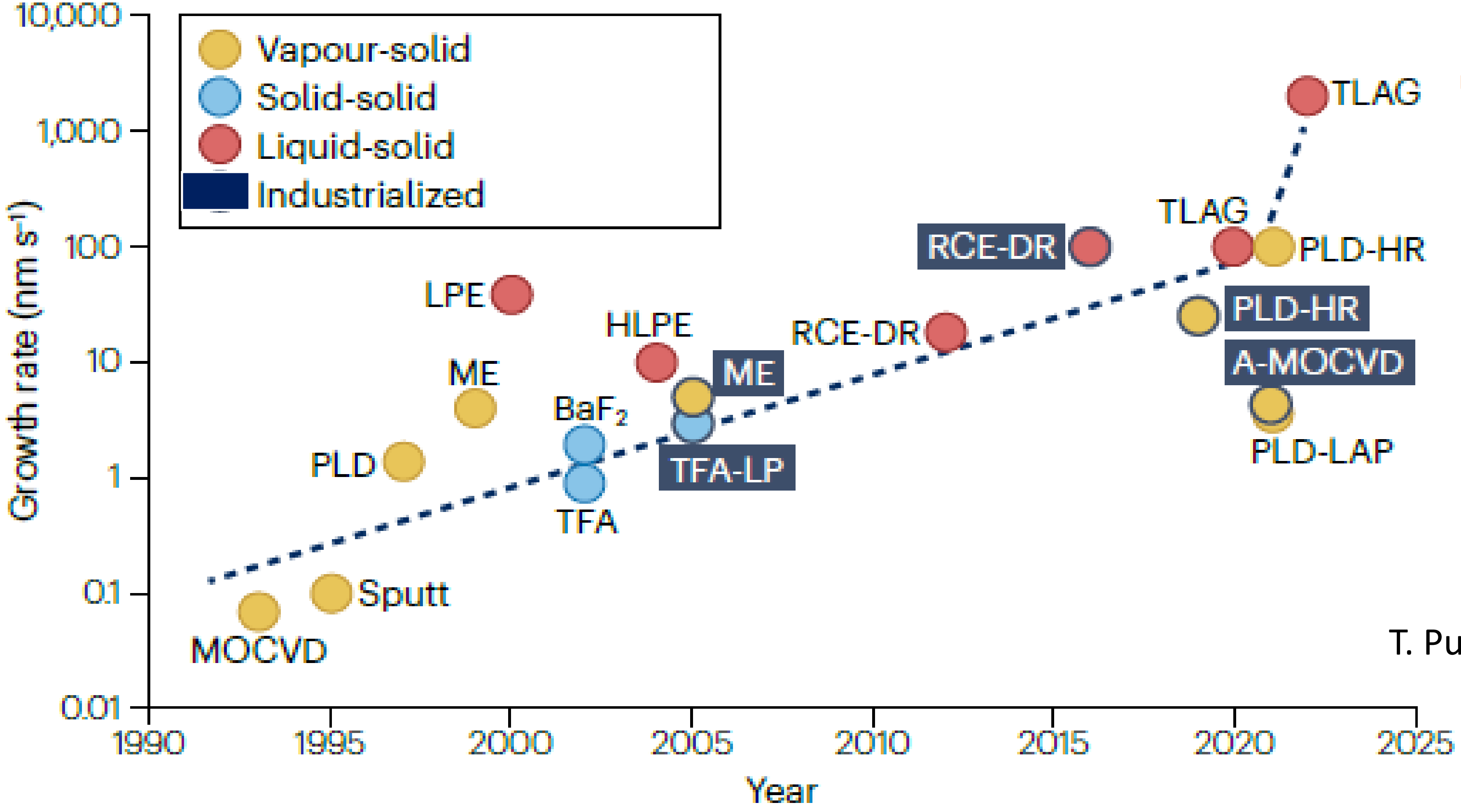
A path towards cost reduction: Reaching high Growth Rate

Figure of merit: $$\frac{Cost}{Perfomance} = \frac{total\ cost\ per\ year}{\boldsymbol{G}\ x\ L\ x\ W\ x\ (I_{c-w}/d)} = \frac{€}{kA\ x\ m}$$

G= growth rate
W= tape width
L= tape length
d=tape thickness



suman SUPERCONDUCTING MATERIALS AND FUNCTIONAL NANOENGINEERED STRUCTURES

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

IEEE-CSC, ESAS and CSSJ SUPERCONDUCTIVITY NEWS FORUM (global edition), Issue No. 57, Oct 2024. Presentation given at ASC 2024, Sept 2024, Salt Lake City, Utah, USA.

High growth rates with nanorods vs nanoparticles

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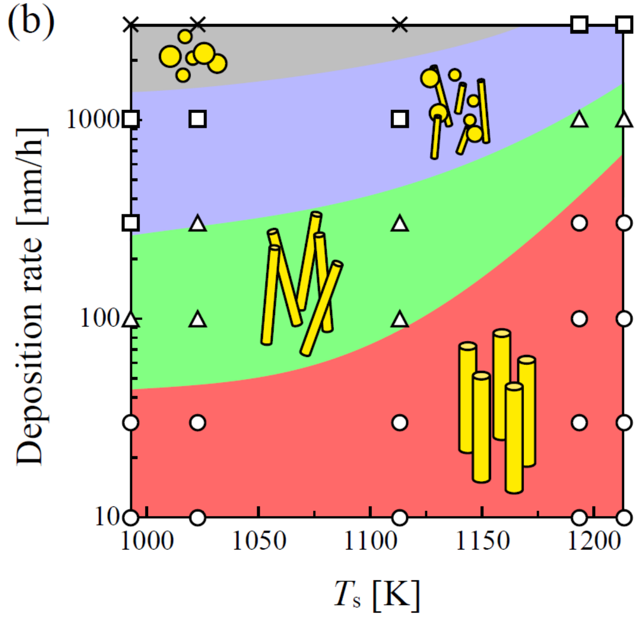
Nanorods by PLD

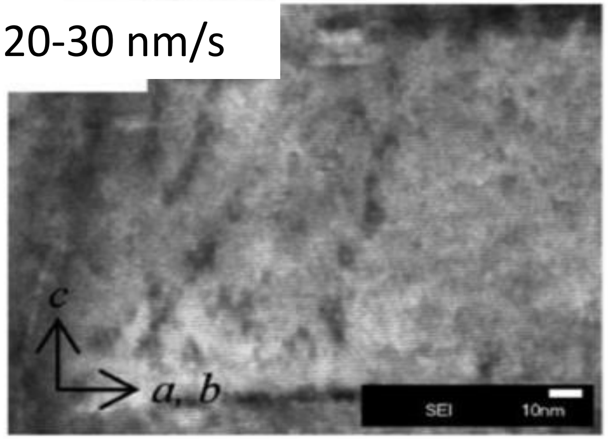
Montecarlo simulations



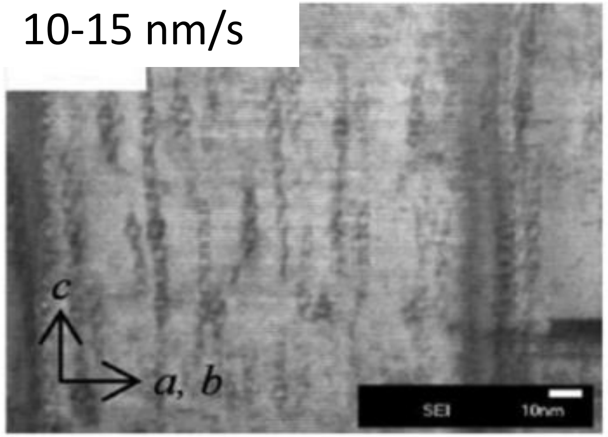
Y Ichino et al J.JAP 56 (2017)

EuBCO + $HfBaO_3$ nanorods

20-30 nm/s



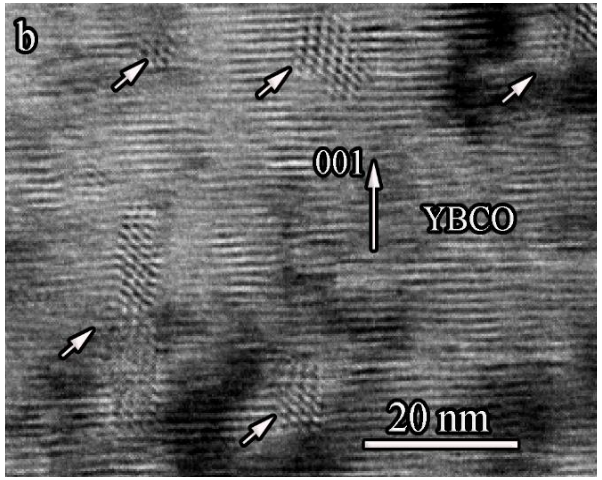
10-15 nm/s



Fujita, S. et al. IEEE TAS 29 (2019)

Aligment and length of nanorods are strongly affected by high growth rates

Nanoparticles by PLD

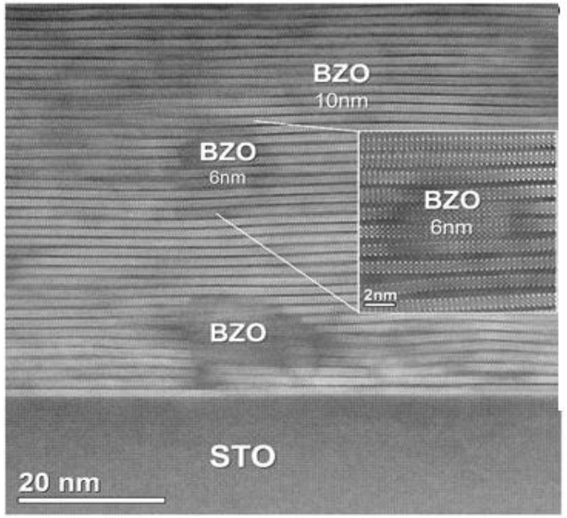


Nanoparticles are less affected

20-40 nm/s

A. Molodyk et al, Sci. Rep 11 (2021)

Nanoparticles by TLAG-CSD



50-2000 nm/s

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

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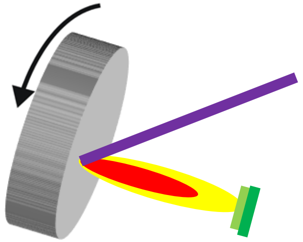
Transient Liquid Assisted Growth (TLAG)

A new high throughput non-equilibrium kinetically controlled growth process

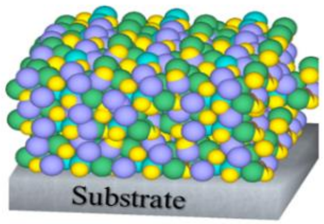
CSD

L. Saltarelli et al, ACS Appl. Mat. & Interf. (2022)
Patent EP22382741

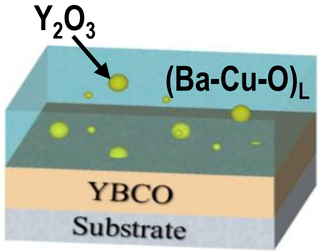


Low Temp PLD

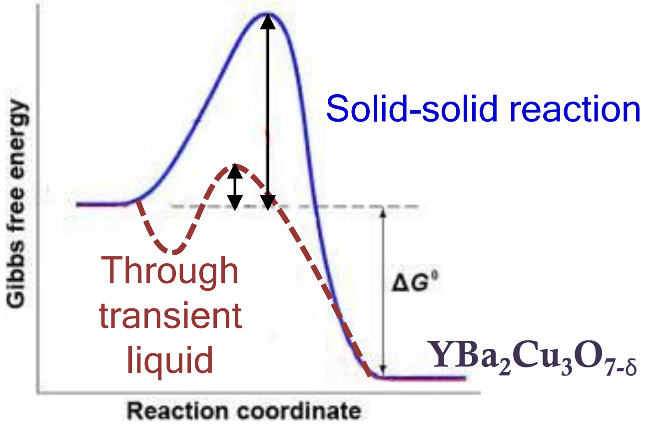
A. Quetalto et al, SUST (2023)



Nanocrystalline precursors



YBCO growth

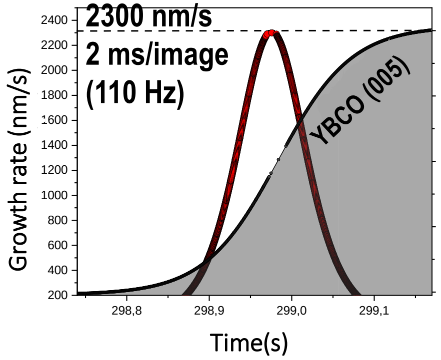


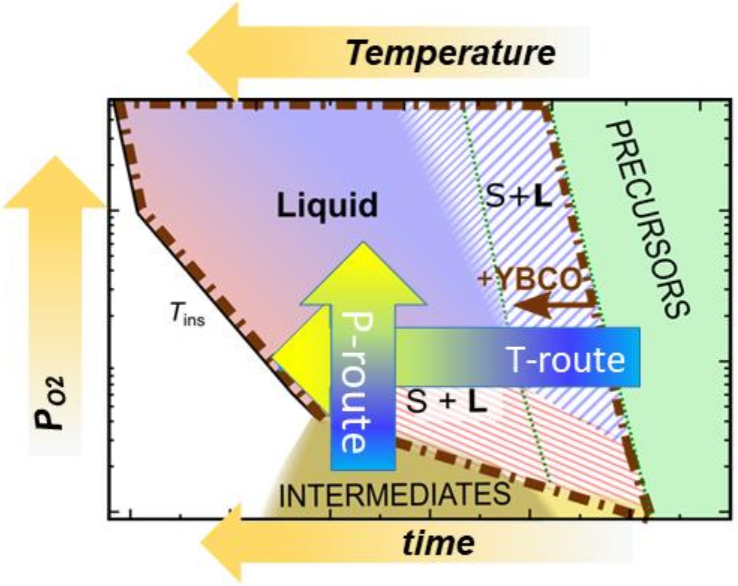
CSD: $BaCO_{3(s)} + CuO_{(s)} + Y_2O_{3(s)}$
(mostly studied)

PLD: $(Ba\text{-}Cu^{I/II}\text{-}O)_s + Cu_2O_{(s)} + Y_2O_{3(s)}$

$\rightarrow (Ba - Cu^{I/II}\text{-}O)_{(l)} + Y_2O_{3(s)} \rightarrow YBa_2Cu_3O_{7-\delta}$

- **High throughput**
- **High growth rate** (2300 nm/s demostrated)
- **High performance** (2-5 MA/cm^2 at 77K)
- **Uses simple reactor**
- **Large area processing**
- **Low cost/performance ratio**





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

REBCO TLAG film nucleation and growth

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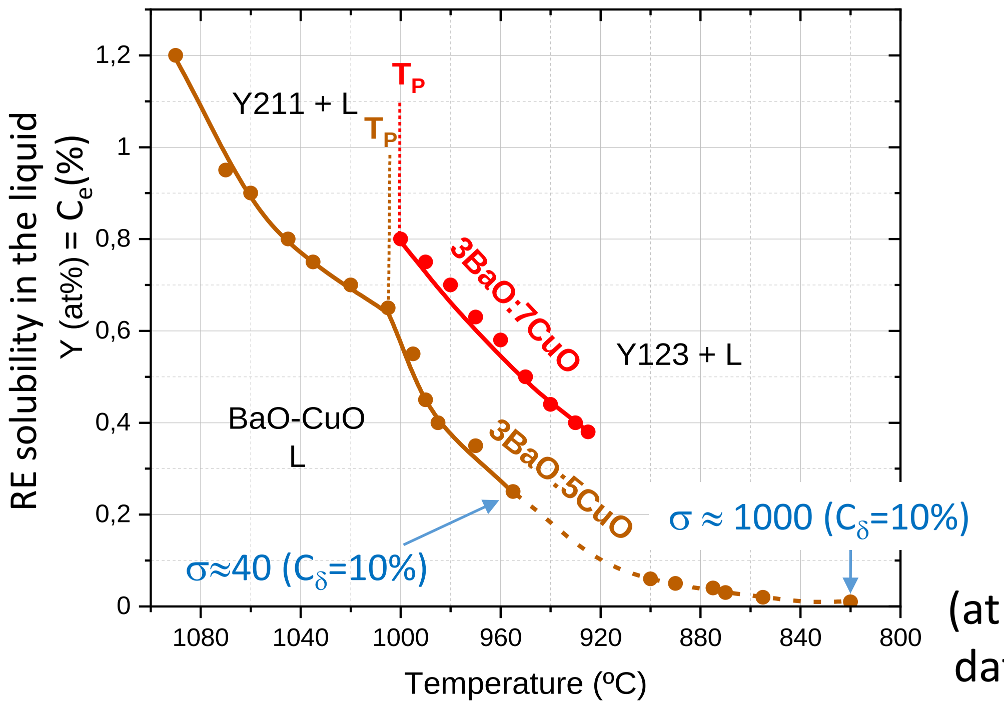
Kinetically governed by RE liquid supersaturation

$$\sigma = (C_\delta - C_e) / C_e$$

C_e = RE equilibrium concentration in the liquid (**thermodynamic parameter** that depends on T, P_{O2}, Ba/Cu liq. composition, RE ion)
C_δ = RE actual concentration (**kinetic parameter** that depends on heating ramp, pressure ramp, …)

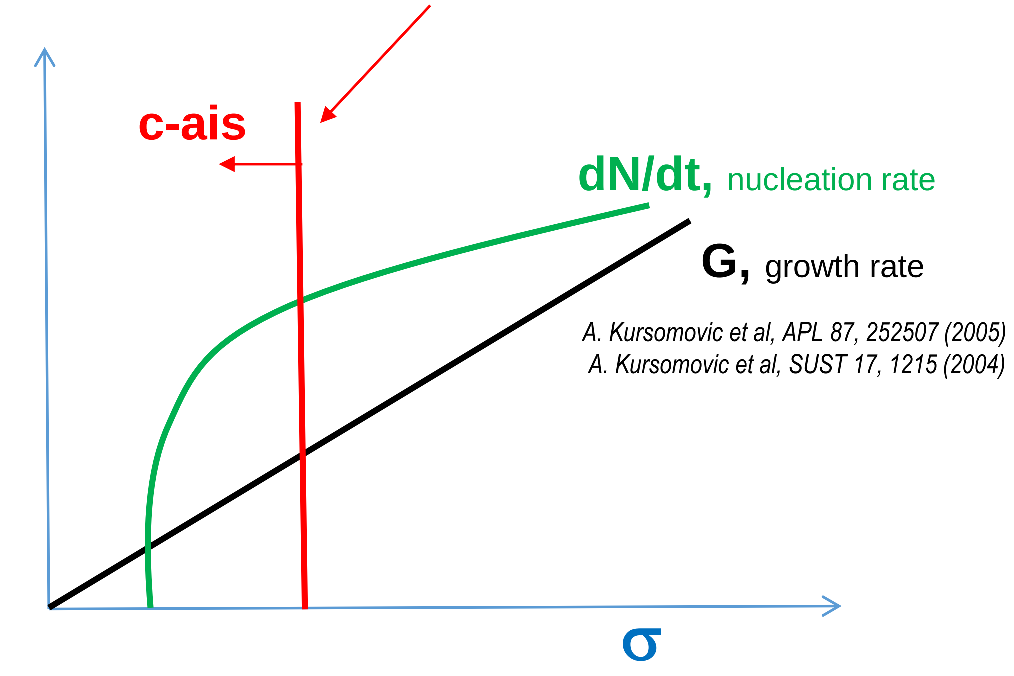
0.2 bar O_2



T	↓	σ	↑
PO_2	↑	σ	↑
Ba/Cu	↑	σ	↑
r_{RE}	↑	σ ??	

