

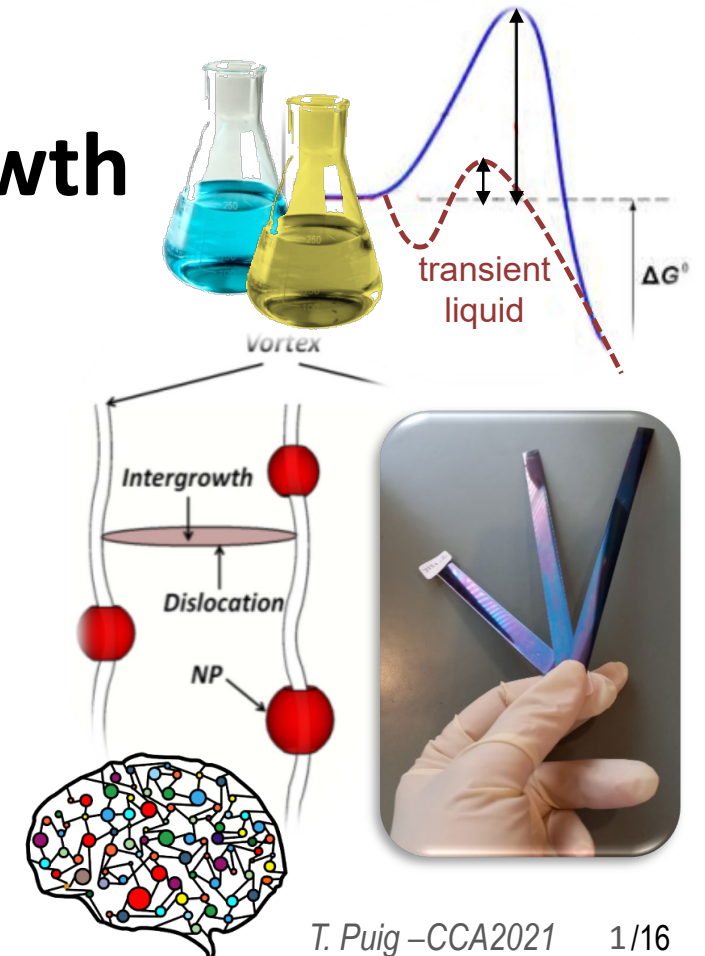
# Materials opportunities to boost HTS-CC

## Example of Transient liquid Assisted Growth

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# Materials Science opportunities for CC

## Decrease the cost/performance ratio



- 1. Decrease the cost/performance ratio by developing high throughput growth methods:**  
Transient Liquid assisted growth, TLAG-CSD process (100-1000 nm/s)
- 2. Use of High Throughput Experimentation strategies for fast optimization of processes:**  
Combinatorial approaches and Machine Learning algorithms
- 3. Foster higher critical currents by engineering the vortex pinning landscape for each application:**  
Understanding the  $(H, T, \theta)$  pinning landscape for engineered nanocomposites
- 4. Increase pinning force by increasing charge carrier density:**  
Tune charge carrier density by oxygen overdoping

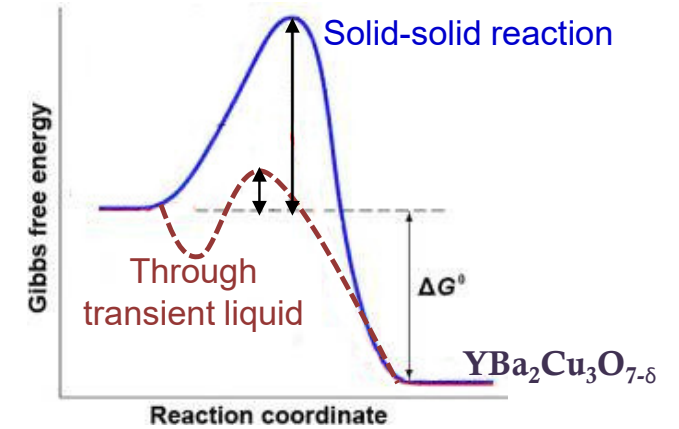
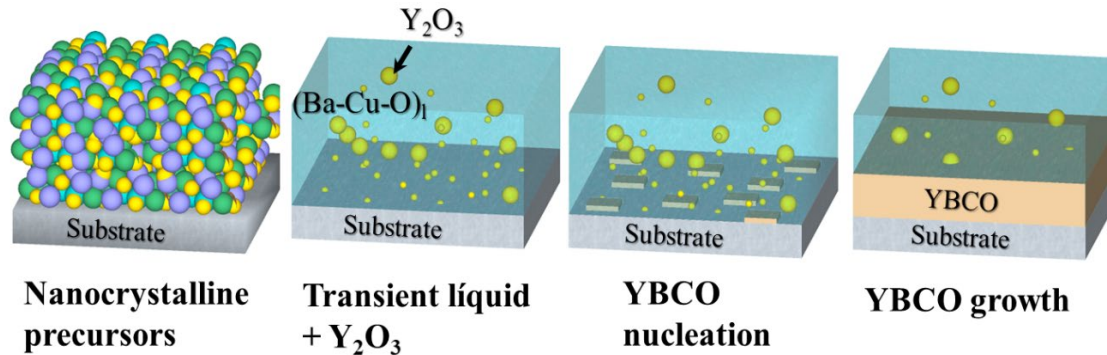


# 1. TLAG-CSD process

An opportunity for high-throughput, scalable and low cost production of Coated Conductors at **ultrafast growth rate**



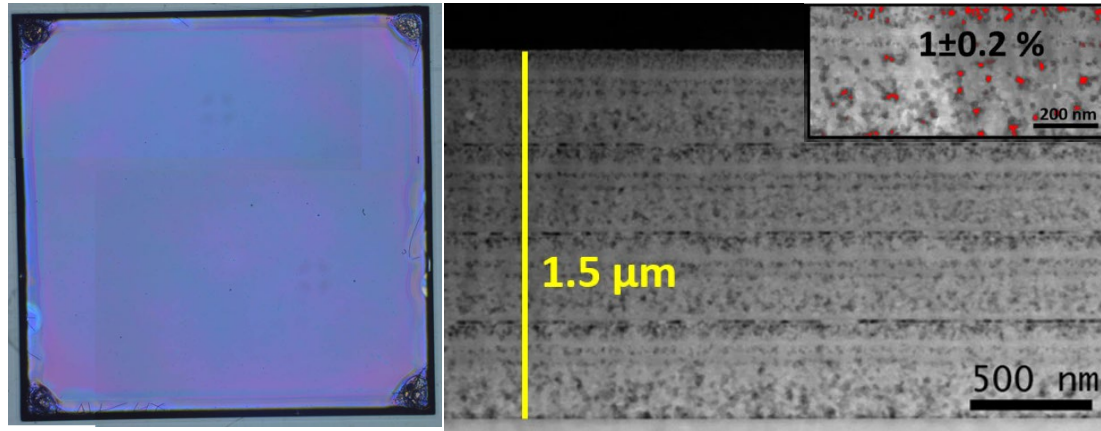
Propionate based metalorganic solution



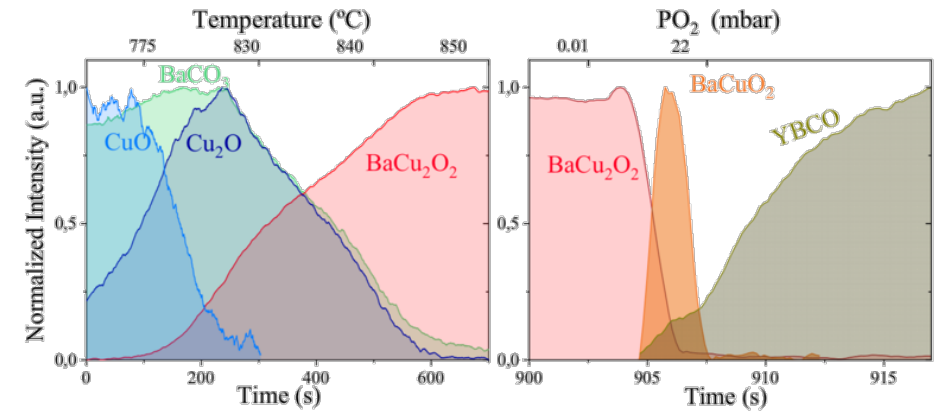
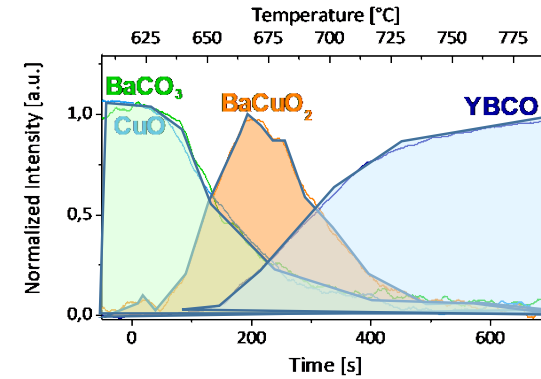
- Non-equilibrium process kinetically controlled by supersaturation reaching ultrafast growth rates 100 nm/s – 1000 nm/s from a **transient liquid-solid reaction**
- **Supersaturation** is the driving force for nucleation, controls liquid properties, substrate wettability and c-axis window. It can be modified by: Ba-Cu-O liquid composition, RE solubility, T and PO<sub>2</sub> ramps and (T, PO<sub>2</sub>) values reached
- Environmentally friendly **fluorine free precursors** based on BaCO<sub>3</sub>- CuO reaction
- Well-matched with **large area** deposition, simple and large area reactor of low cost investment

L. Soler et al., Nat Comm (2020), S. Rasi et al., J. Phys Chem C (2020), A. Queralto et al., ACS Appl. Mater. Interfaces (2021)