(at TLAG conditions data is in progress)

Ichino, K. *IEEE Trans. Applied Supercond.* **13, 2735** (2003)

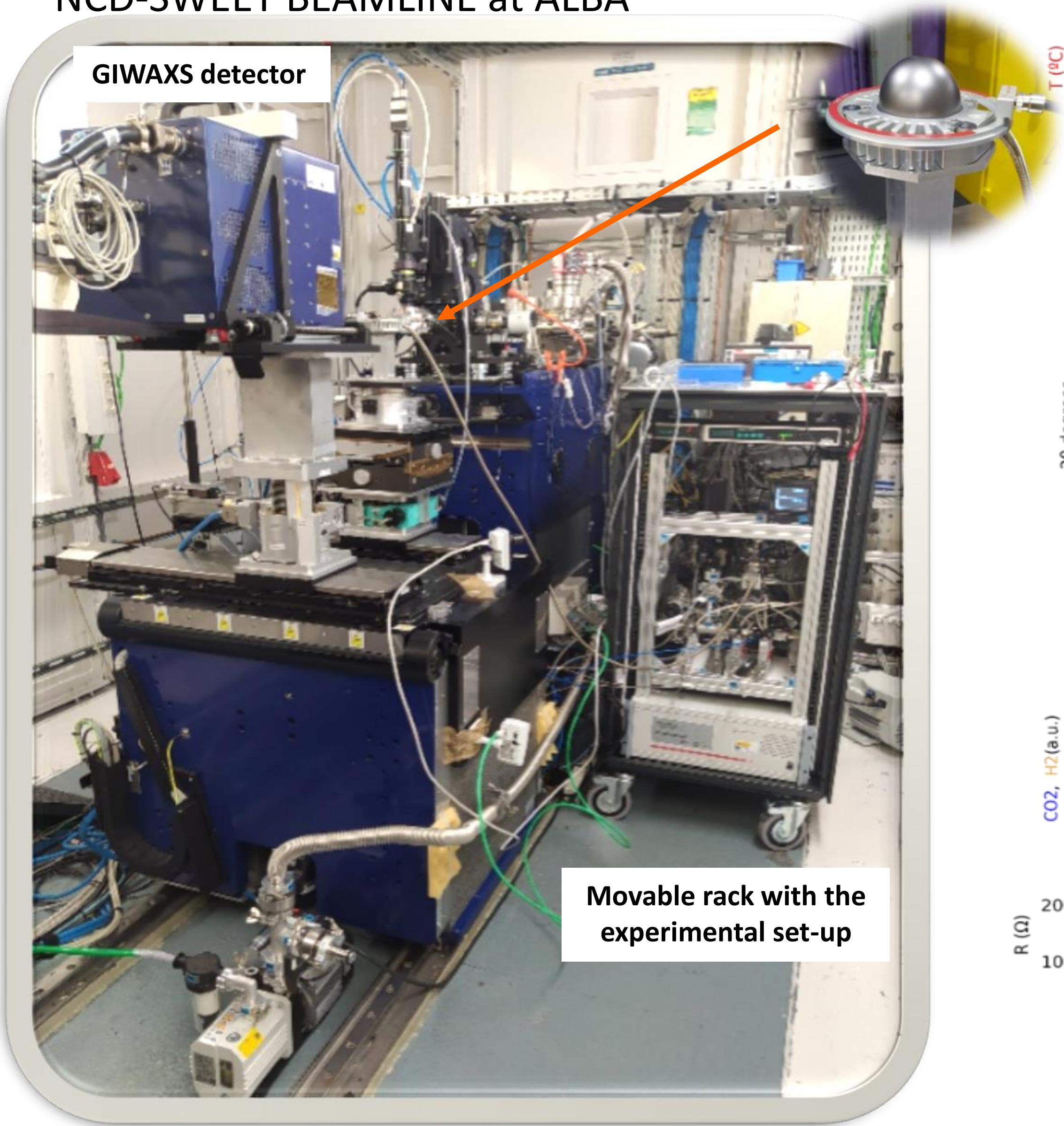


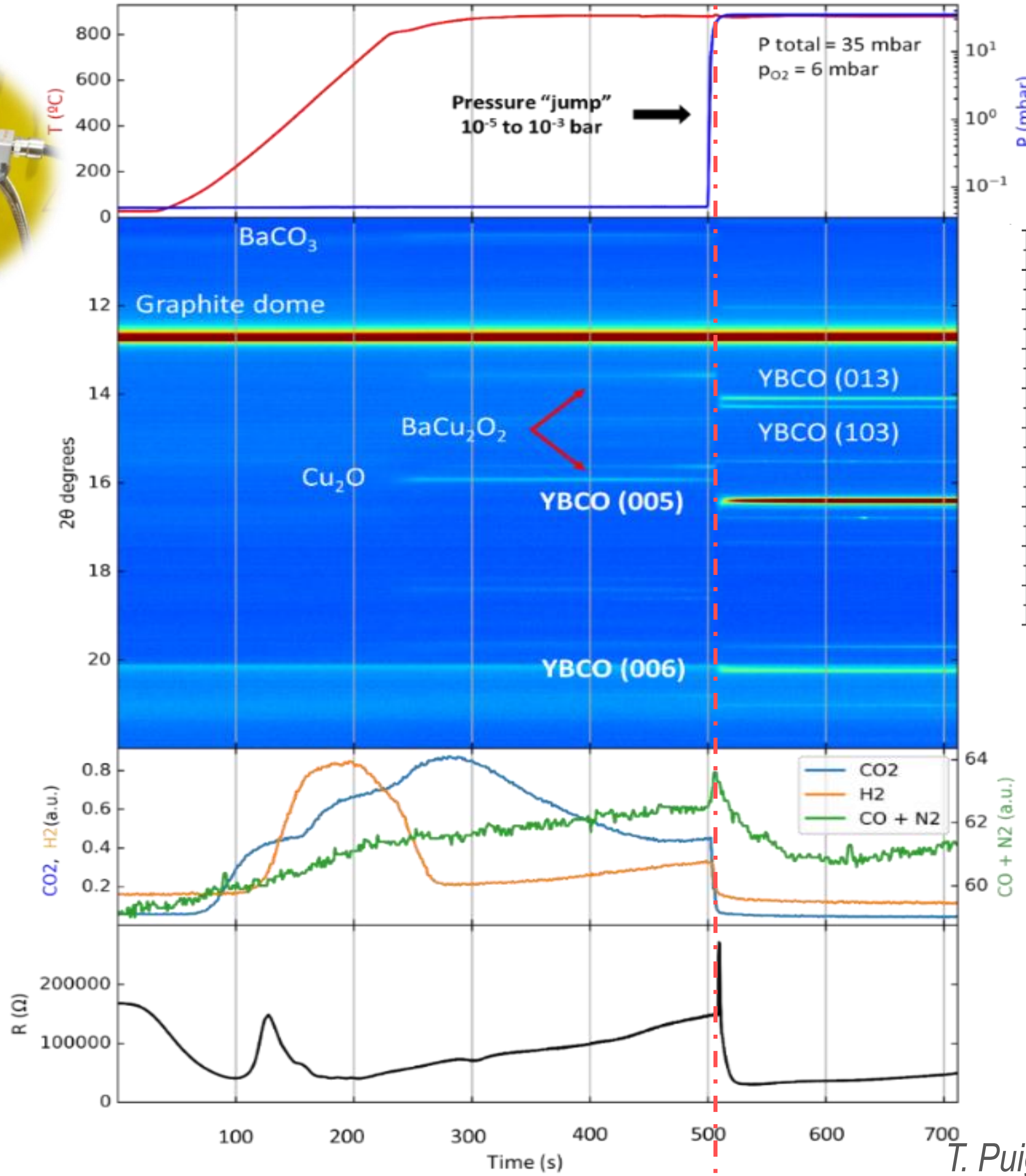
A compromise is needed between ↑σ and epitaxy for each method

Krauns, Shiohara; Z. Phys. B 96, 207-212 (1994) ***(MTG at 0.2 bar O_2)***
Kursumovic, MacManus-Driscoll; Appl Phys Letters 87, 252507 (2005) ***(HLPE at 0.2 bar O_2)***

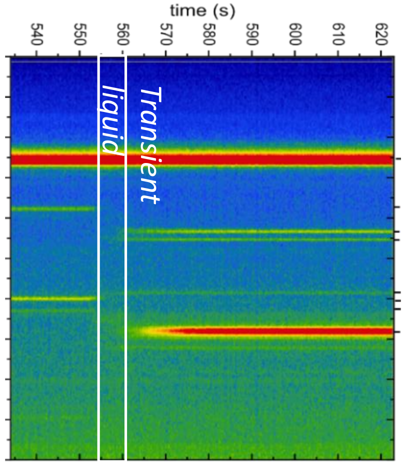
Synchrotron installation

DIFFABS BEAMLINE at Soleil
NCD-SWEET BEAMLINE at ALBA





Experiment conditions



Synchrotron XRD

Mass spectroscopy

Resistivity

suman
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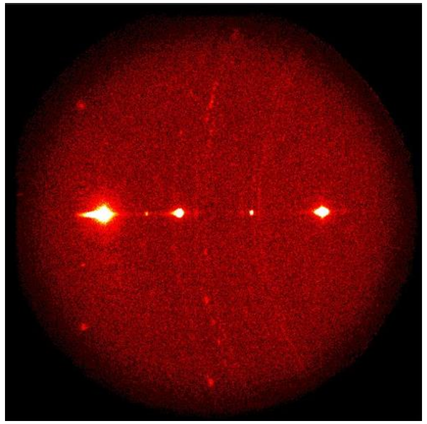
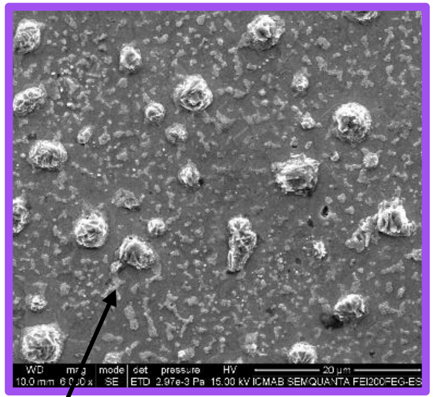
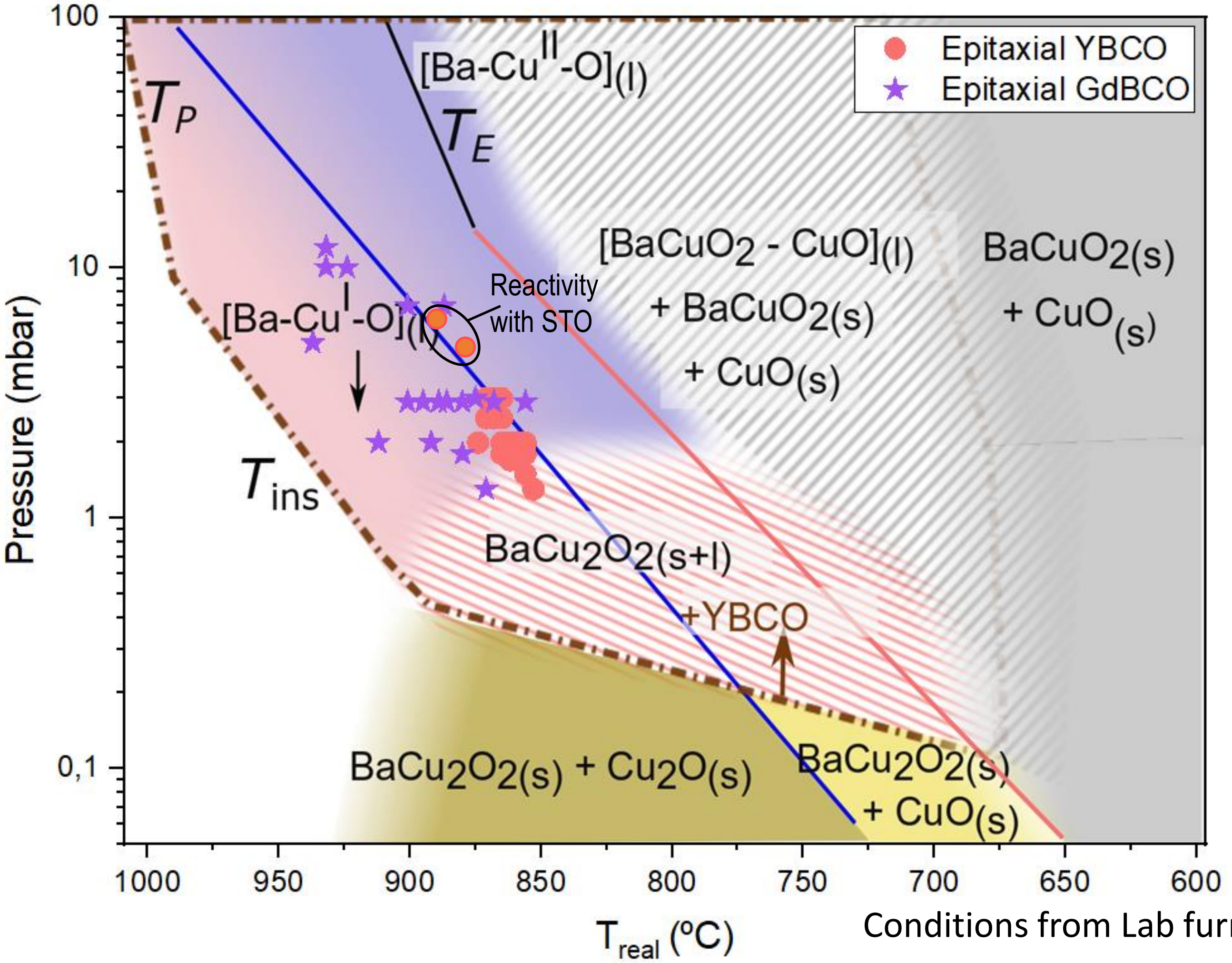
TLAG-CSD: Versatile, large epitaxial window

GdBCO/STO



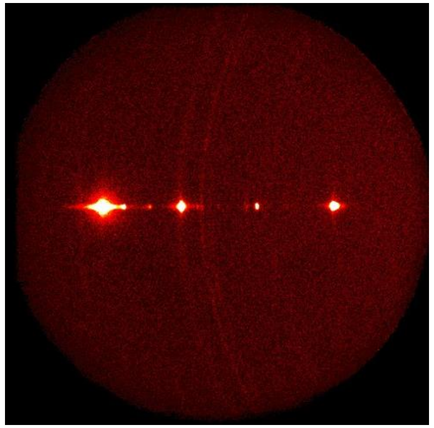


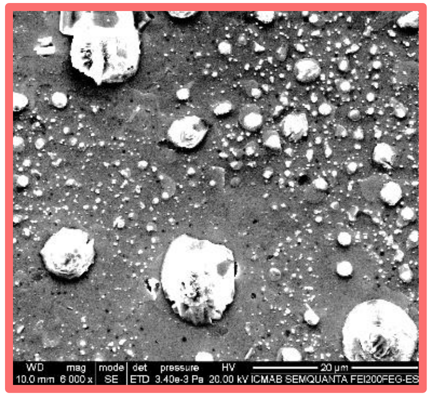
CuO Surface precipitates from the Cu rich-stoichiometry