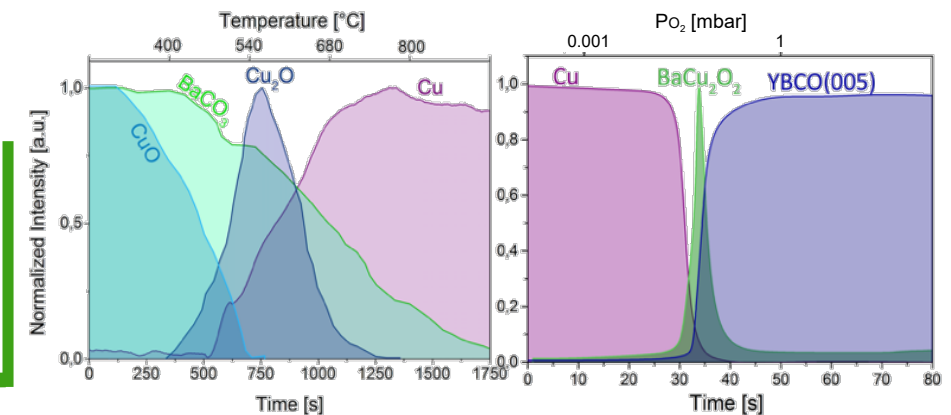
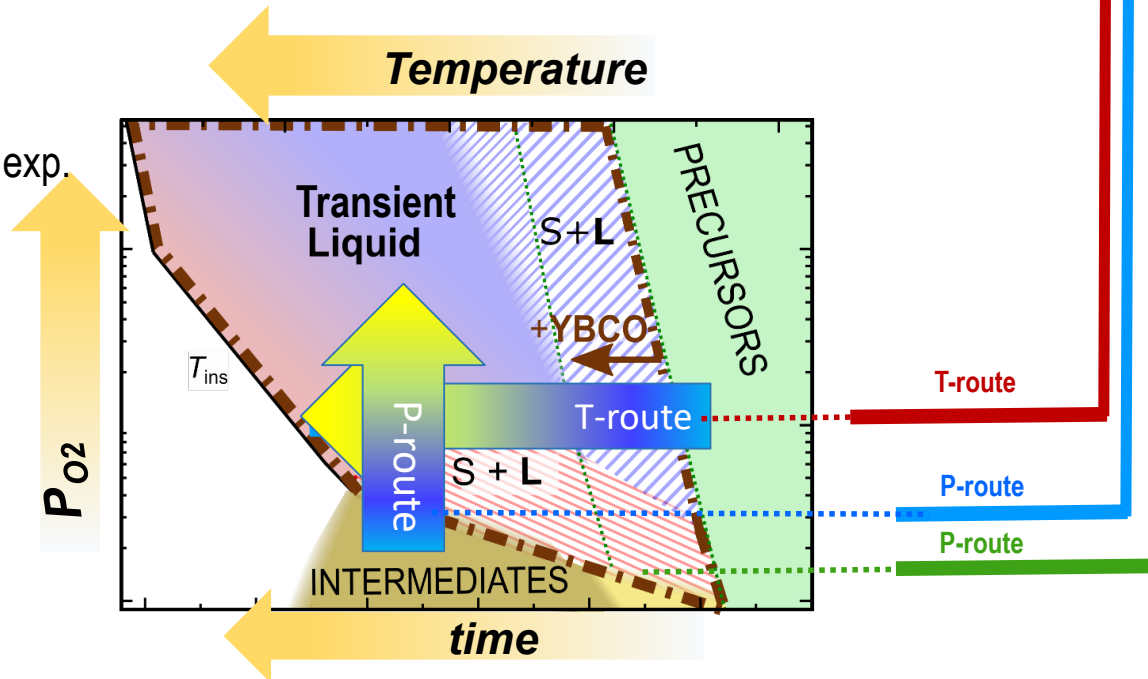
# Pyrolyzed films and intermediate phases



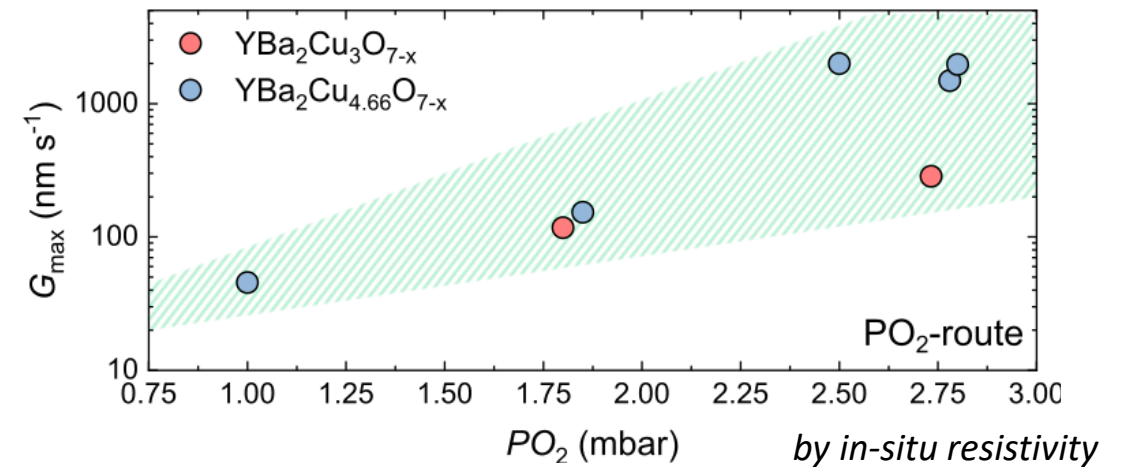
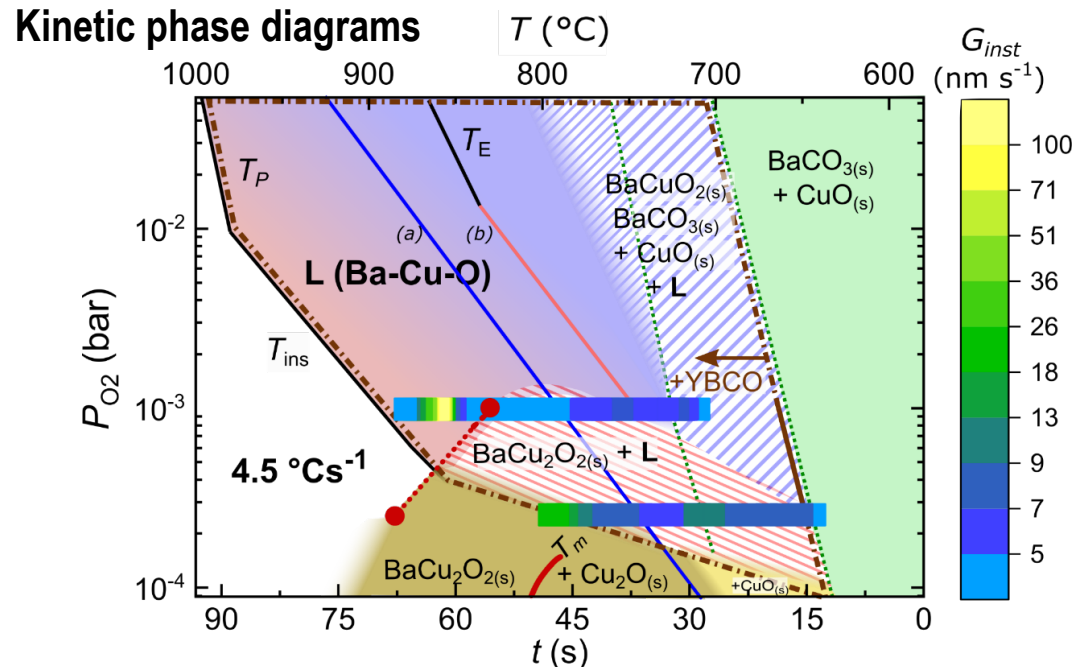
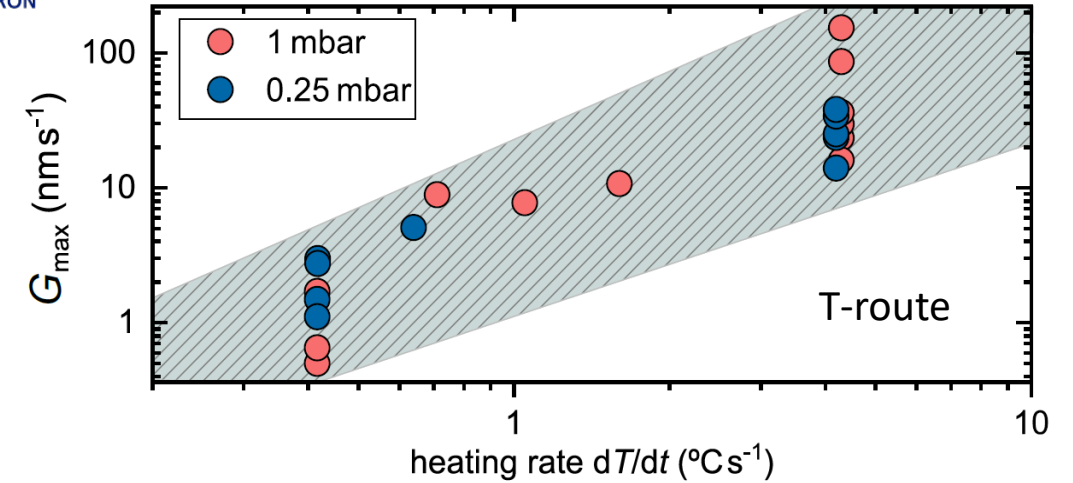
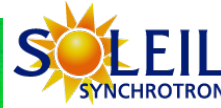
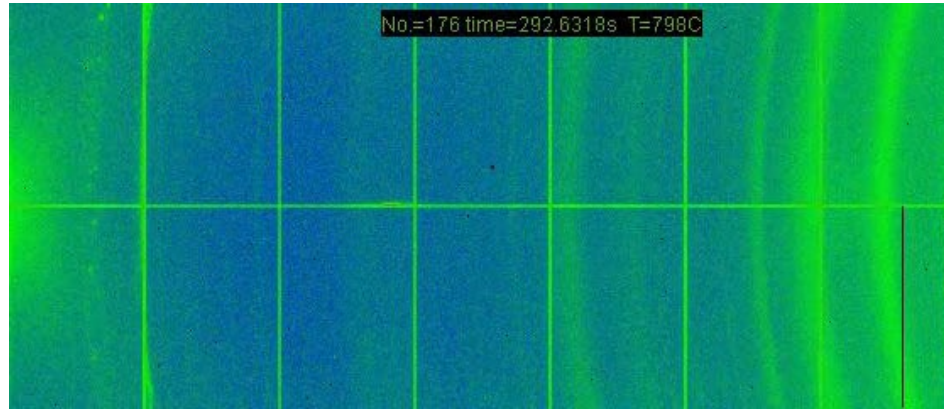
BaCO<sub>3</sub> decomposition verified in 2.5 μm thick pyrolyzed layers



In-situ XRD  
synchrotron exp.