Conditions from Lab furnace

YBCO/STO





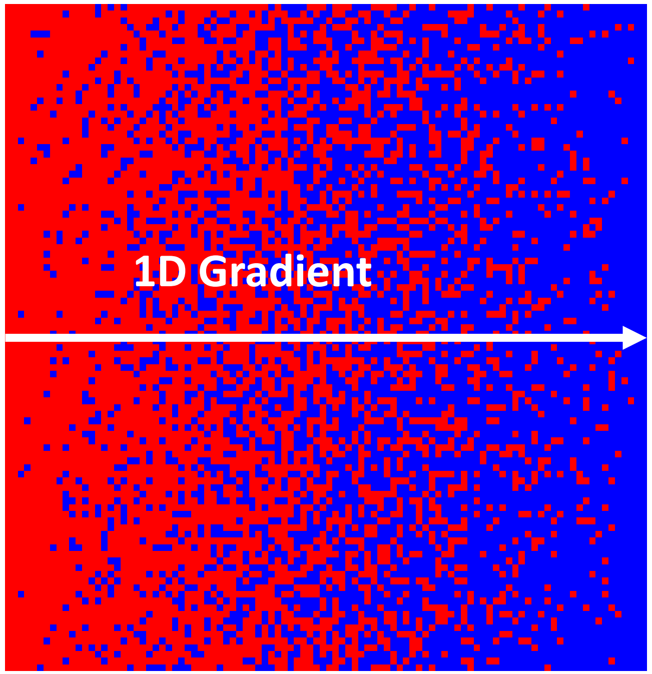
RE solubility defines the supersaturation values, nucleation density, growth rate and epitaxial window

REBCO combinatorial compositional gradients

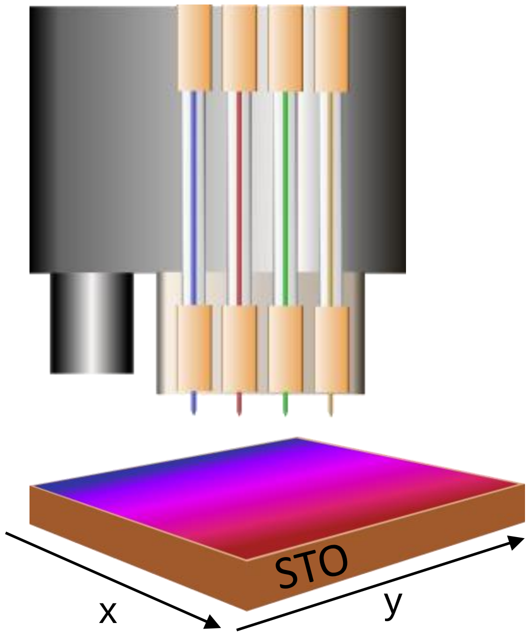
Allows for fast screening of compositions and process parameters

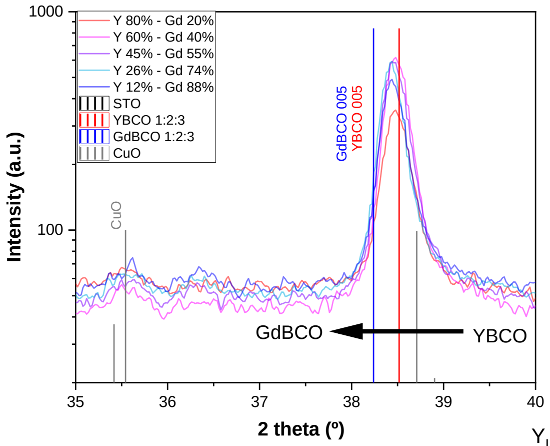
See 5MOr2A-02, C. Pop

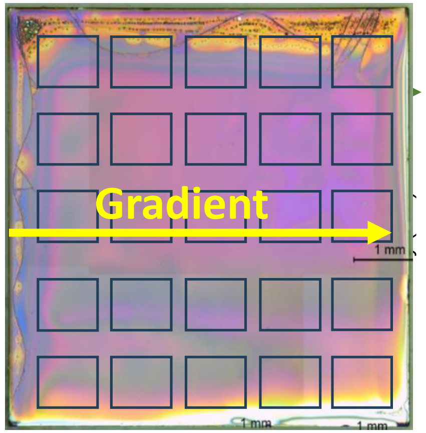
Bitmap Image supplied to the printer to obtain a combinatorial sample

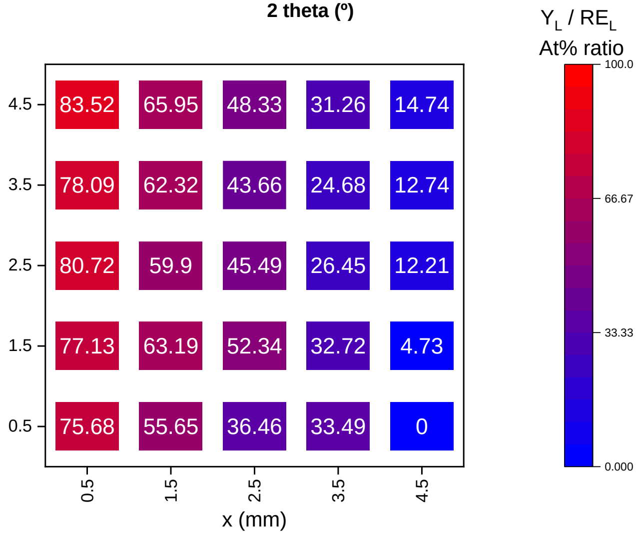


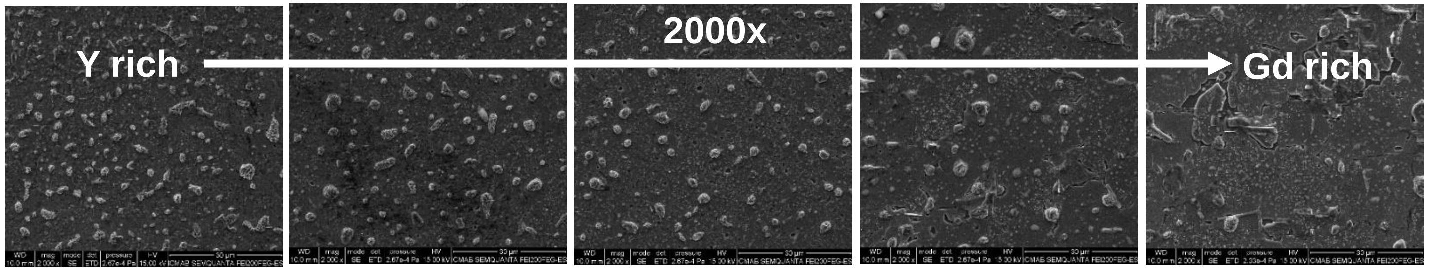
Via inkjet printing





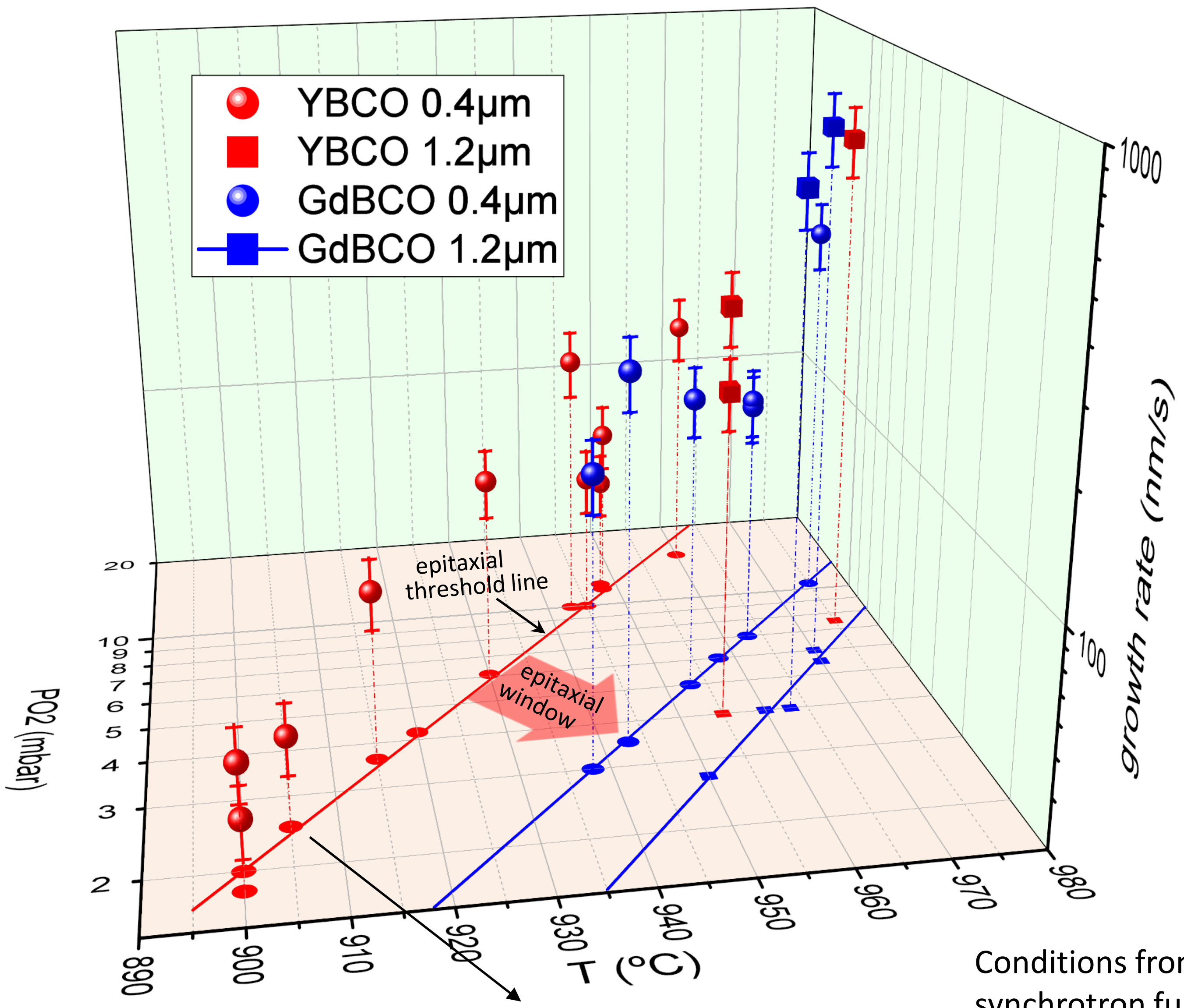






Colaboration with T. Kiss for superconducting analysis by SHPM

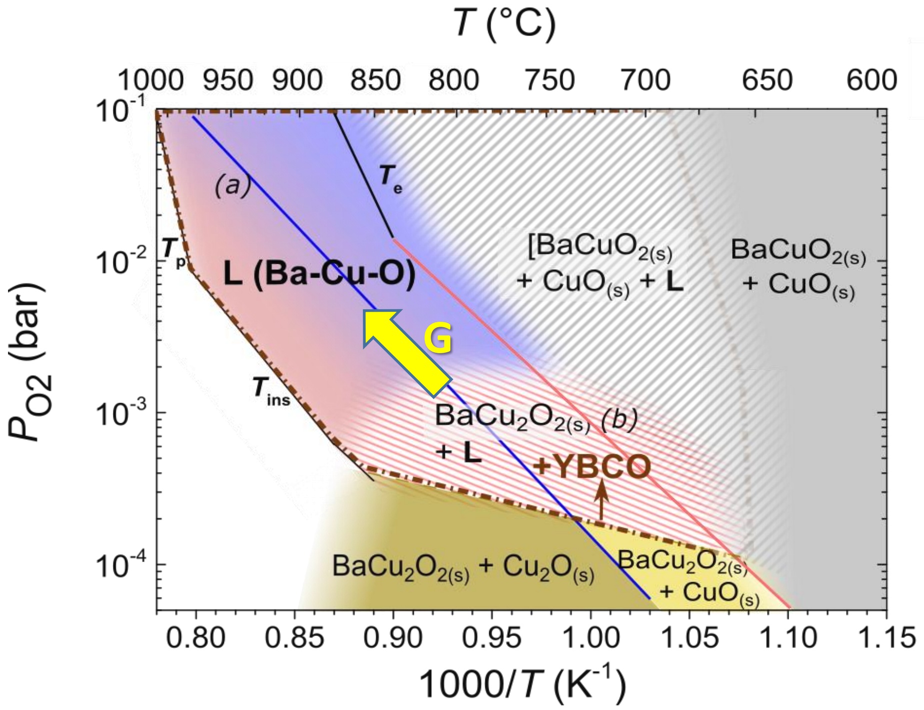
Growth rate dependence on TLAG growth conditions



$LnP_{O2} = m \cdot T - n$

Conditions from ALBA synchrotron furnace

Growth rate, G, ↑ if P_{O2} (+ T) ↑



From magnetic granularity analysis of TLAG films

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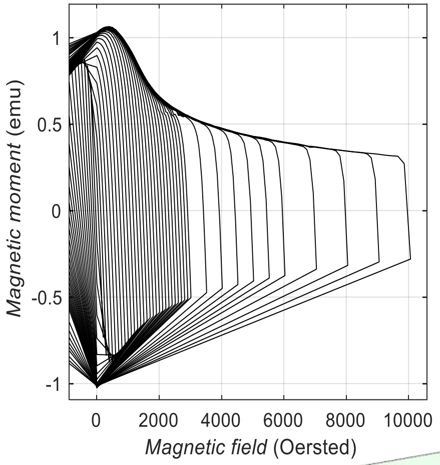
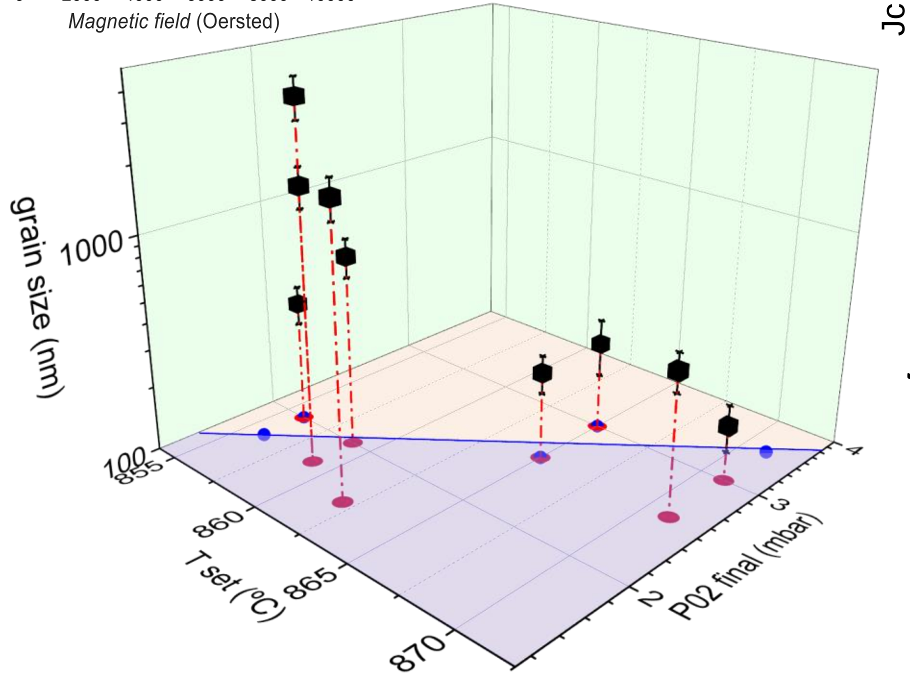
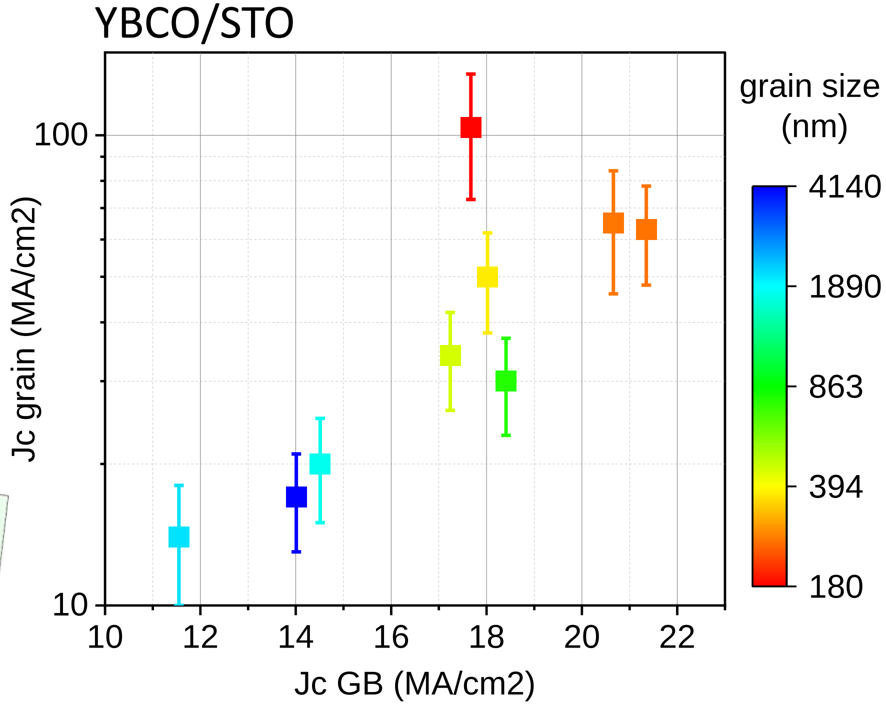


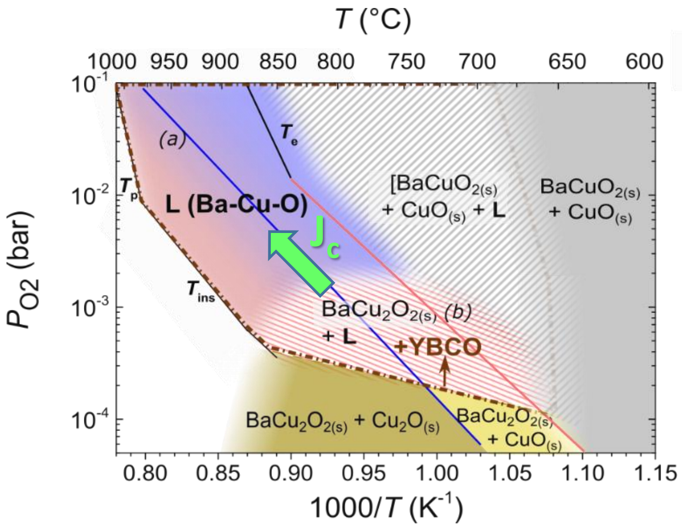
A. Palau et al, Phys. Rev. B (2007)



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



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



High growth rates (high supersaturation conditions) are beneficial for TLAG films

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Microstructure of TLAG pristine films at high growth rates

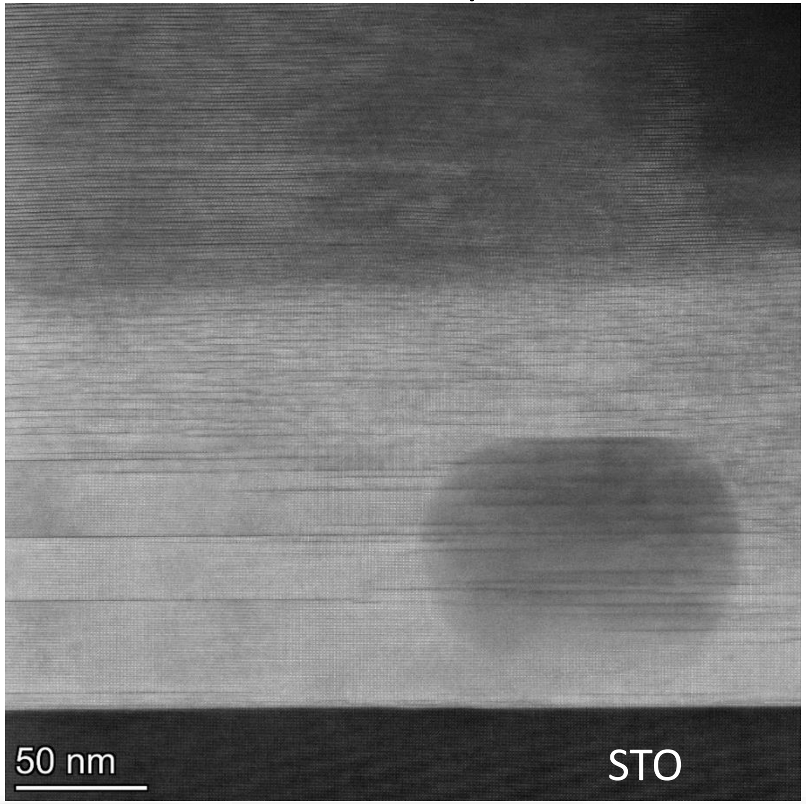
Main defects:
- -SF
- -partial disloc.
- -strain
- -TB
- -oxygen and cluster vacancies

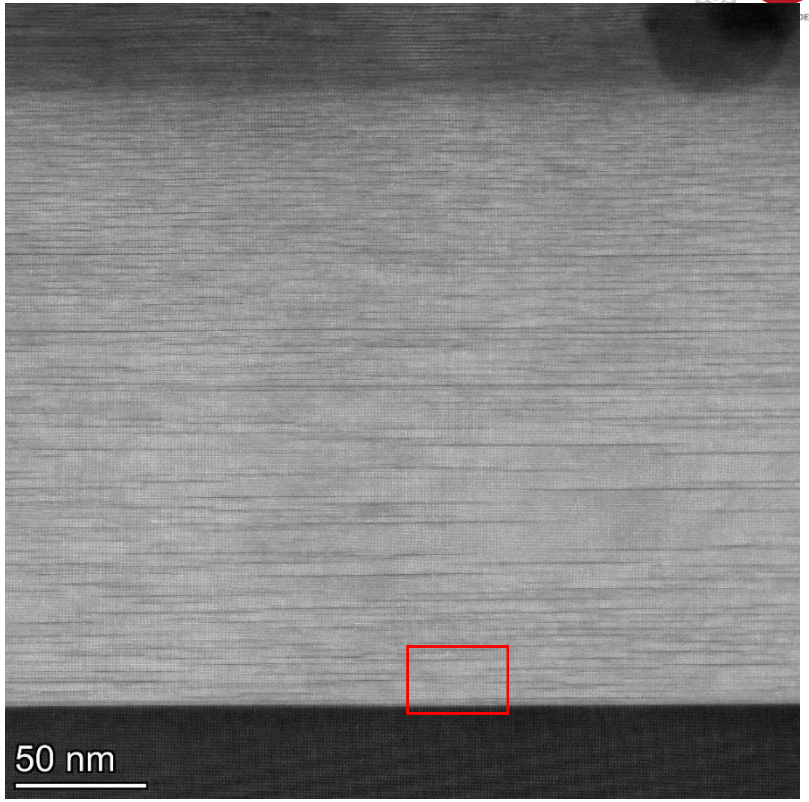
STEM-HAADF YBCO/STO

<u>High</u> density of **<u>long</u>** intergrowths (SF)

<u>High</u> density of **<u>short</u>** SFs

<u>Low</u> density of **<u>long</u>** SFs





<u>High</u> density of **<u>long</u>** SFs

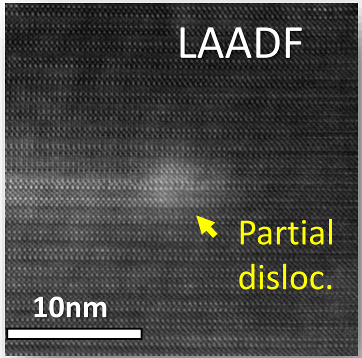
<u>High</u> density of **<u>short</u>** SFs

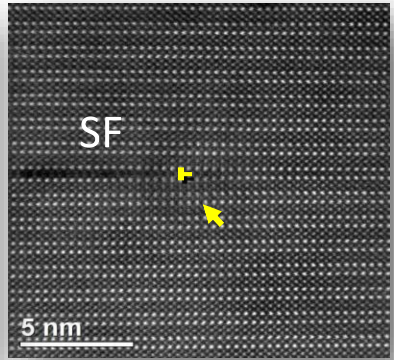
<u>Medium</u> density of **<u>long</u>** SFs

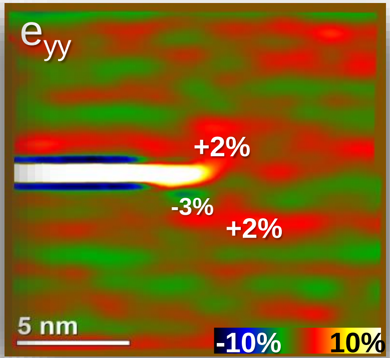
<u>High</u> density of **<u>short</u>** SFs

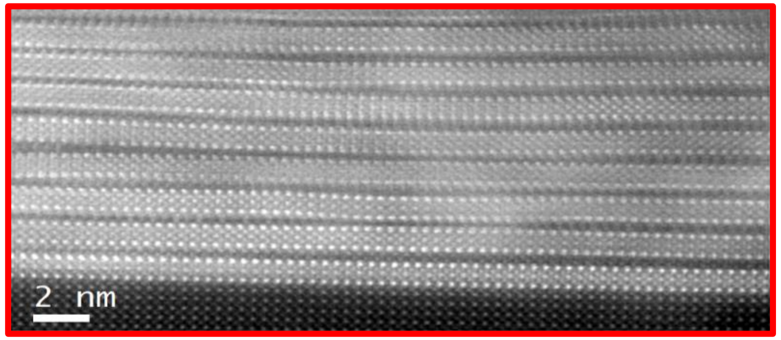
Strain accumulated at the partial dislocation surrounding the SF (**NANOSTRAIN**)

Also measured by XRD (Williamson-Hall)









T. Puig et al, Nat Rev Phys (2023), L. Soler et al., Nat Comm (2020), A. Llordes et al, Nat Mat (2012)

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STEM-HAADF from TLAG films at different conditions

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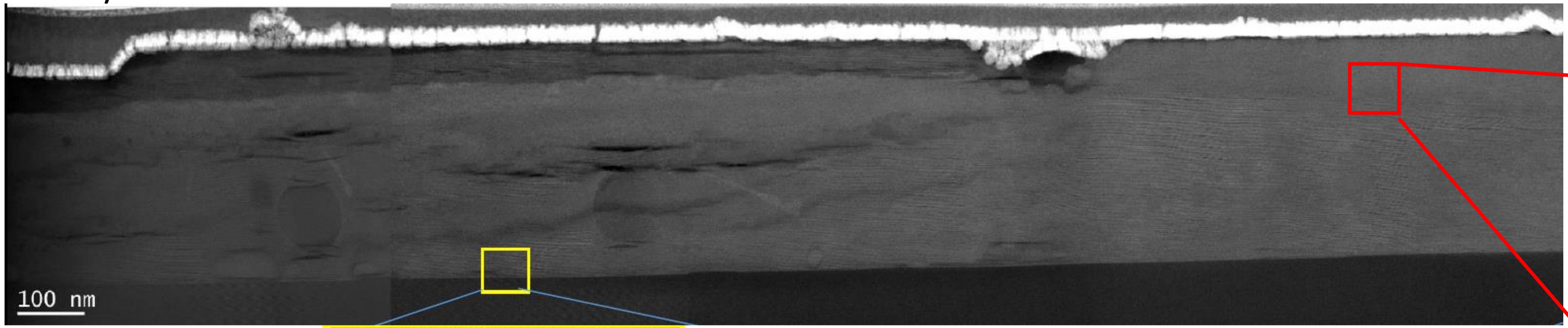
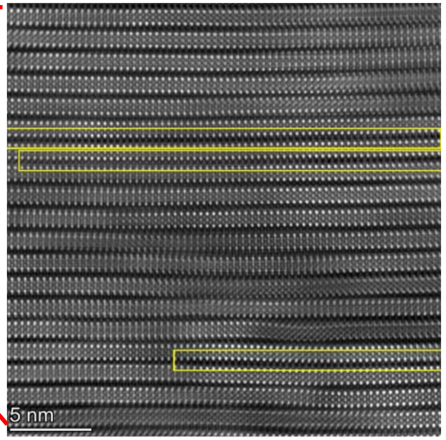
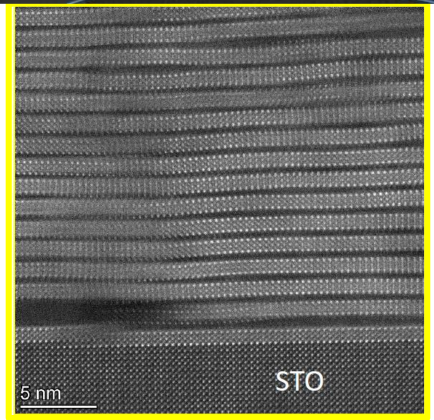
YBCO/STO



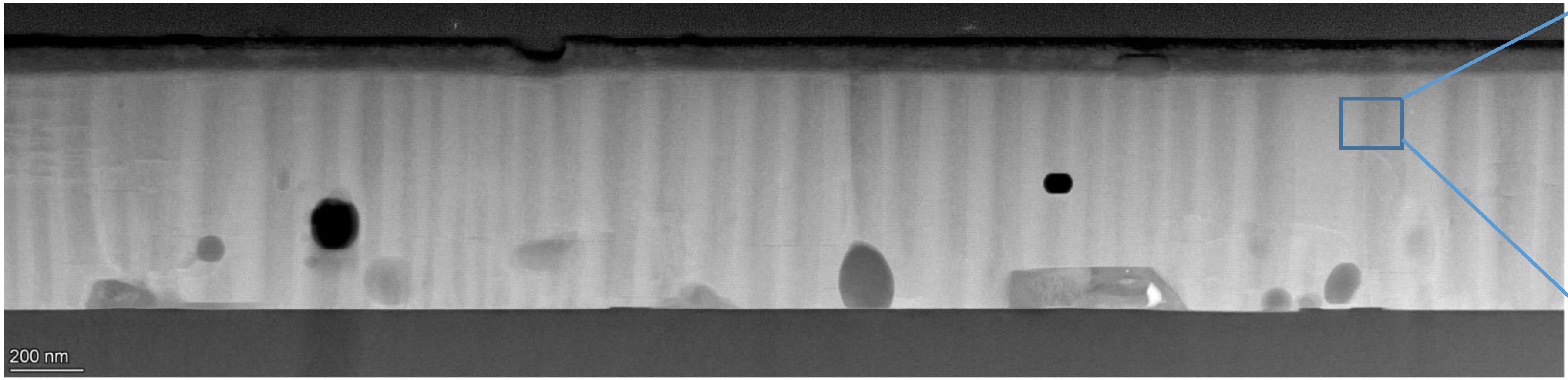


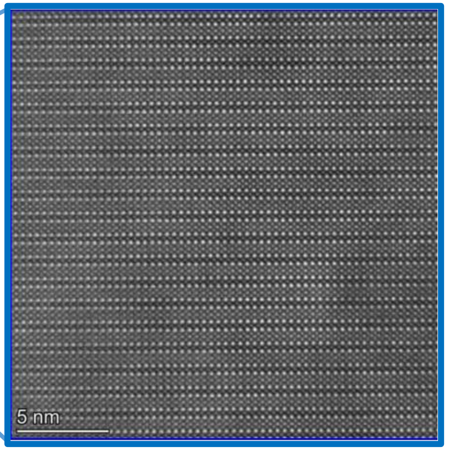


350 nm, J_c(77K)=2.5 MA/cm^2

Stablishing the correlation between defects microsctructure, process conditions, supersaturation and growth rate is the next step

750 nm, J_c(77K)=2 MA/cm^2





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suman SUPERCONDUCTING MATERIALS AND FUNCTIONAL NANOENGINEERED STRUCTURES
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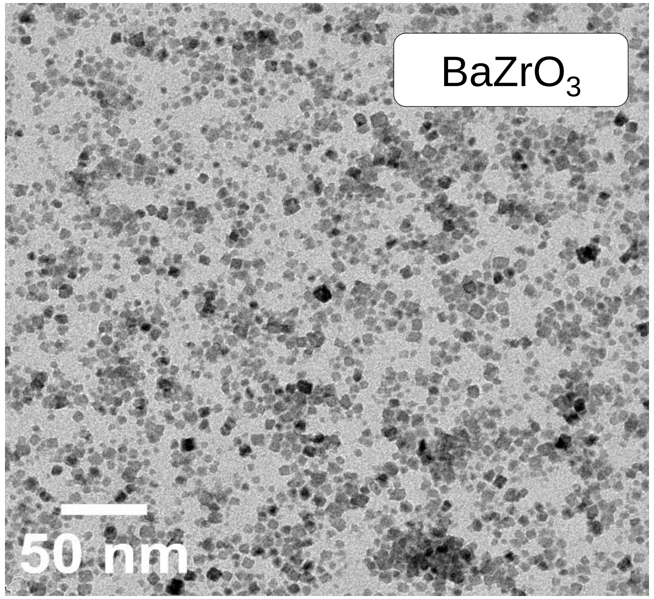
erc European Research Council Executive Agency

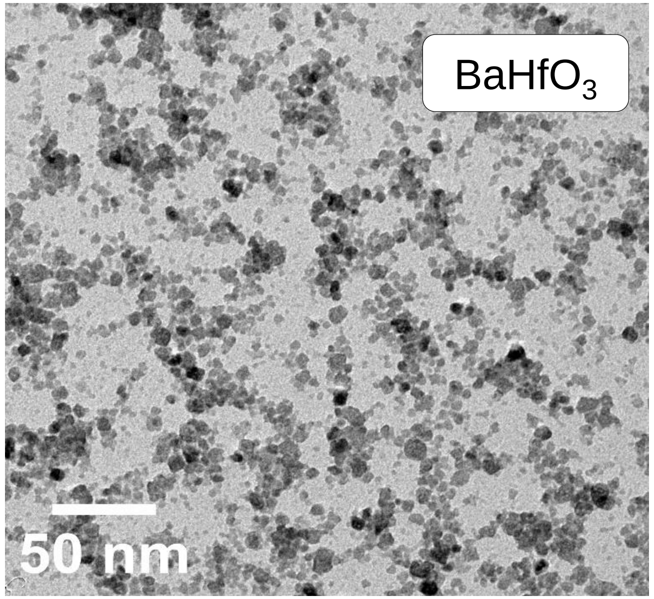
TLAG nanocomposites with Multifunctional Colloidal Inks

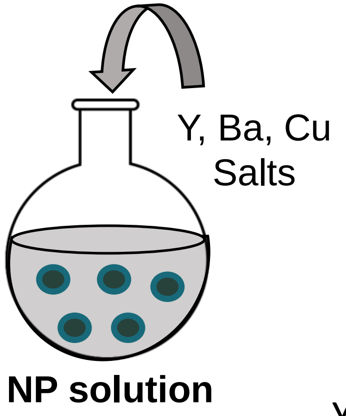
Hybrid Hydrolitic-Solvothermal Synthesis (H2S2)

$BaMO_3$ (M= Zr, Hf)