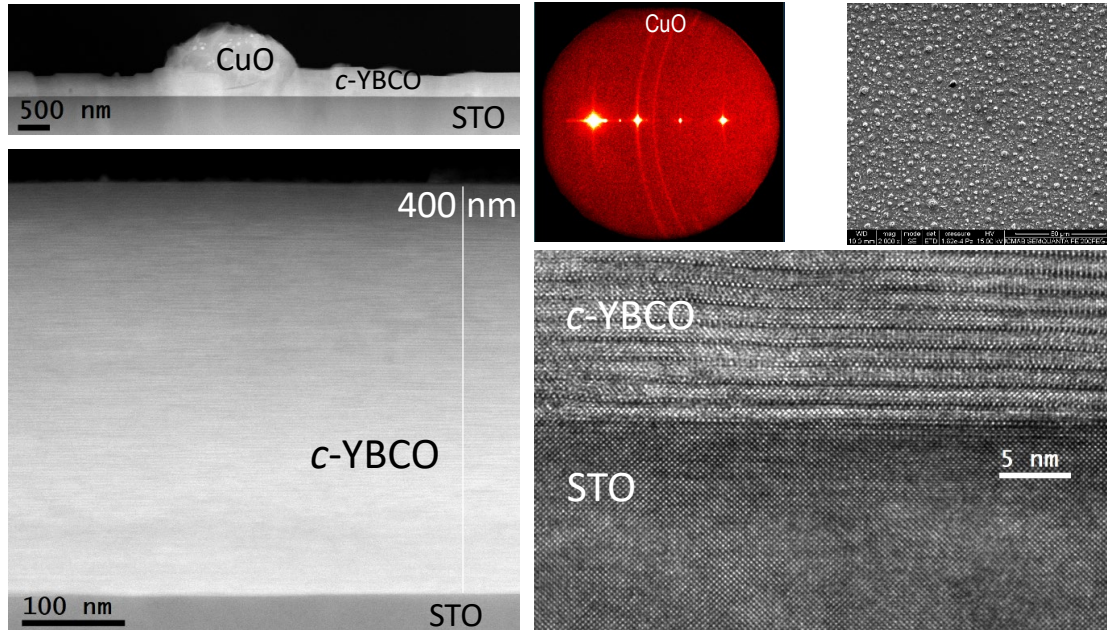


# Ultrafast (non-equilibrium) growth by TLAG-CSD



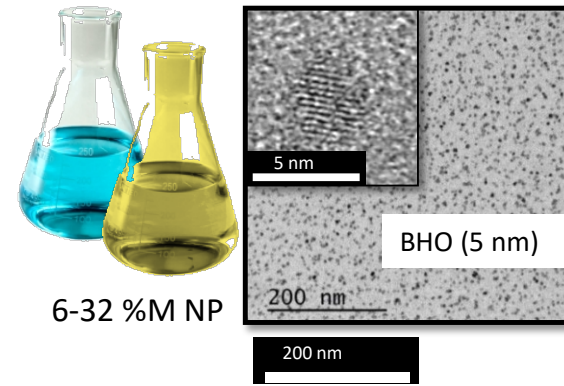


# TLAG-CSD films properties

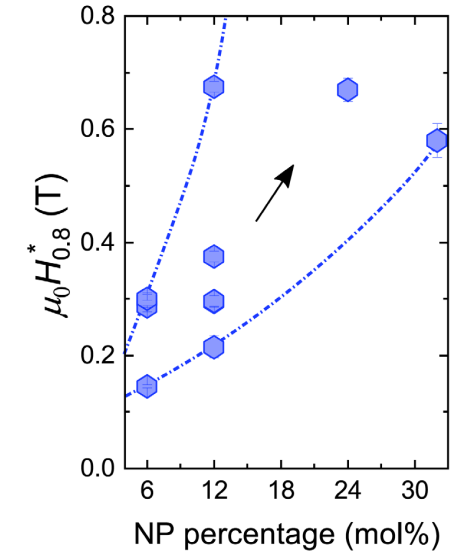
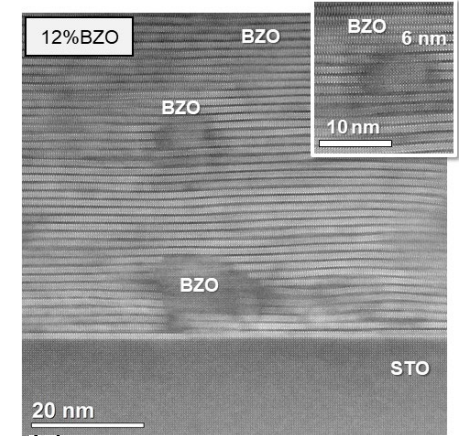
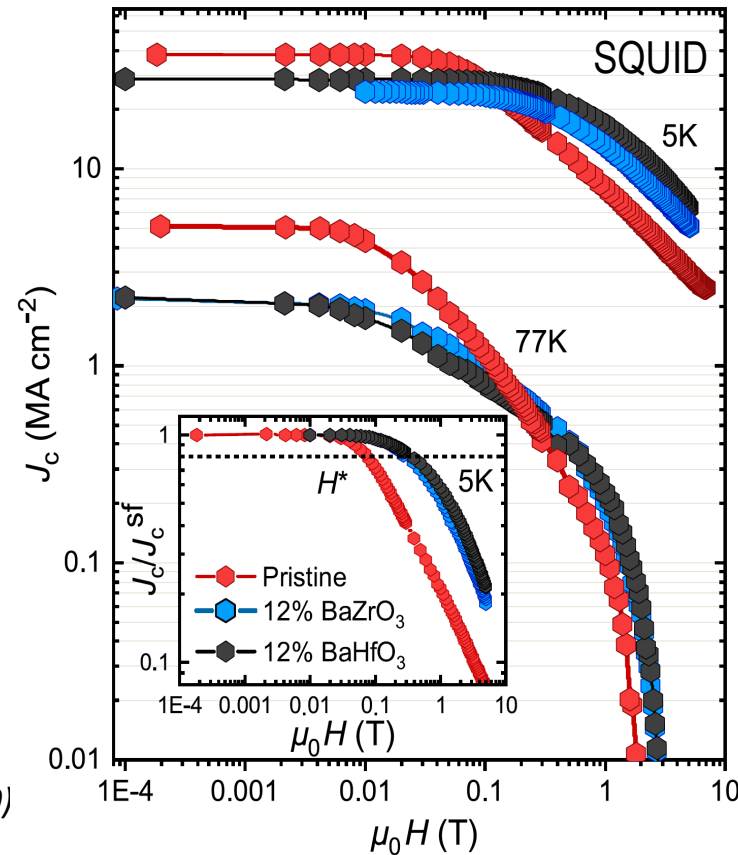


$J_c$  (77K, sf) = 2-5 MA/cm<sup>2</sup> and higher in-field performance in TLAG nanocomposite

*L. Soler et al, Nature Communications (2020), N. Chamorro, RSC Adv (2020)*  
*L. Saltarelli et al (to be published), J. Banchewski et al, to be published*



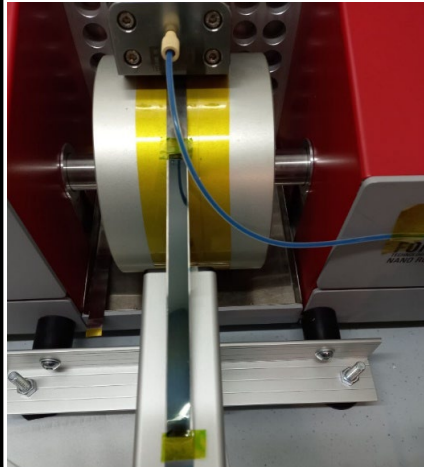
6-32 %M NP



# TLAG Coated Conductors

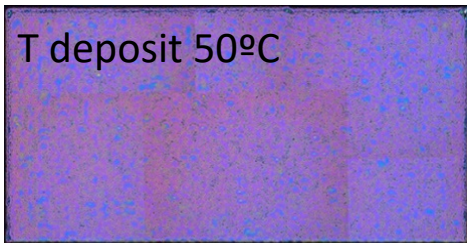
## Slot-die coating deposition

15 cm, 1  $\mu\text{m}$  pyrolyzed film (2 layers)



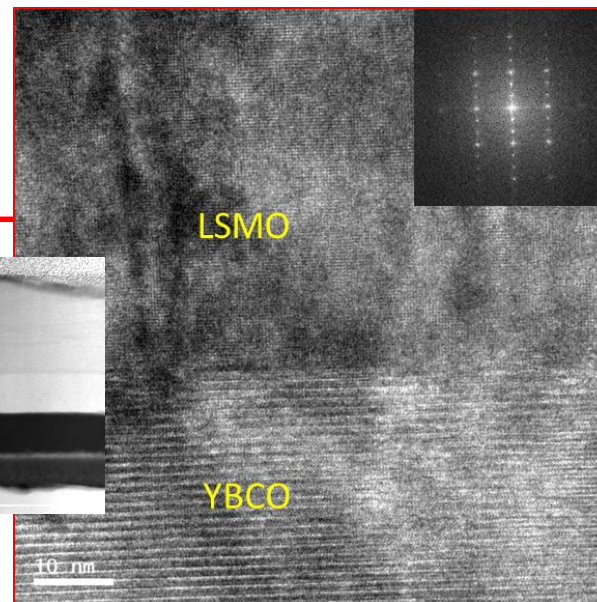
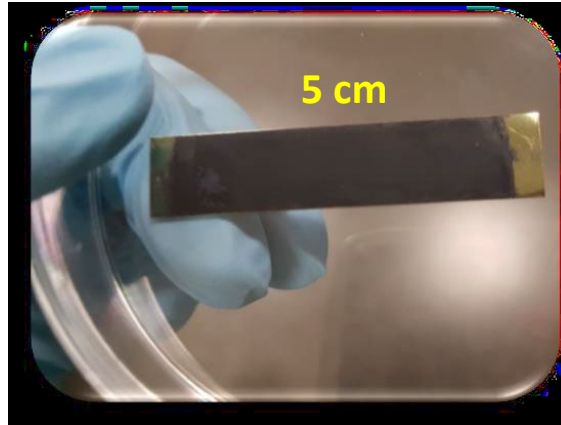
## IJP deposition

1  $\mu\text{m}$  pyrolyzed film (1 layer)

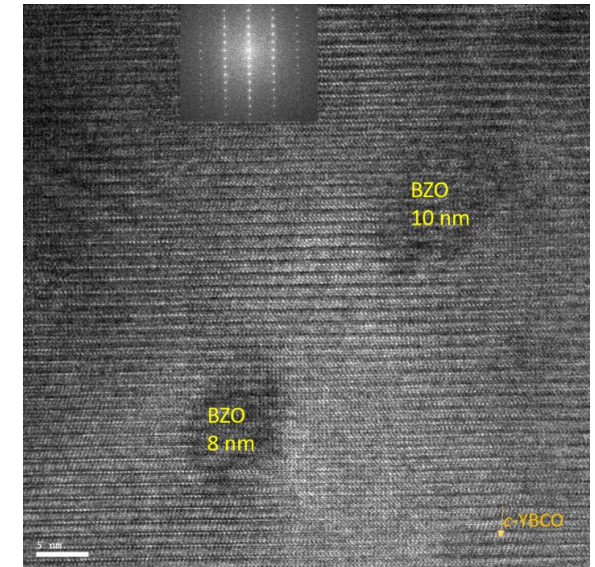


- Homogeneous depositions and pyrolysis

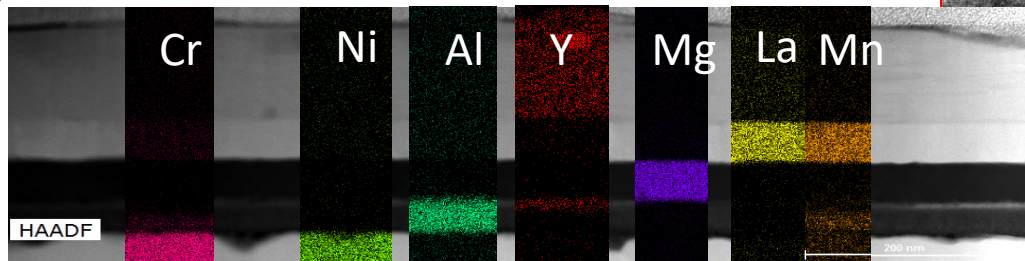
## TLAG CC growth on provider 1 (5 cm long tapes tested)- P-route



## TLAG CC growth of nanocomposites provider 2 – T-route



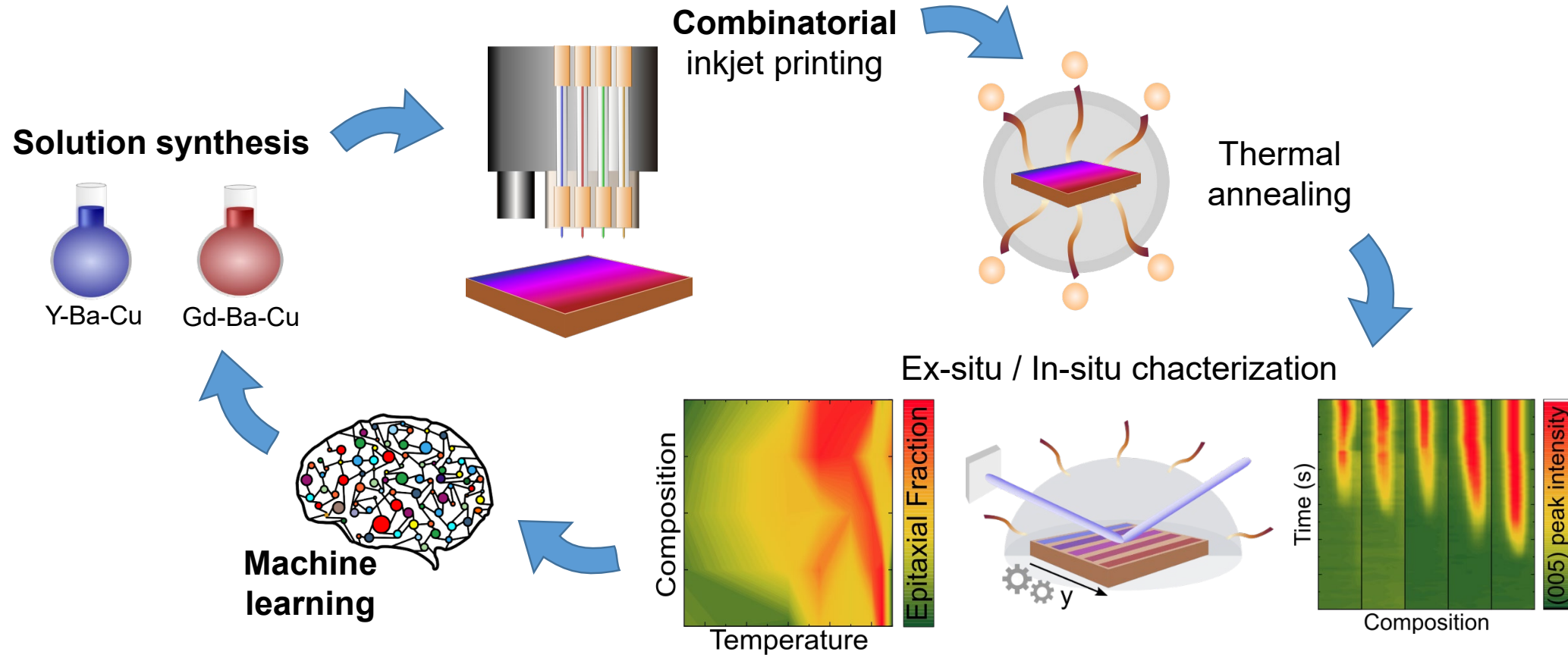
- Small NPs (8-10 nm)
- Epitaxial and dispersed BZO NPs



- Smooth interfaces
- $J_c \sim 1 \text{ MA/cm}^2$  at 77 K for 500 nm



## 2. High Throughput Experimentation by combinatorial approaches and Machine Learning algorithms



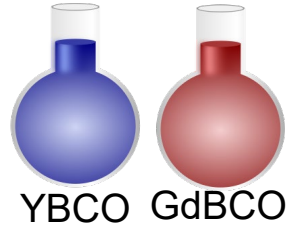




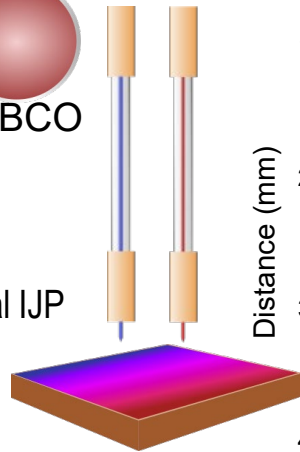
# Fast optimization using compositional gradient samples