N. Chamorro et al, RSC Adv. (2020)





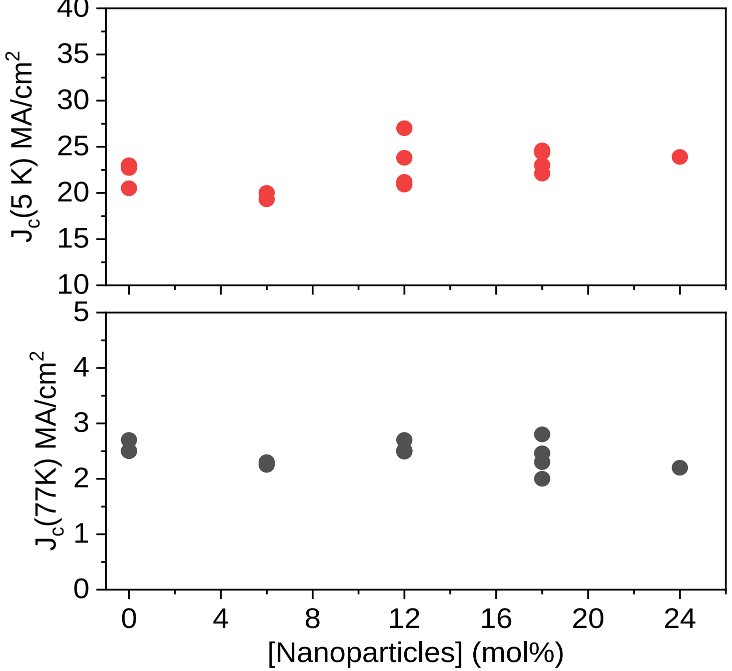


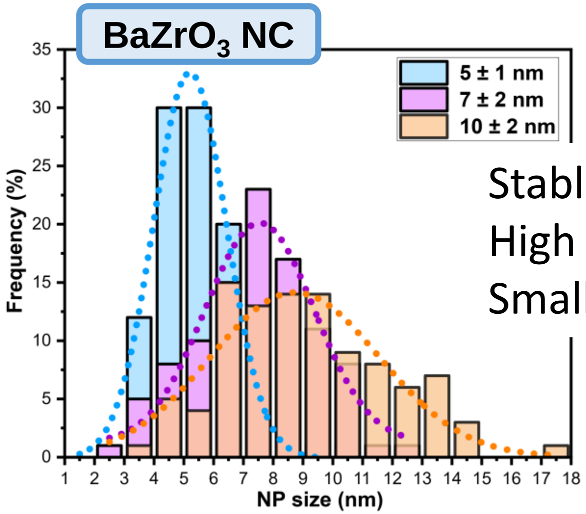
Colloidal- metalorganic ink stable for months and scaled up to 1L

(Patent EP22382741)

See 5MOr2A-02, C. Pop

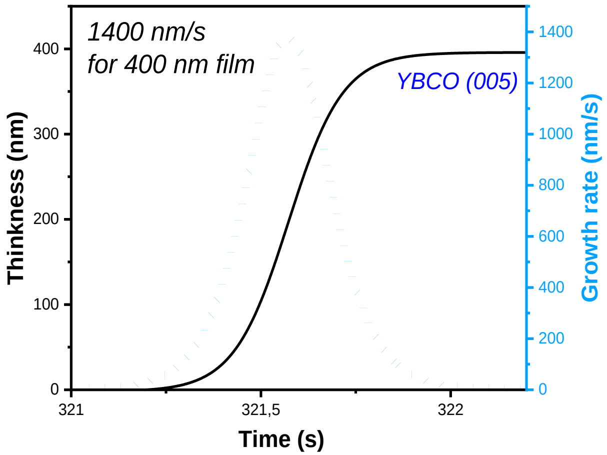
High performance up to 24%M BMO
YBCO/STO 0.4-0.9 µm thick





Stable
High concentrations
Small size and distribution

Ultra-high growth rates



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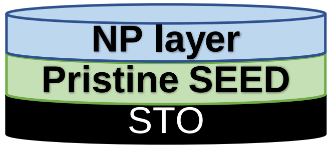
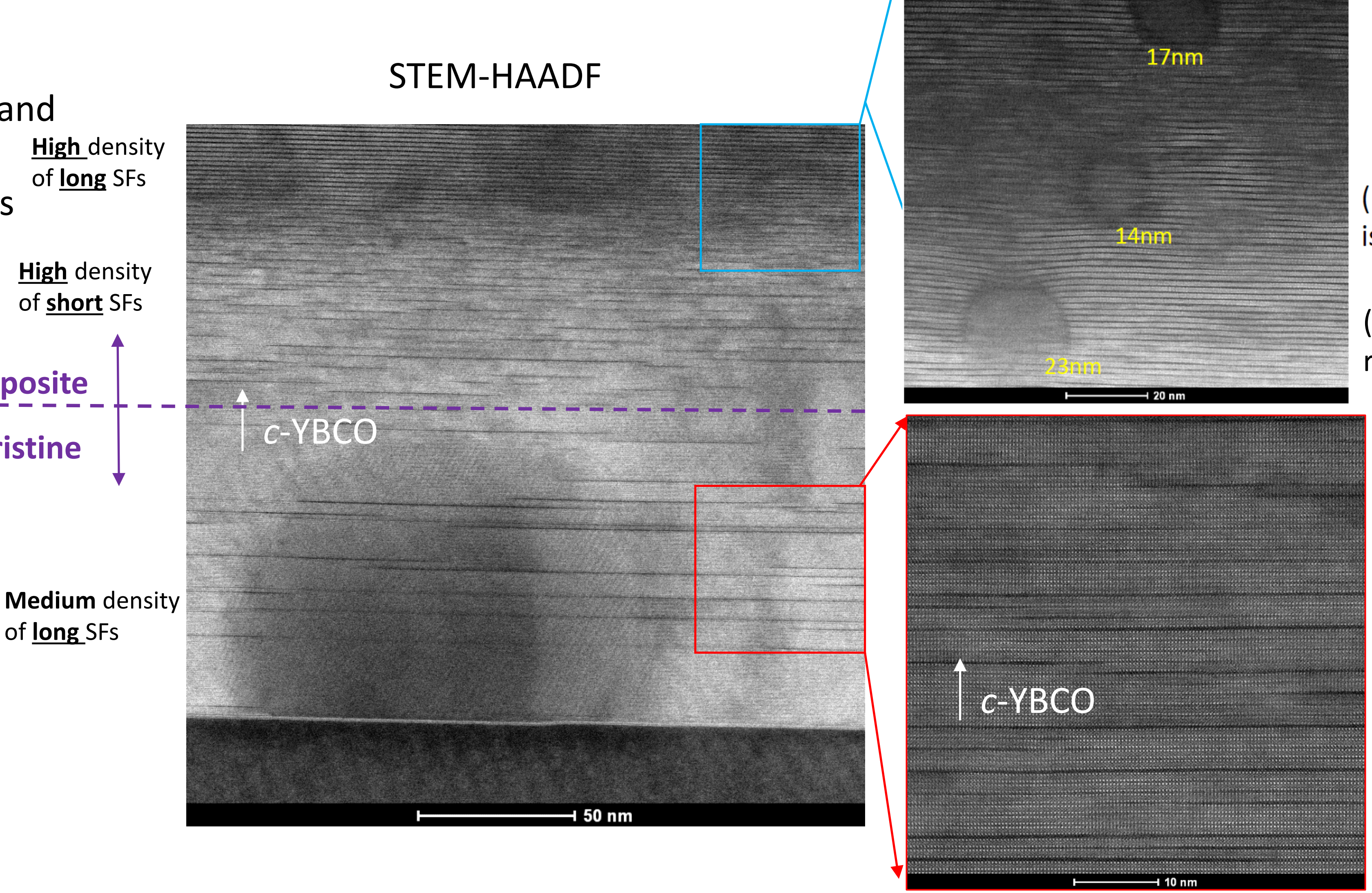
Microstructure of TLAG nanocomposite films

Main defects:

- -SF
- -partial disloc.
- -strain
- -TB
- -oxygen and cluster vacancies
- -NP



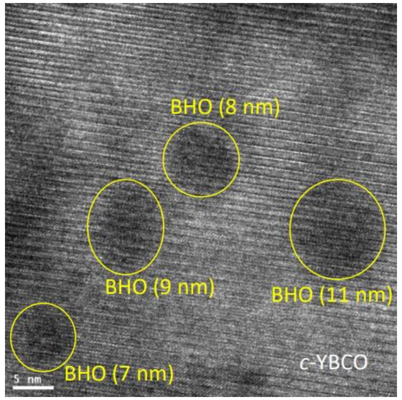
STEM-HAADF



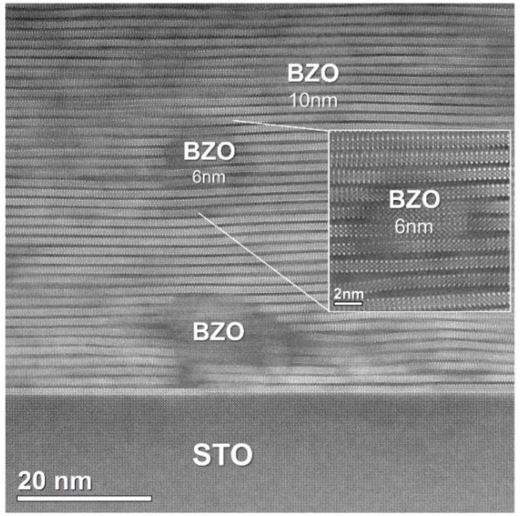
(14 nm BZO is epitaxial)

(23 nm is random)

BHO NPs in YBCO



BZO NPs in YBCO



T. Puig et al, Nat Rev Phys (2023), L. Soler et al., Nat Comm (2020)

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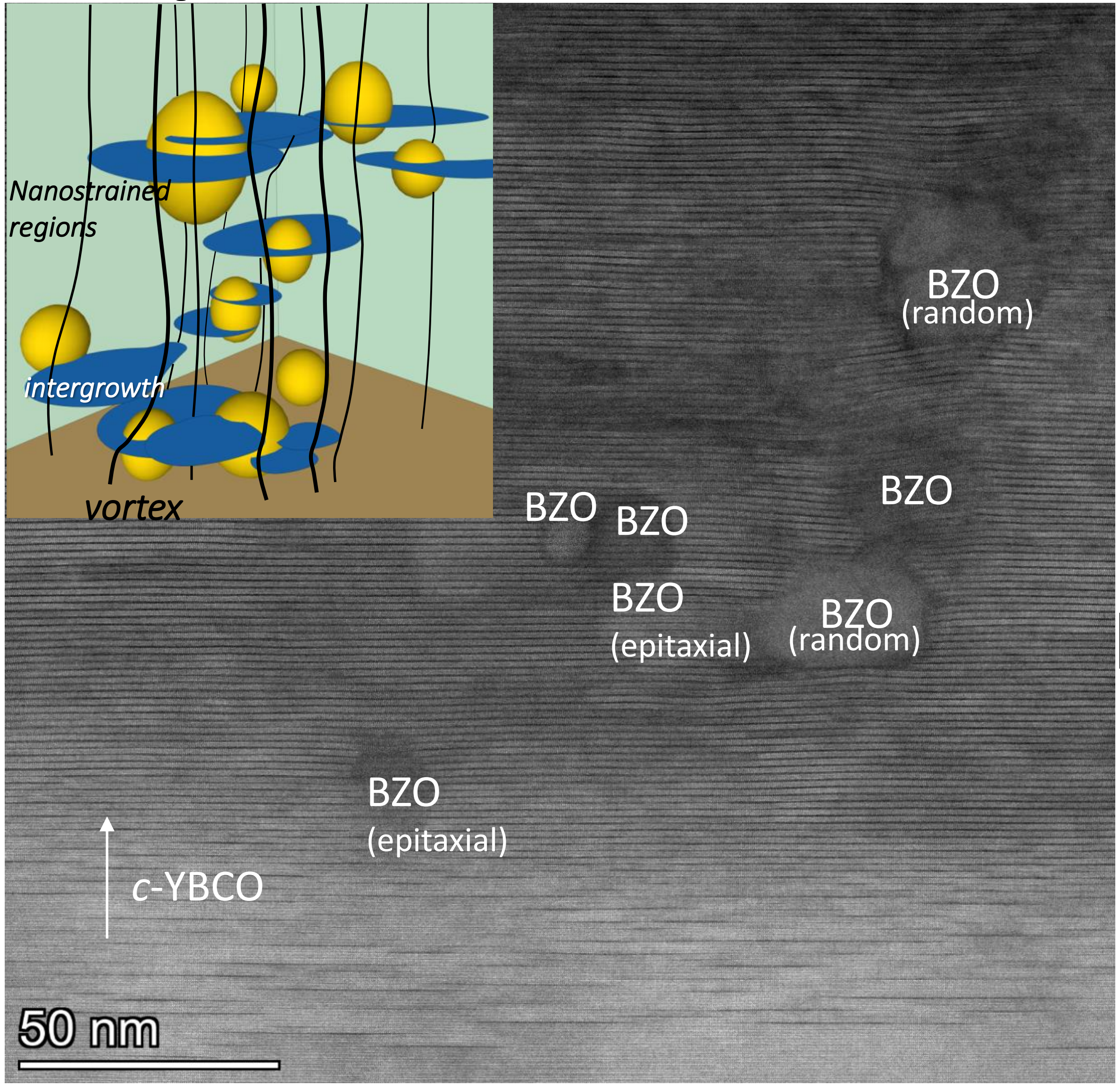
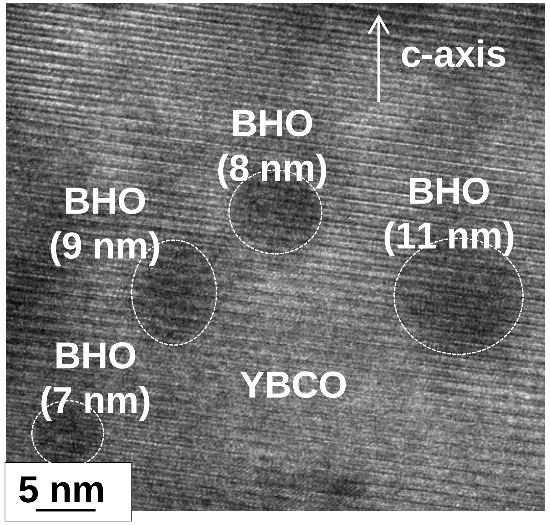
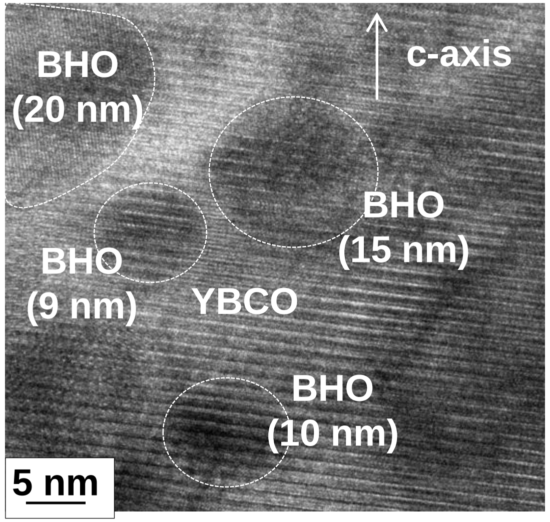
High throughput TLAG-CSD epitaxial superconducting nanocomposite films







Epitaxial NPs (7-11 nm < 20 nm)

Favourable to rotate at the liquid/solid interface

Random NPs ≥ 20 nm

T. Puig et al, Nat Rev Phys (2024), L. Soler et al., Nat Comm (2020)

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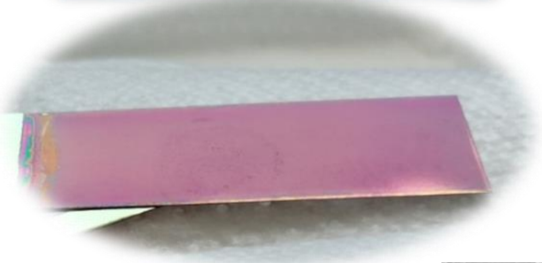
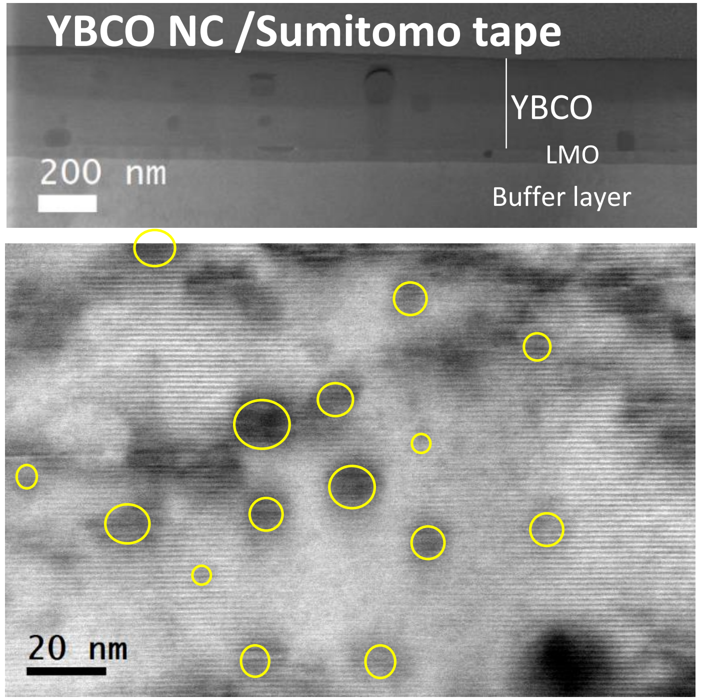
TLAG Coated conductors