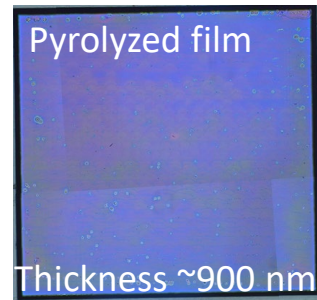
Solution synthesis



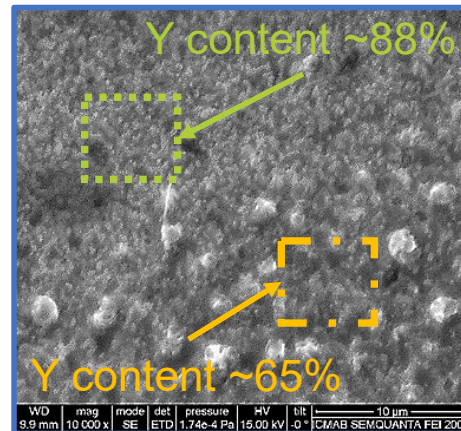
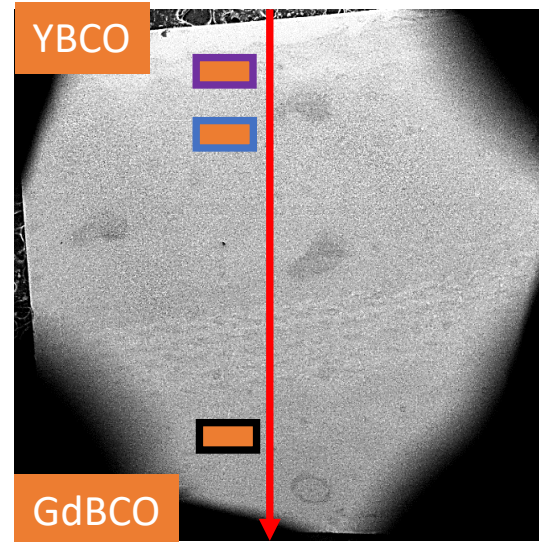
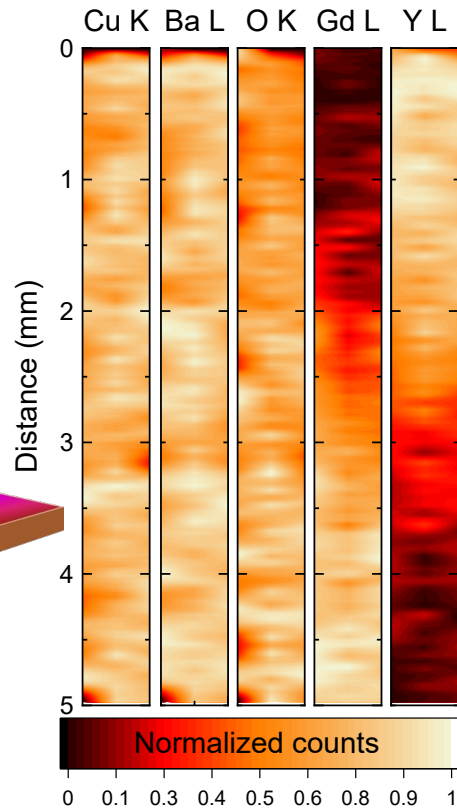
Combinatorial IJP



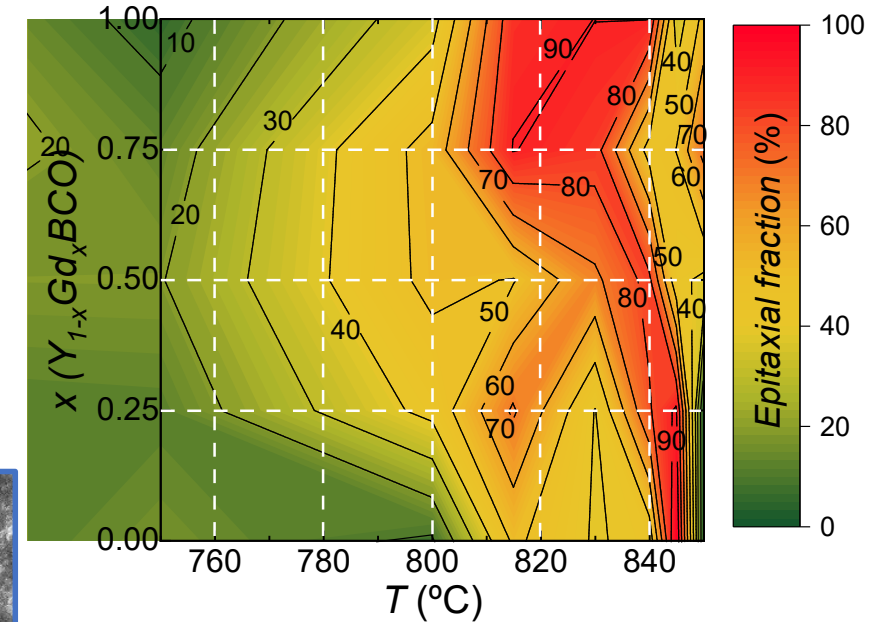
YBCO (0 mm)



GdBCO (5 mm)



Morphological changes



13 combinatorial samples were used instead of 78 regular samples to reach the same mapping resolution



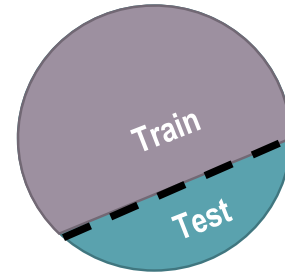
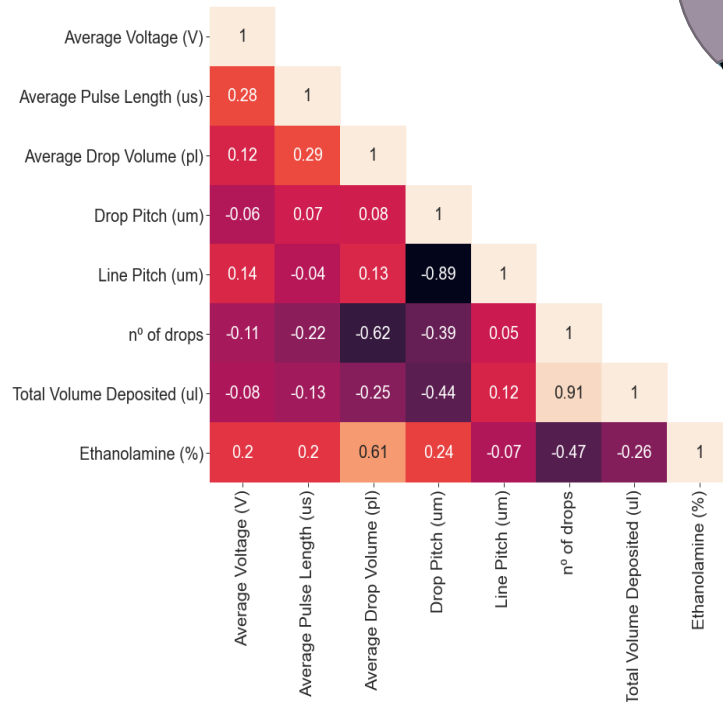
# Machine learning for predicting process magnitudes



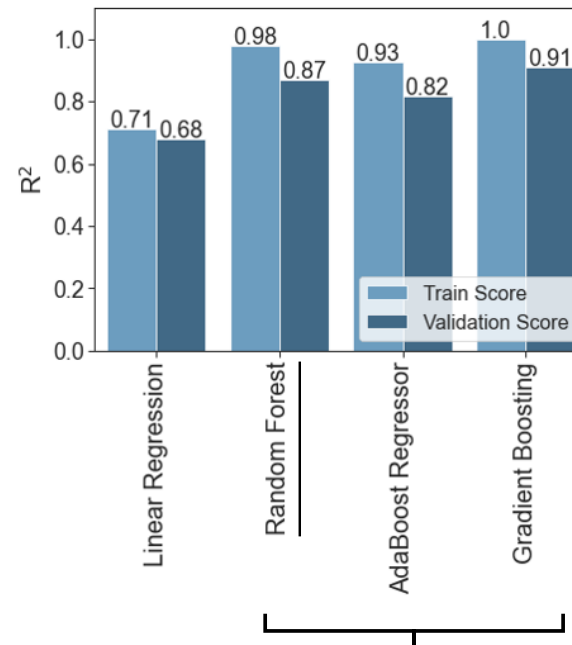
## Case: Predicting the Total Volume Deposited

IJP deposition data of 231 samples

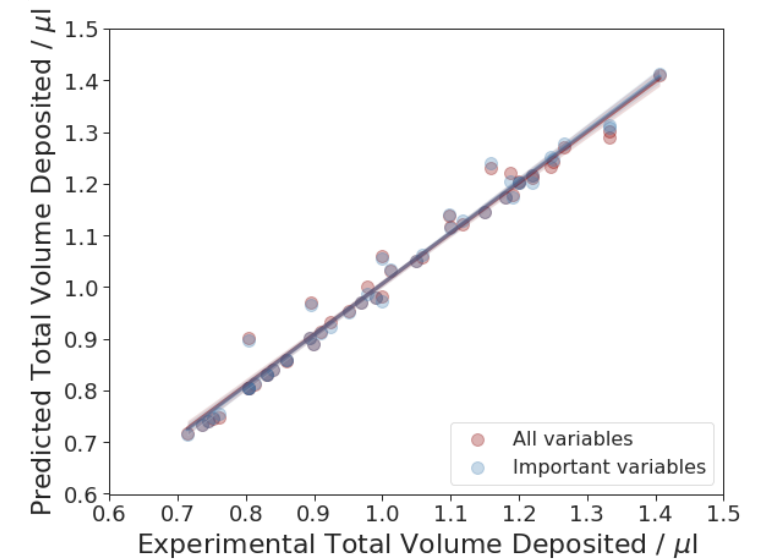
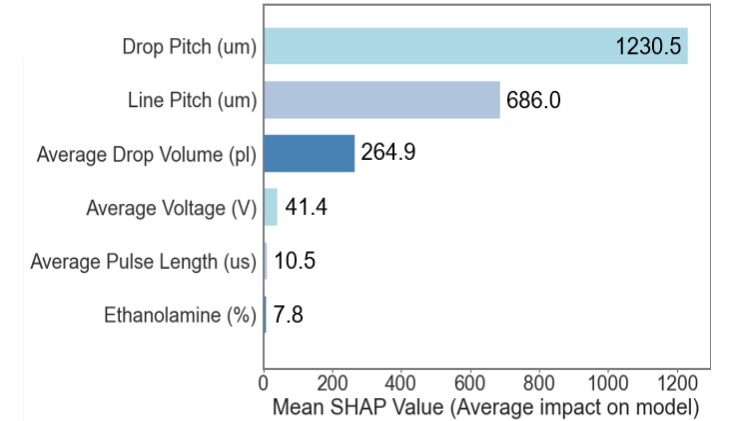
Correlation matrix