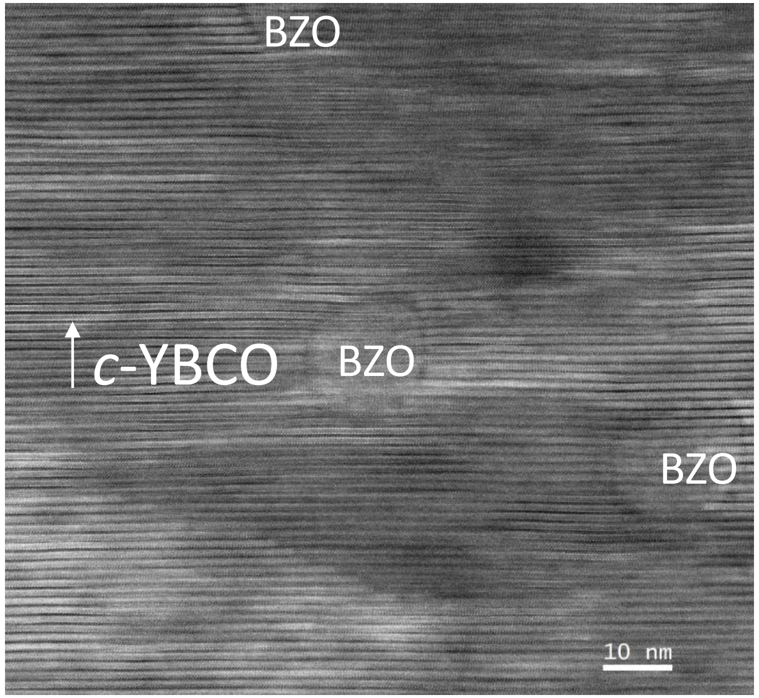
SUMITOMO ELECTRIC | SuNAM | ICMAB | CSIC

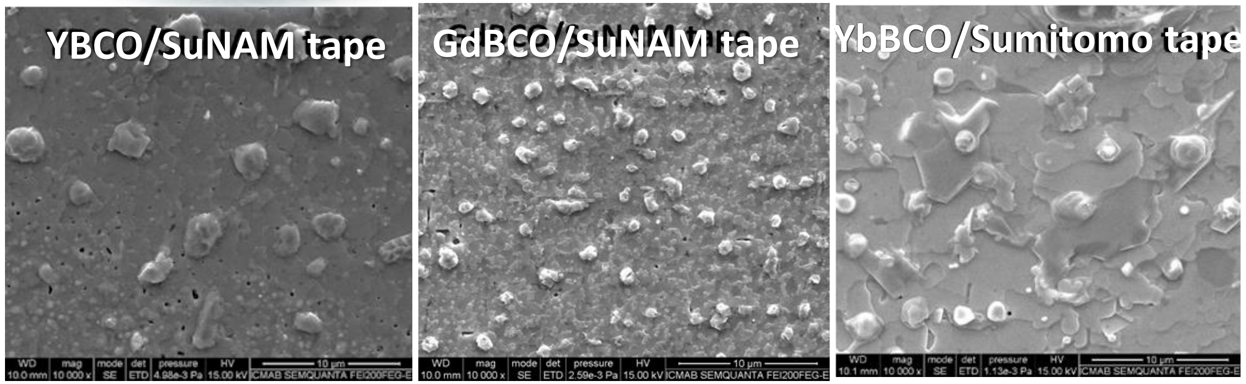


See 2MOr1C-05, R. Vlad

Slot die
now with 40 mm-width printhead capabilities



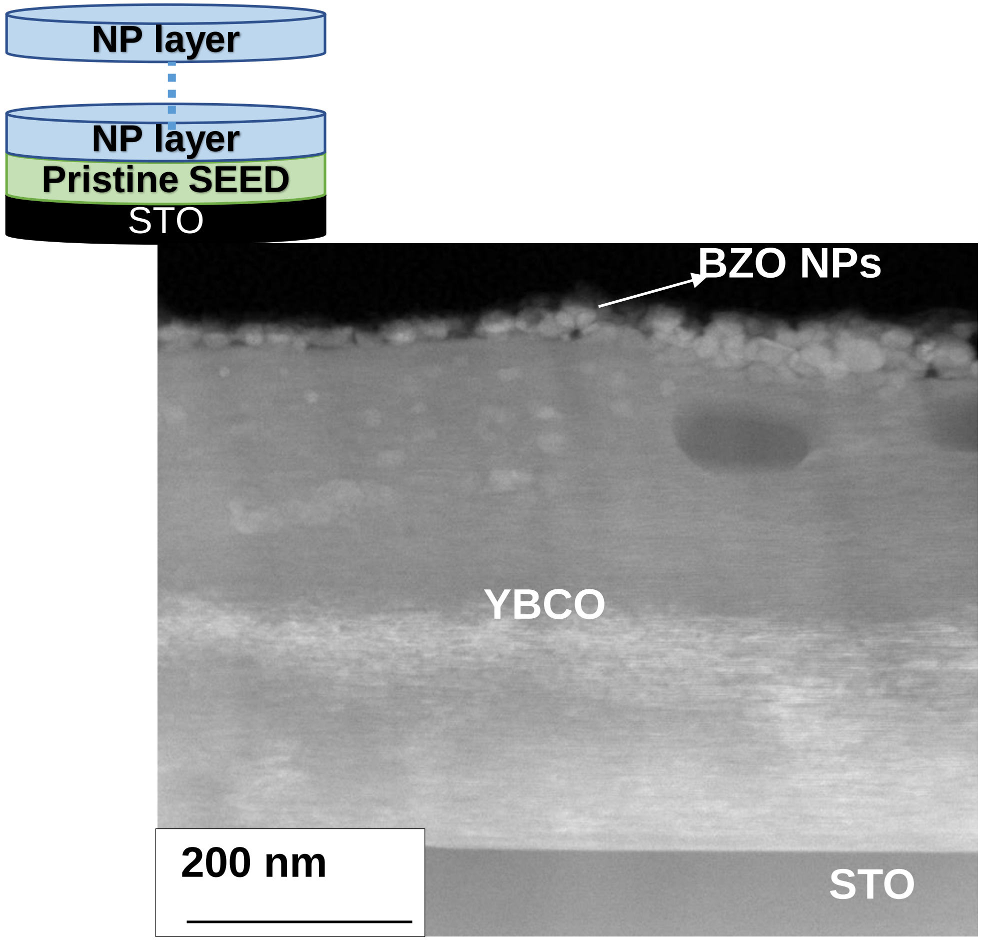


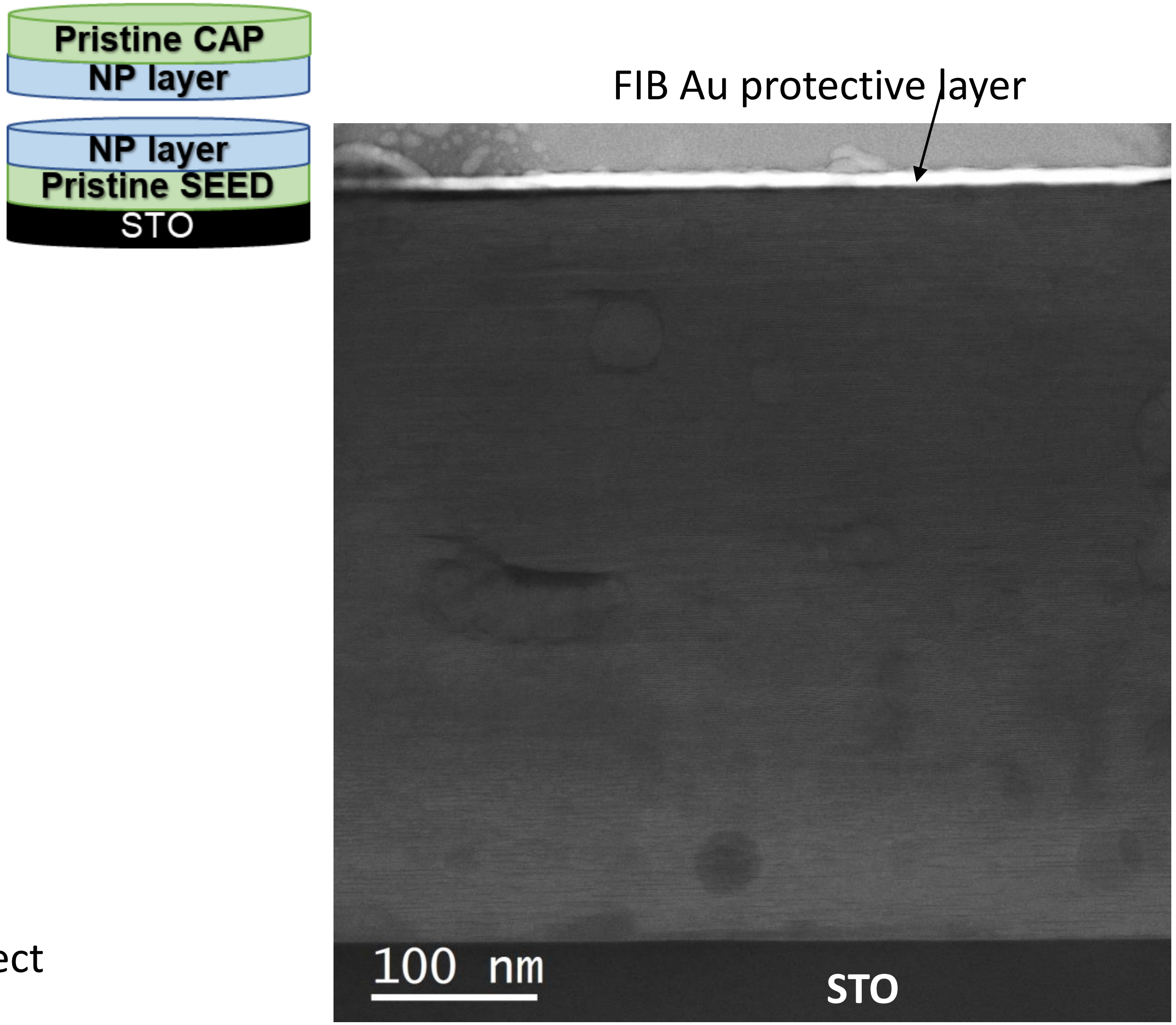


Microstructure is reproduced in 250-750 nm CC
High superconducting properties: **J_c(77K) =1.7-2 MA/cm^2**

Furnace for 40 mm-width tape on going

YBCO TLAG-CSD nanocomposite films with small nanoparticles





-Small nanoparticle (5 nm) tend to promote some pushing effect
-A pristine thin cap layer is able to avoid it
-The different liquid viscosities and densities (Pristine vs NC with the additional NPs solid part) might be at the origin of liquids non-miscibility

Superconducting properties of YBCO Nanocomposites with small BZO Nanoparticles

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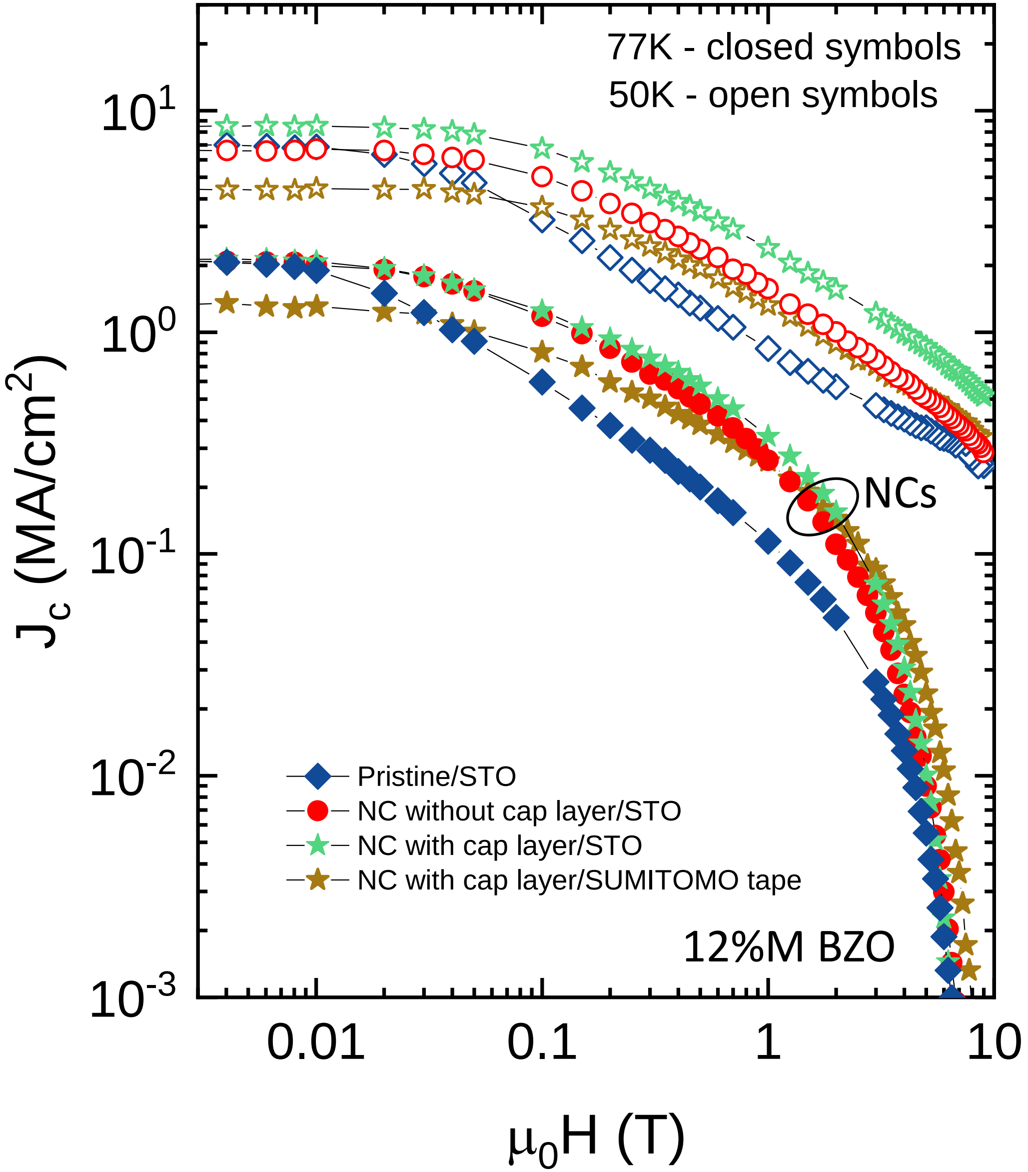