train/test splits  
80/20

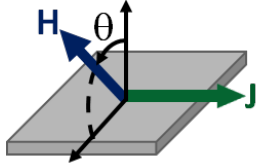


Tree-based ensemble algorithms



A Queraltó et al, to be published

# 3. Understanding the (H,T,θ) pinning landscape for engineered nanocomposites

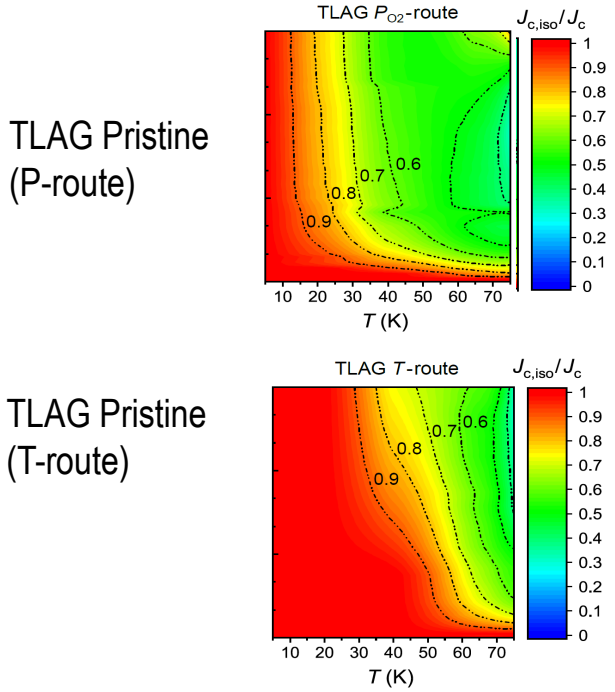
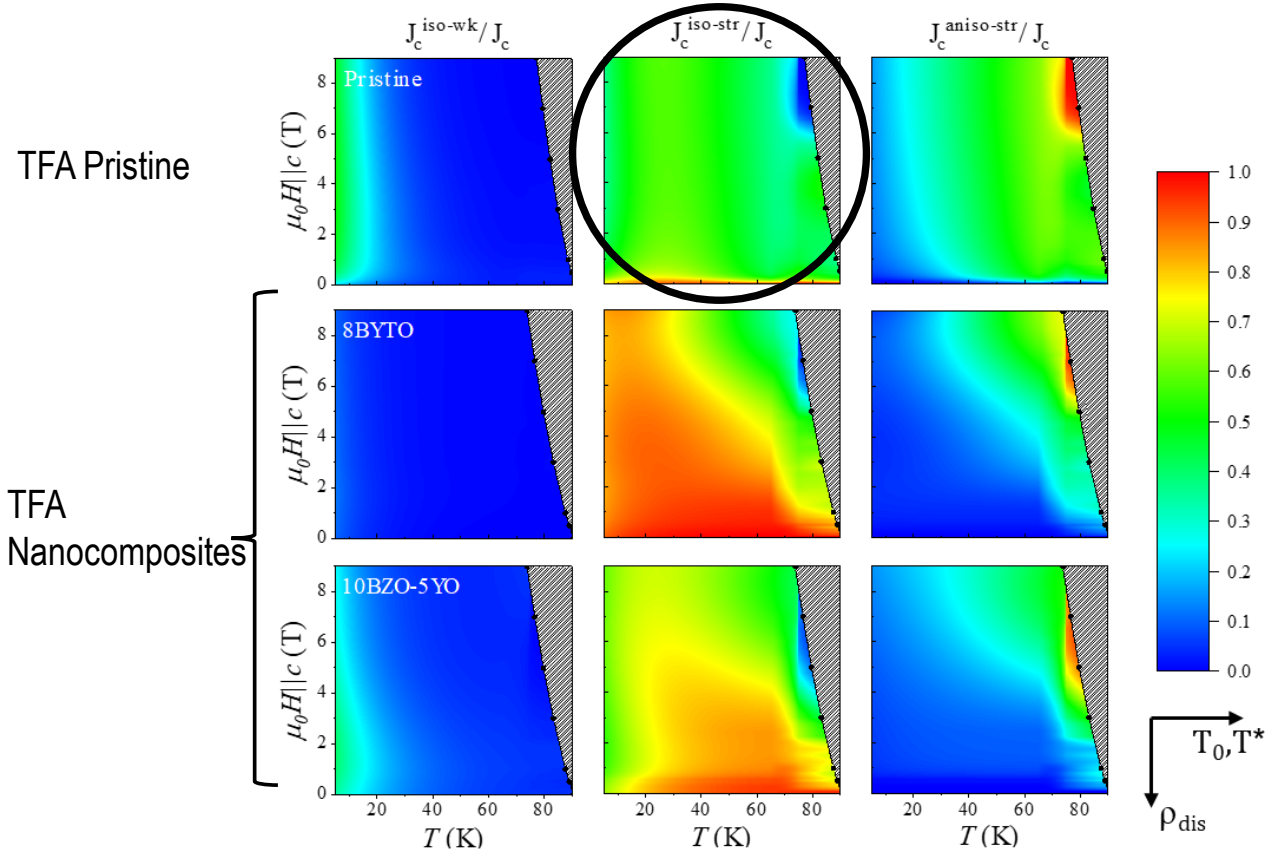


$$J_c(T) = J_c^{iso-wk}(T) + J_c^{iso-str}(T) + J_c^{aniso-str}(T)$$

- $J_c^{iso-wk}/J_c$
- $J_c^{iso-str}/J_c$
- $J_c^{aniso-str}/J_c$



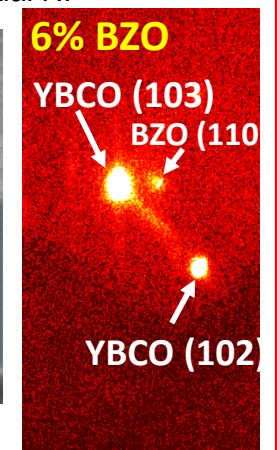
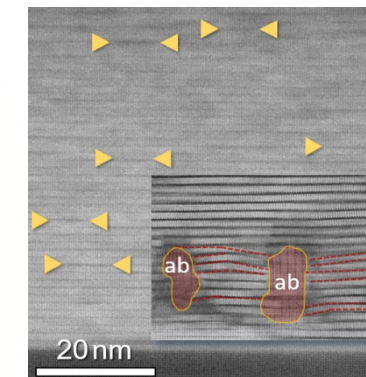
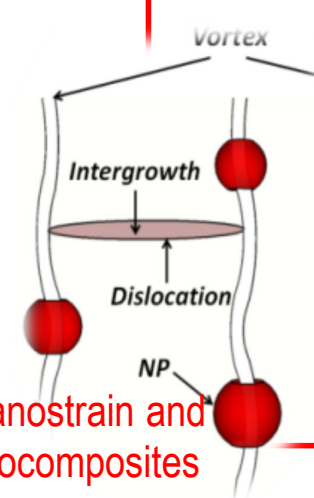
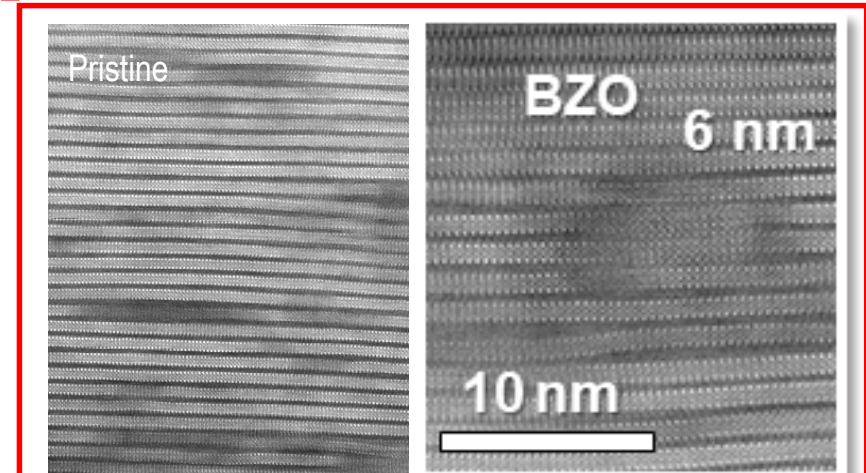
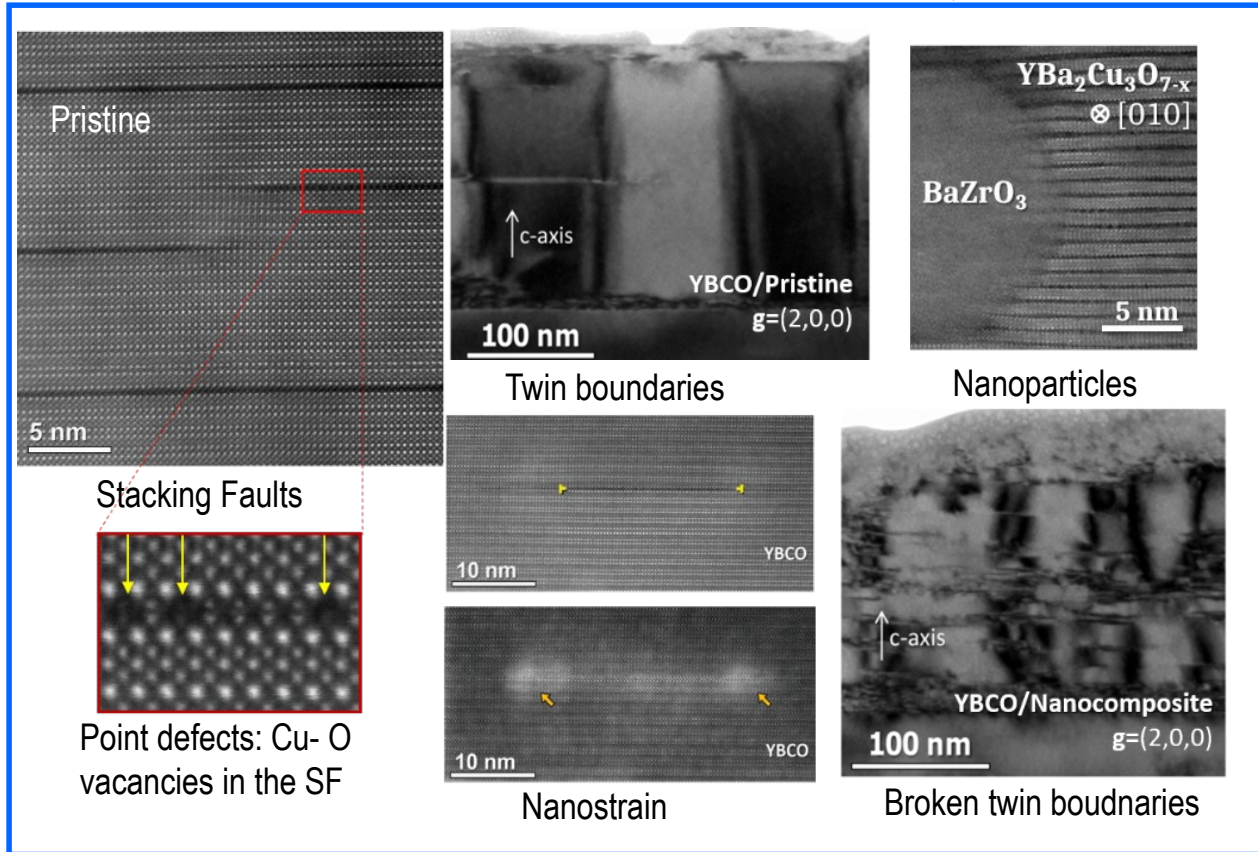
- defect dimensionality
- orientation
- size



J. Banchewski et al, to be published

Lot's of opportunities for TLAG Nanocomposites

# Vortex pinning defects in TFA/TLAG - CSD films



**Synergistic effect of nanostrain and small NP in TLAG nanocomposites**

J. Gutierrez et al, Nat Mat (2007)  
 A. Lordés et al, Nat Mat (2012)  
 A. Palau et al., SUST (2018)  
 Z. Li et al, Sci Rep. (2019)  
 L. Soler et al, Nat Comm (2020)

J. Gazquez et al., Adv. Sci. (2016)  
 R. Guzman et al, APLMat (2017)  
 S.T. Hartman et al, PRMat (2019)



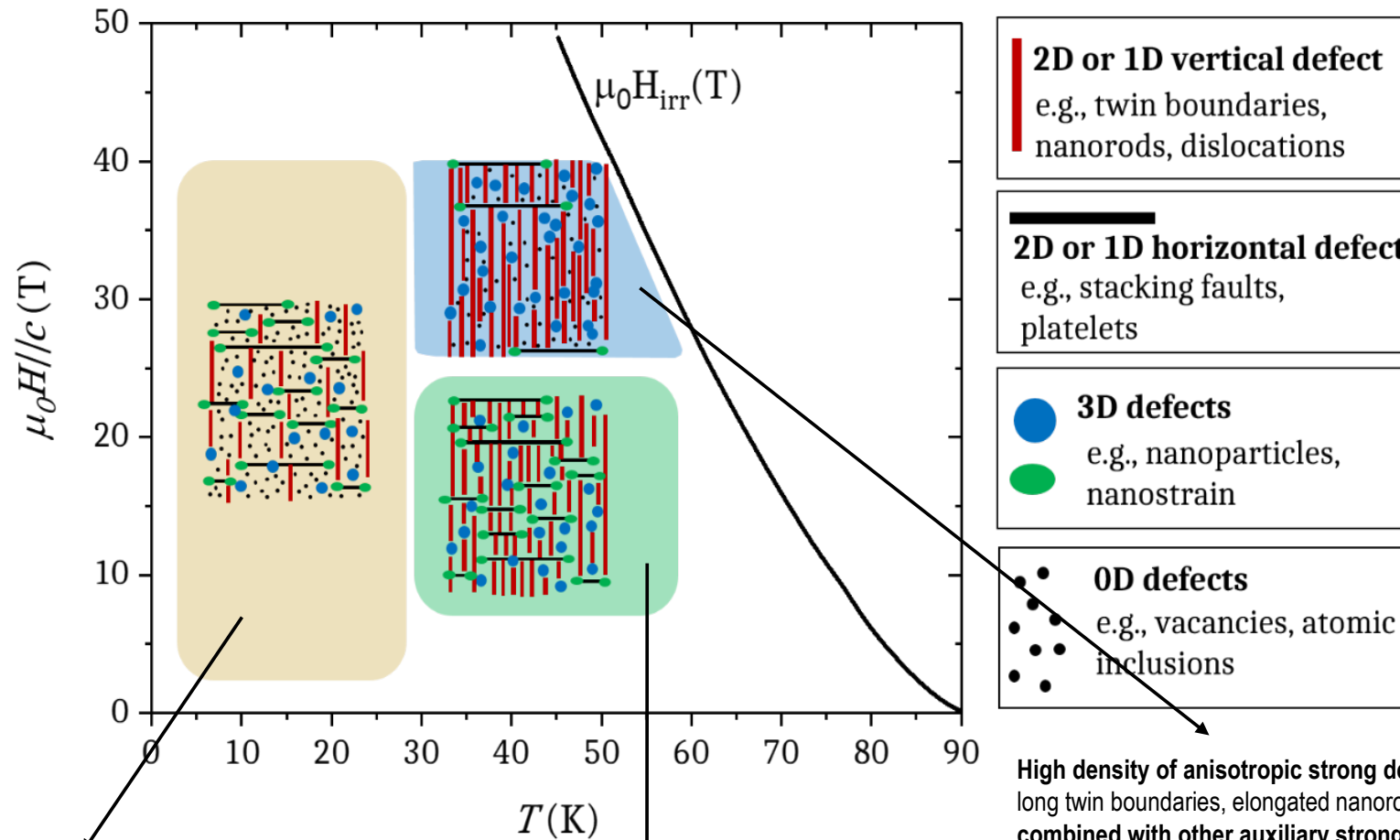
# Optimized pinning landscapes

Low and intermediate T - Intermediate and high H

14 years of measurements



*D. Abraimov*  
*J. Jaroszynski*  
*D. Larbalestier*



**High density of isotropic defects:**  
Cu-O vacancies, short SF, small NP  
*(better Np than nanorods)*

**Large density of strong isotropic and anisotropic defects with long vertical coherence:** nanoparticles, nanostrain, long nanorods, long twin boundaries  
*(Nanorods and nanoparticles will add effects)*

**2D or 1D vertical defect**  
e.g., twin boundaries, nanorods, dislocations

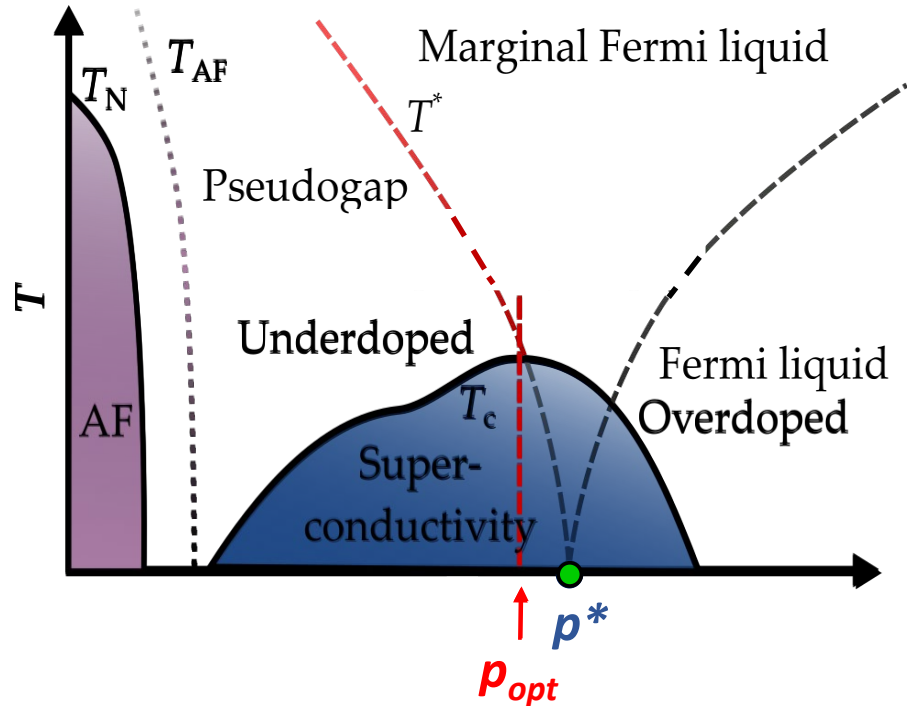
**2D or 1D horizontal defect**  
e.g., stacking faults, platelets

**3D defects**  
e.g., nanoparticles, nanostrain

**0D defects**  
e.g., vacancies, atomic inclusions

**High density of anisotropic strong defects with very long vertical coherence:** long twin boundaries, elongated nanorods, thick CSD nanocomposites  
**combined with other auxiliary strong or weak isotropic defects to lessen vortex creep excitations**  
*(1D-2D mandatory but all defects will help to diminish creep)*