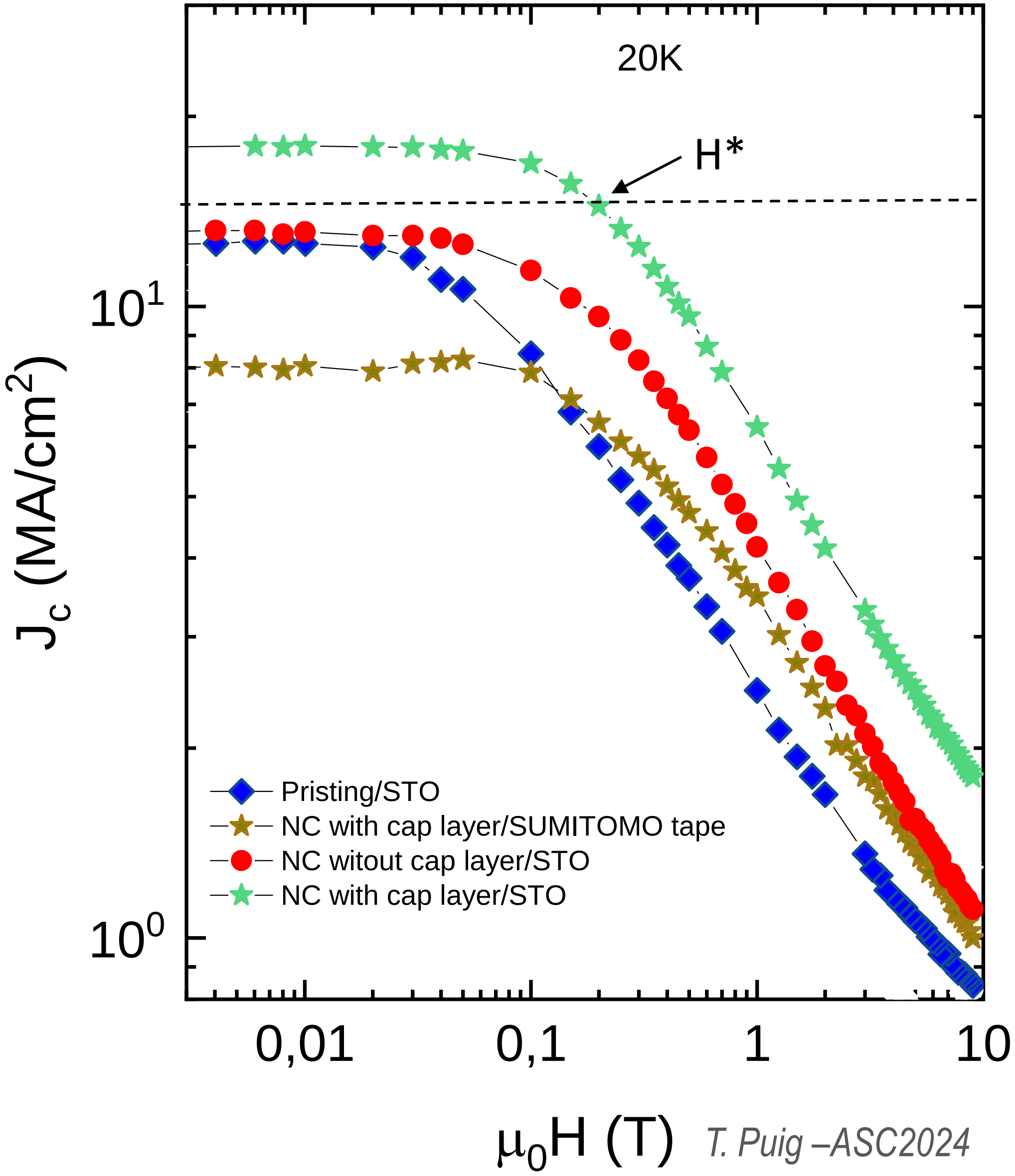

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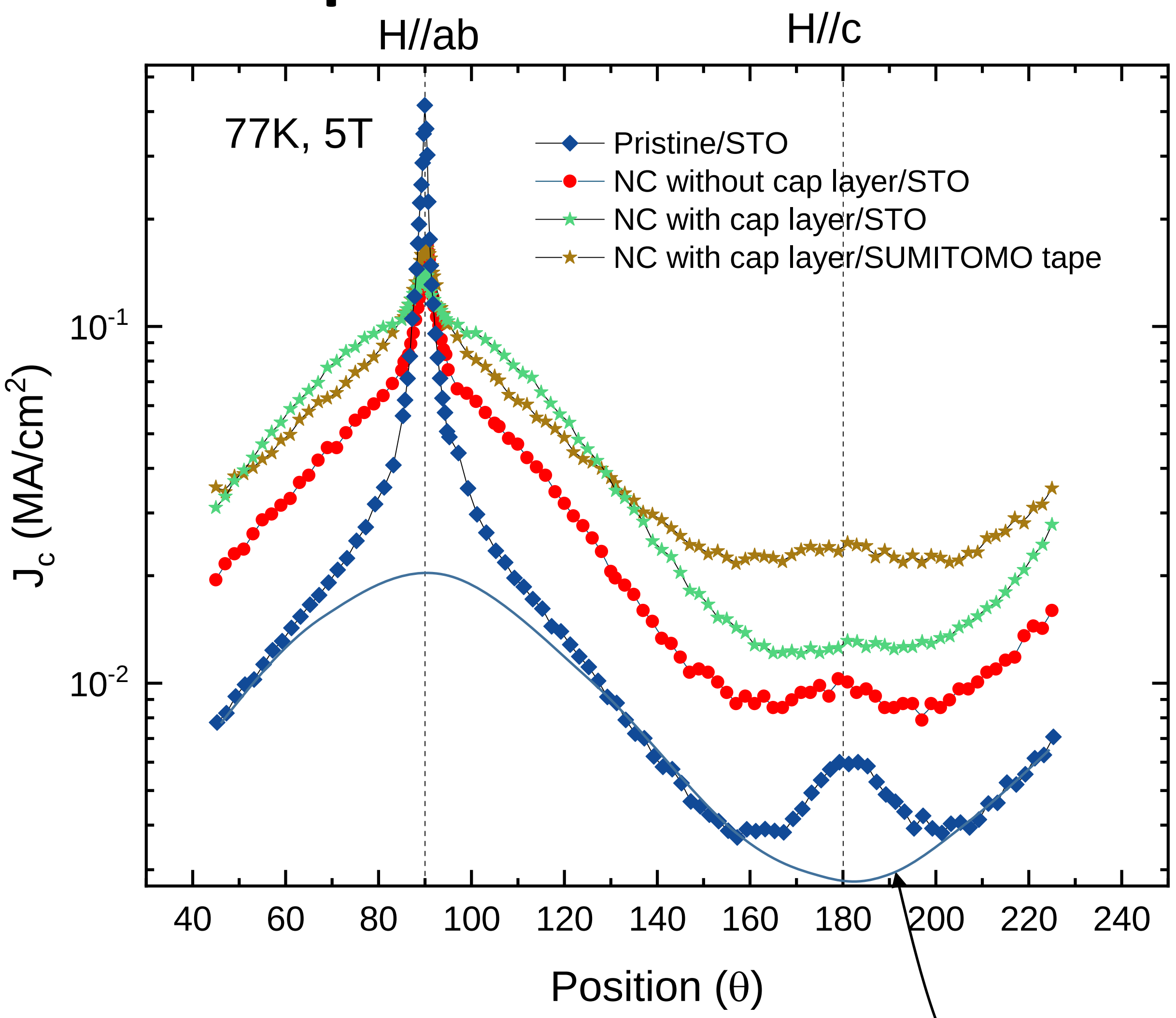
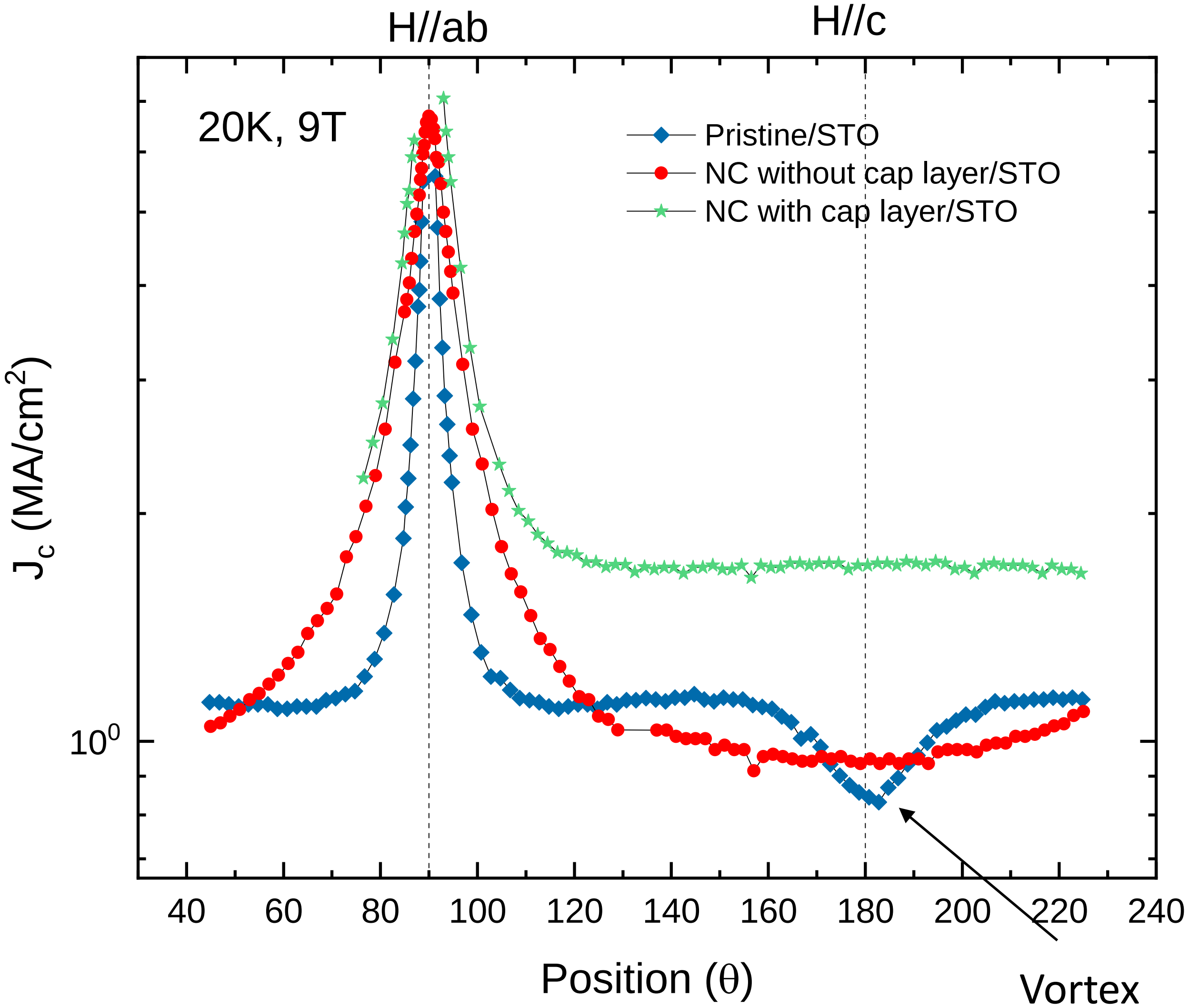
Superconducting properties of YBCO Nanocomposites with BZO Nanoparticles





Blatter scalling approach

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

Vortex channelling

Superconducting properties of YBCO Nanocomposites with BZO Nanoparticles

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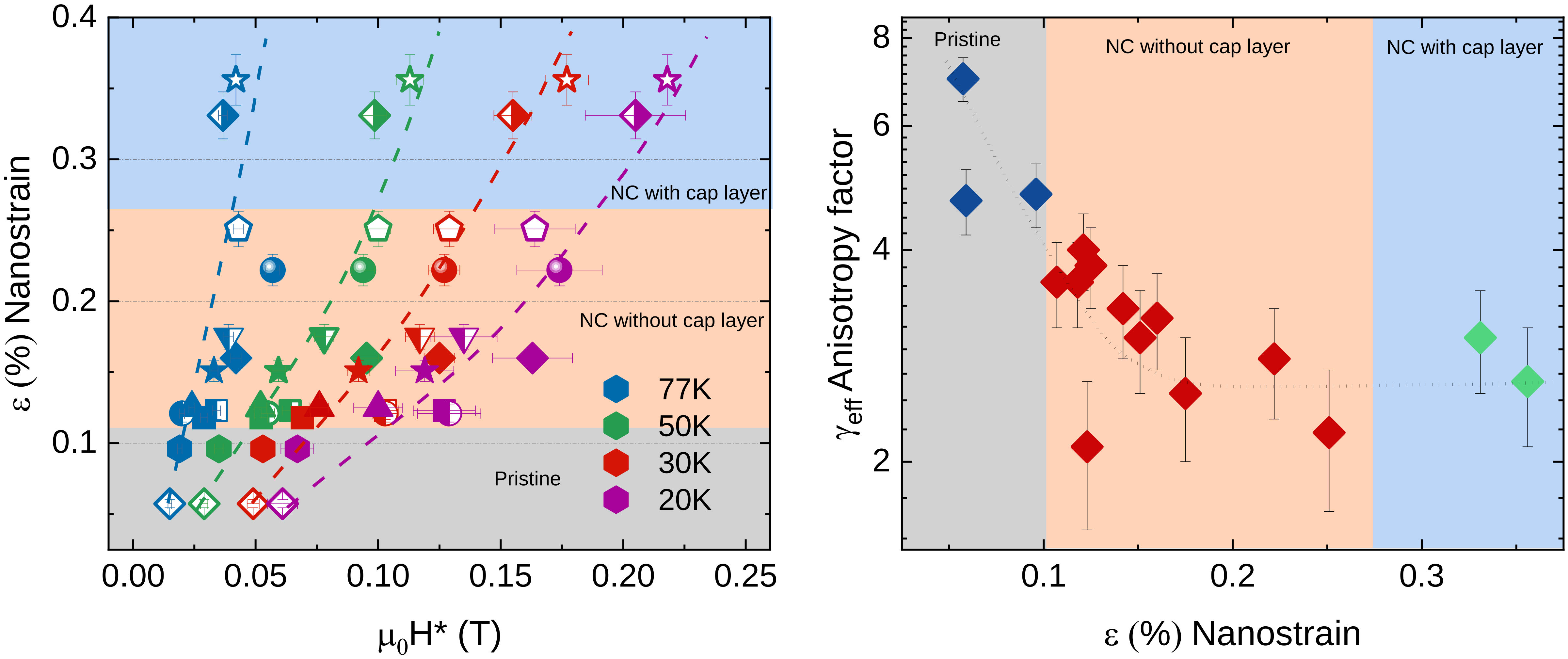

Strong correlation between Nanostrain and H* and effective anisotropy factor

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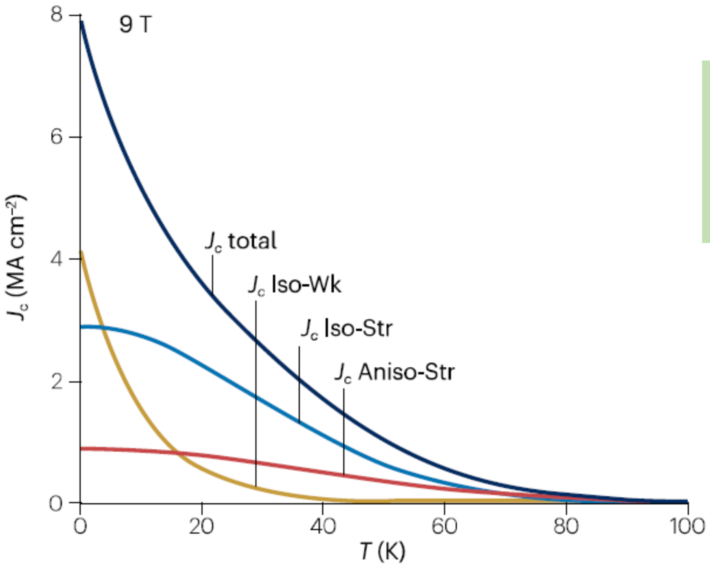
Pinning Model Analysis

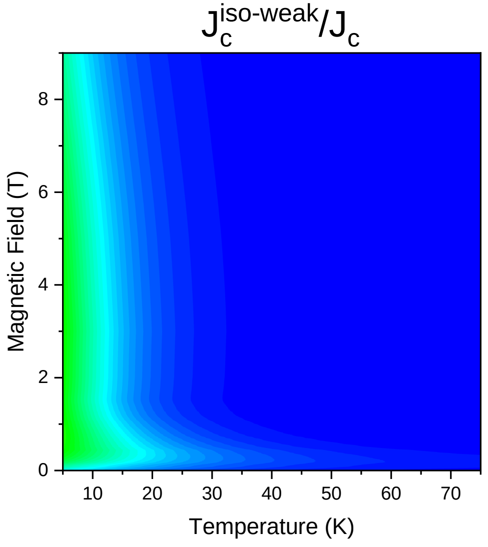


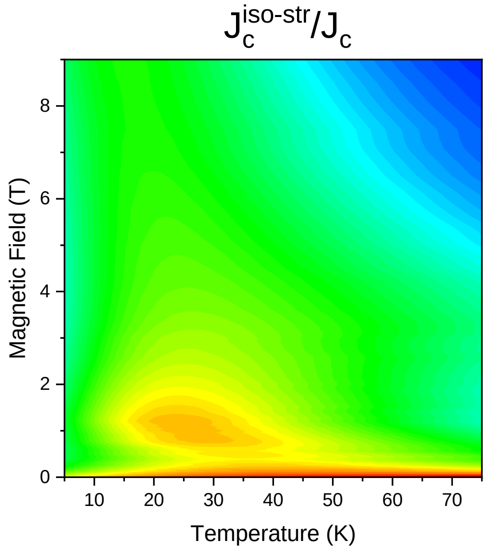
$$\mathbf{J_c(T) = J_c^{\mathit{iso\text{-}wk}}(\mathit{T}) + J_c^{\mathit{iso\text{-}str}}(\mathit{T}) + J_c^{aniso\text{-}str}(\mathit{T})}$$

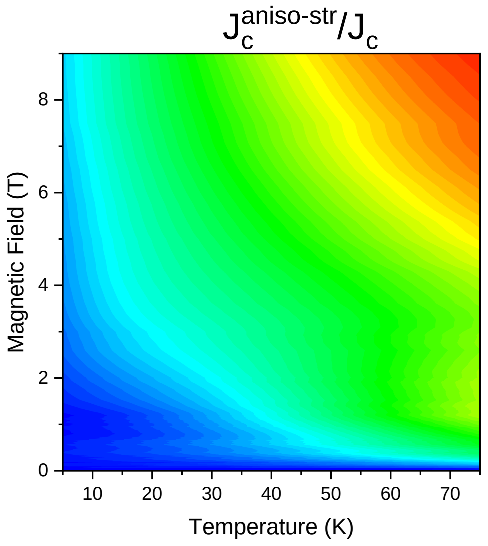
$$J_c^{wk}(T) = J_c^{wk}(0)e^{\left(-\frac{T}{T_0}\right)}$$

$$J_c^{str}(T) = J_c^{str}(0)e^{\left(-3\left(\frac{T}{T^*}\right)^2\right)}$$

Pristine

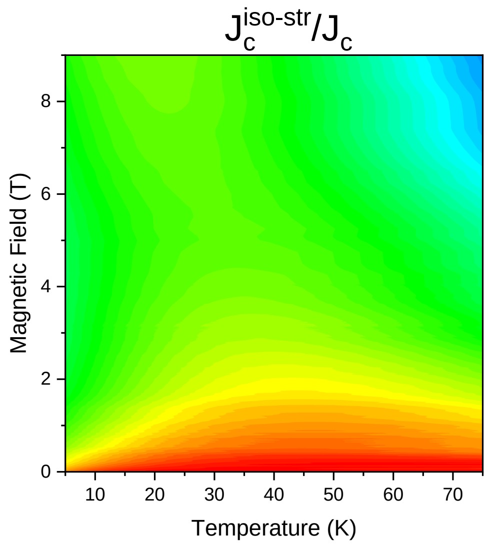


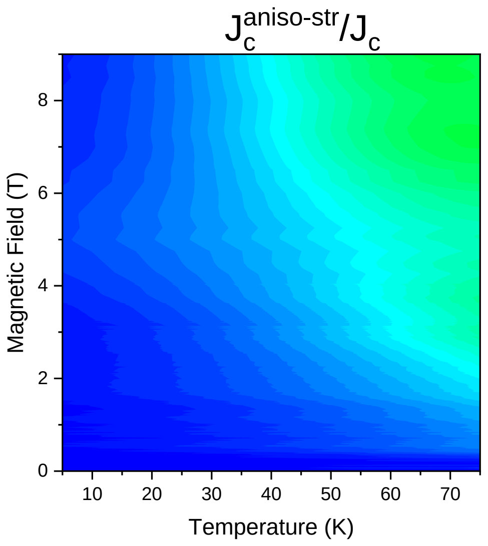




H//c

Nanocomposite

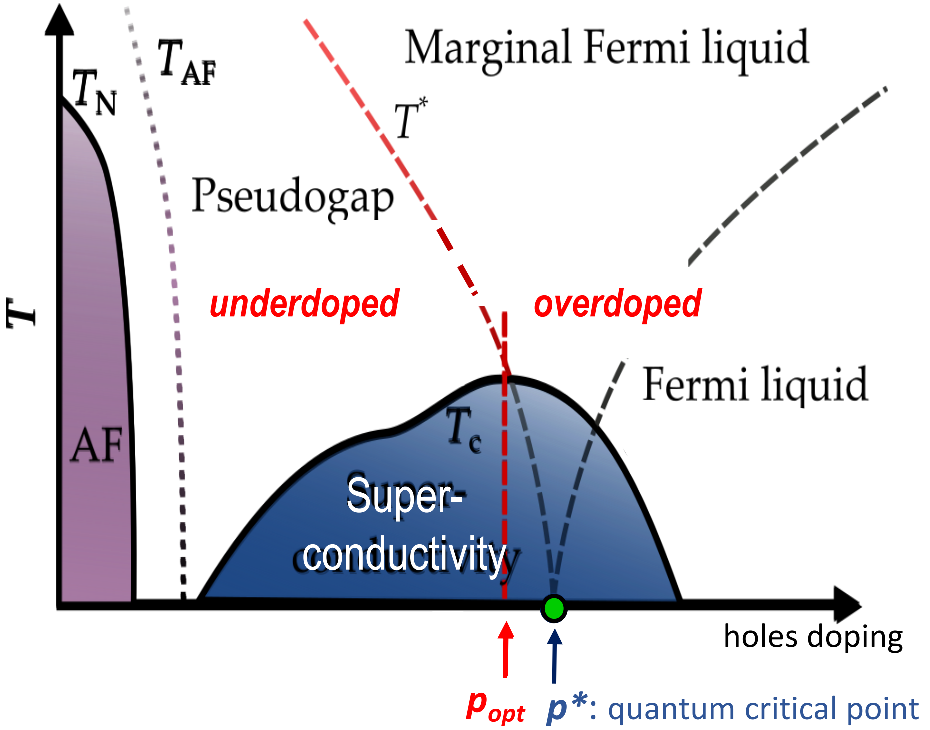


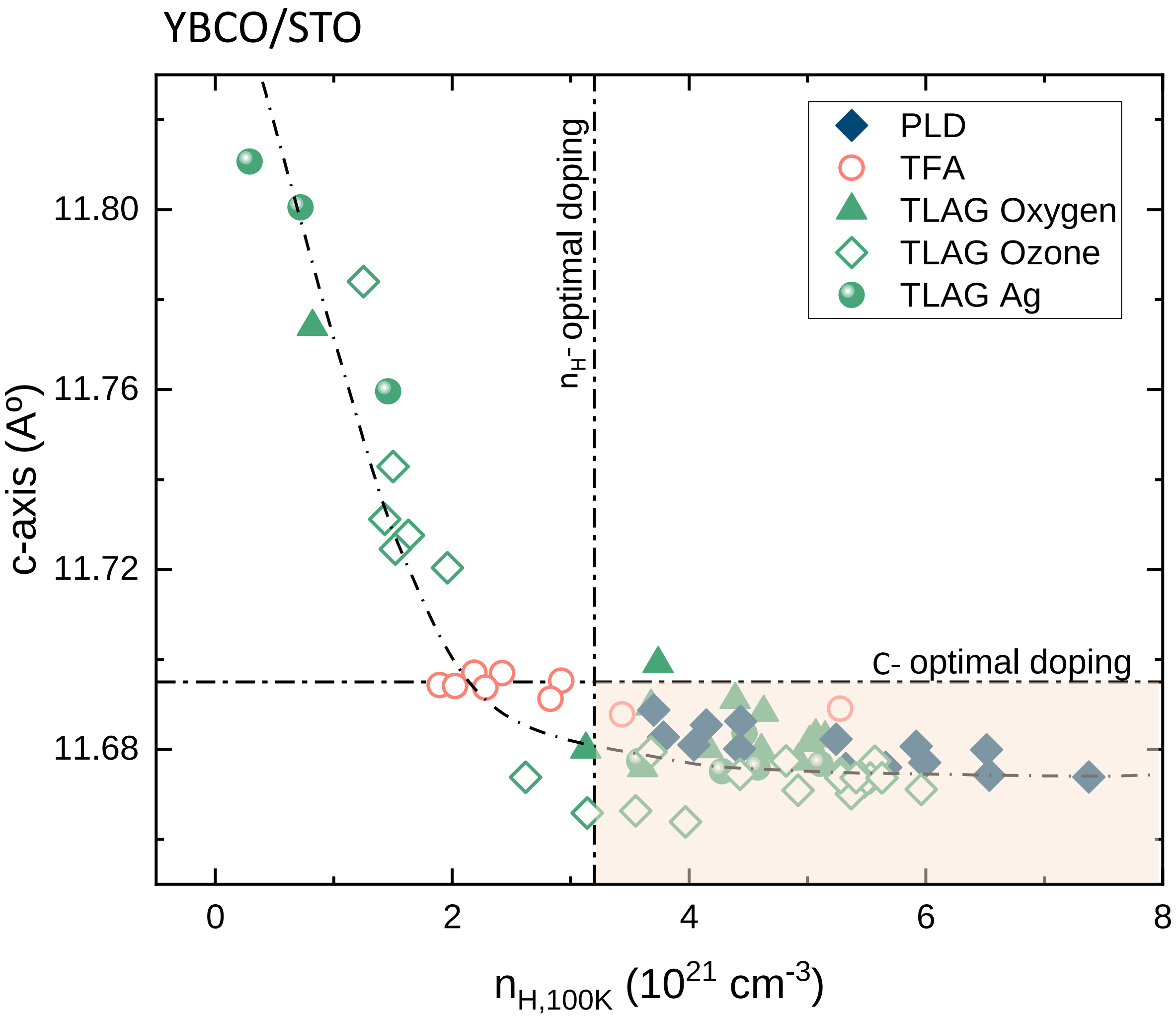


↑ isotropic strong pinning contribution in NCs
↑ anisotropic pinning contribution in pristine

T. Puig et al, Nat Rev Phys (2023), F. Valles et al, Comm Mat 3 (2022)

Doping REBCO films





A. Stangl et al, Sci. Rep. (2021), A. Kethamkuzhi et al (to be published)

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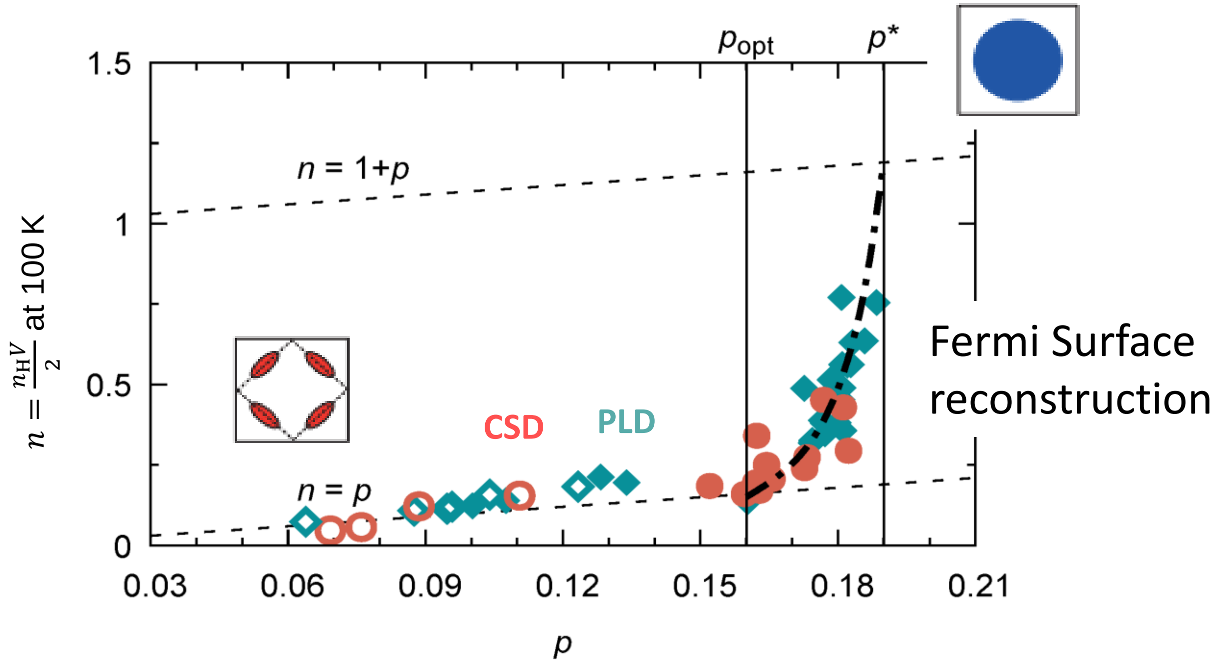
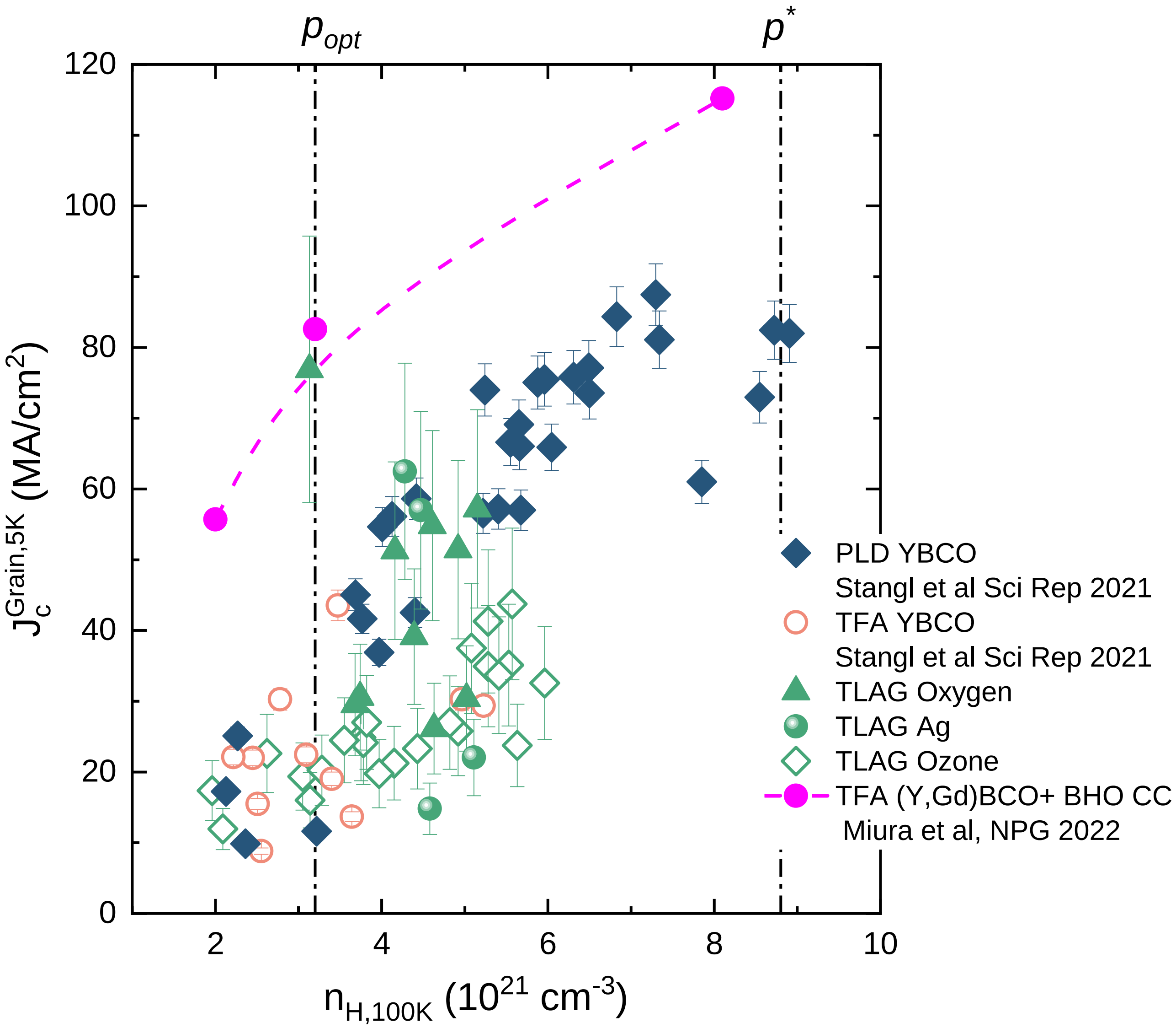
$J_c(n_H, T, H)$ in the Overdoped state: where Condensation energy and charge carrier density increases



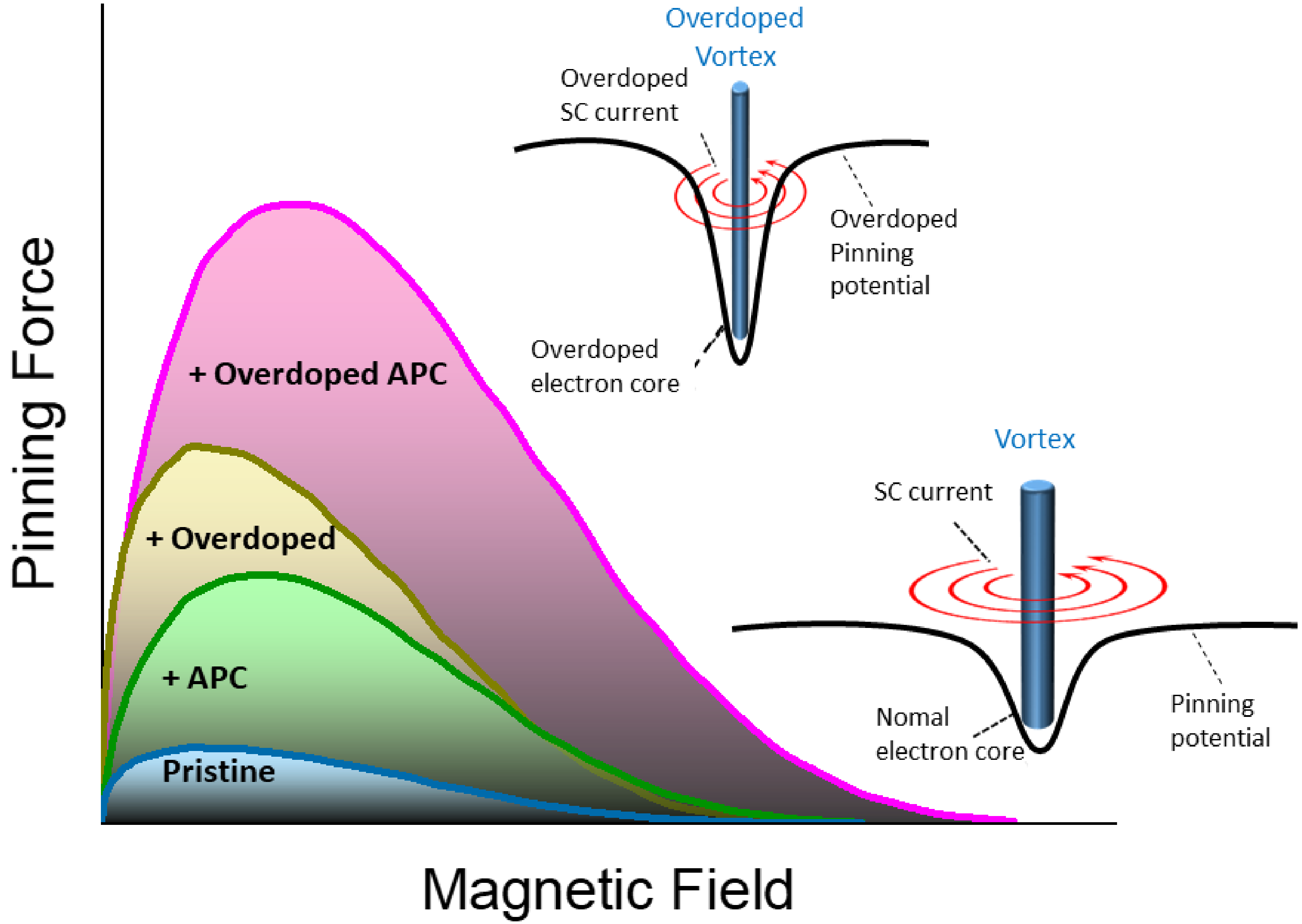
$If\ n_H \uparrow \rightarrow J_c^{sf} \uparrow$

$but\ n_H \propto E_c \propto f_p$



Interest in combining high throughput and nanocomposite CC in overdoped state

Improving vortex pinning in Coated Conductors by APC and electronic charge carriers



Double benefit of overdoping:

- $n_H \uparrow \rightarrow J_c^{sf} \uparrow$
- $\lambda\downarrow, \xi\downarrow \rightarrow E_c \uparrow \rightarrow f_p \rightarrow J_c\,(H) \uparrow$

Vortex pinning defects should have higher efficiency

Need to study vortex physics in the overdoped state

X. Obradors, T. Puig, "Pin the vortex on the superconductor", Nature Mater. News and Views (2024, in press); M. Miura et al, NPG Asia (2022), Miura et al, Nat Mat (2024)

Conclusions

- High growth rate methods, like TLAG, are the future for making CC a persistent enabling technology

- TLAG is a high-throughput low-cost method compatible with solution deposition methods, nanocomposites growth and coated conductors

- The kinetic character enables additional versatility though increasing understanding complexity, thus in-situ growth characterizations and fast screening methods are being employed

- REBCO TLAG films expand to a very broad epitaxial window where supersaturation determines nucleation and growth rate, grain size, microstructure defects and critical currents

- They present a rich microstructure interesting for vortex pinning which efficiency is increased by overdoping

Tomorrow:
5MOr1B “Get together: Challenges and Opportunities of Superconducting Materials”