# 4. Tune charge carrier density by oxygen overdoping



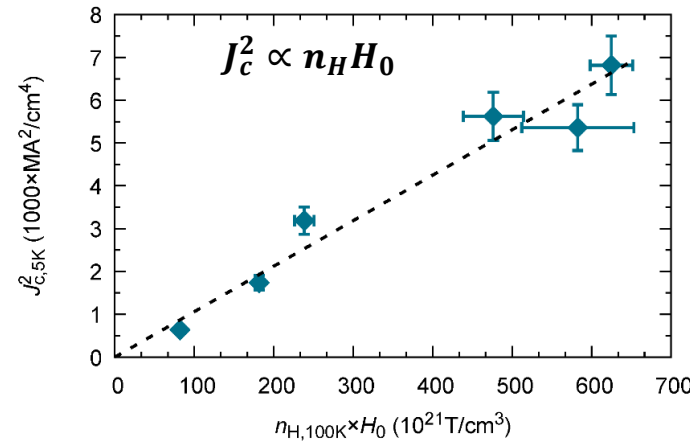
$p_{opt} = 0.16$  holes/ $\text{CuO}_2$ -plane: optimal doping for maximum  $T_c$   
 $p^* = 0.19$  holes/ $\text{CuO}_2$ -plane: Critical doping (QCP)

$$\text{Pinning force } F_p = \sum_i^{N_p} f_{p,i}(B, T) \propto J_c$$

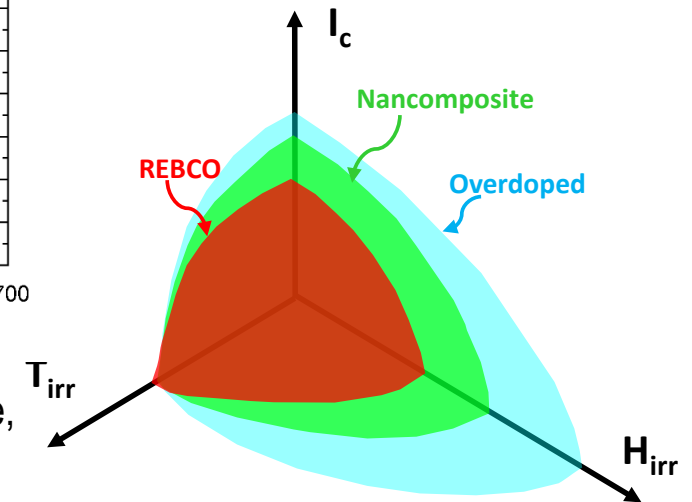
$$f_p \propto E_c \text{ condensation energy}$$

$$J_d^2 \propto n_s E_c \longrightarrow J_c^2 \propto n_H H_0 \text{ (} H_0 \text{ from in-plane magnetoresistance)}$$

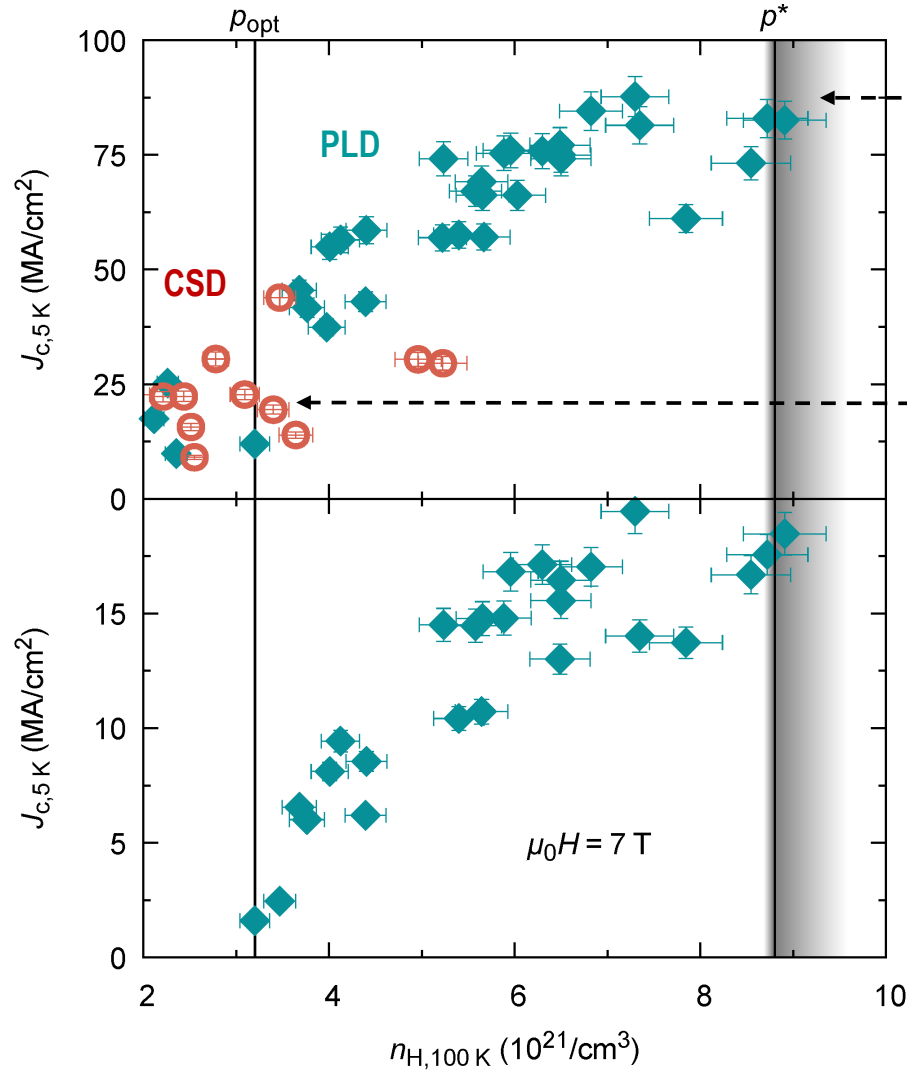
(three independent experimental parameters)



$n_H$  and  $E_c$  increases in the overdoped state, and consequently  $J_c$  should increase



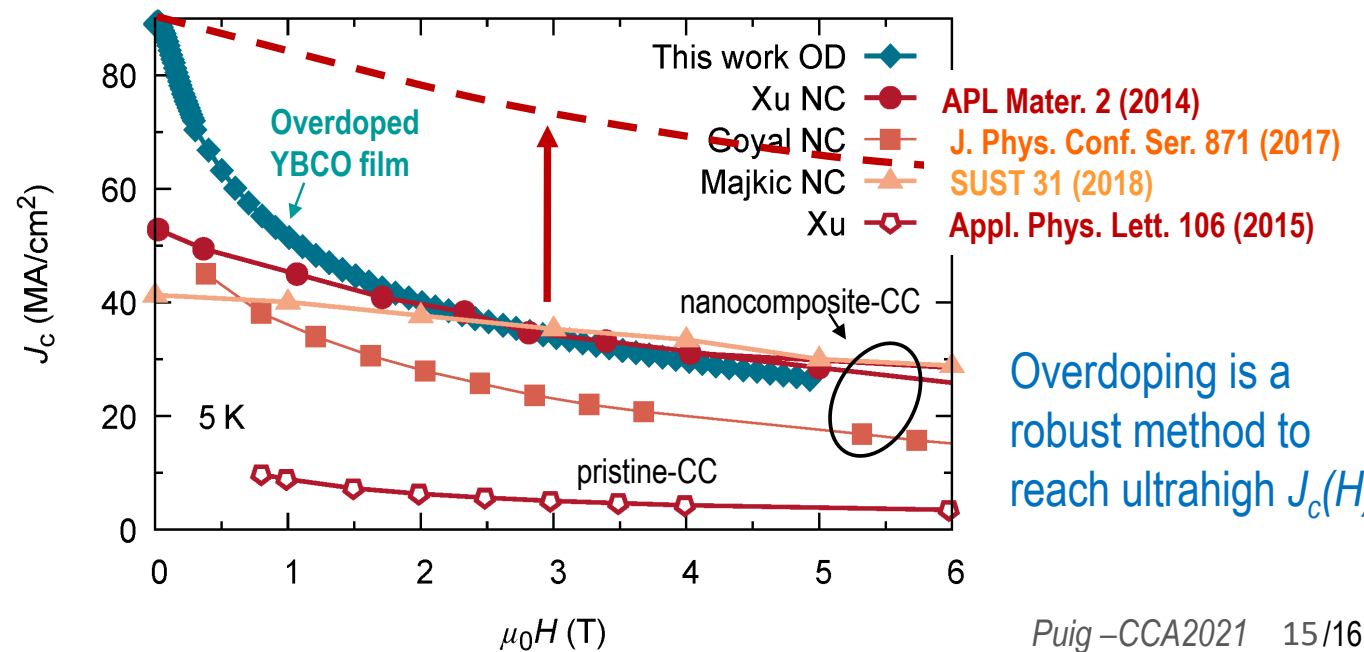
# Strong increase of $J_c$ in the overdoped state



$$J_c(p^*) \approx \frac{1}{5} J_d(p^*) = 90 \text{ MA/cm}^2 \quad J_d(p^*) \approx 500 \text{ MA/cm}^2$$

Strong increase of  $J_c$  with  $n_H$   
 (x4 from  $p_{opt}$  to  $p^*$ )

$$J_c(p^{opt}) \approx \frac{1}{10} J_d(p^{opt}) \quad J_d(p^{opt}) \approx 330 \text{ MA/cm}^2$$



Overdoping is a robust method to reach ultrahigh  $J_c(H)$

# CONCLUSIONS



- Material science should help Coated Conductors to reach the market
- Cost / Performance ratio needs to decrease and high-throughput low cost growth processes with high critical current strategies can be employed
- TLAG-CSD is a non-equilibrium growth method able to grow high performance YBCO nanocomposites using low cost CSD methods at ultrafast growth rates
- Nanoengineering combinations of different defects should be used to optimize pinning landscape for each application
- Overdoping can be a robust and feasible way to further increase  $J_c$  in nanoengineered Coated Conductors