Work is supported by

In-situ growth of superconducting films obtained at the ultrahigh growth rates (100 nm/s) of the transient liquid assisted growth process

Teresa Puig, Xavier Obradors

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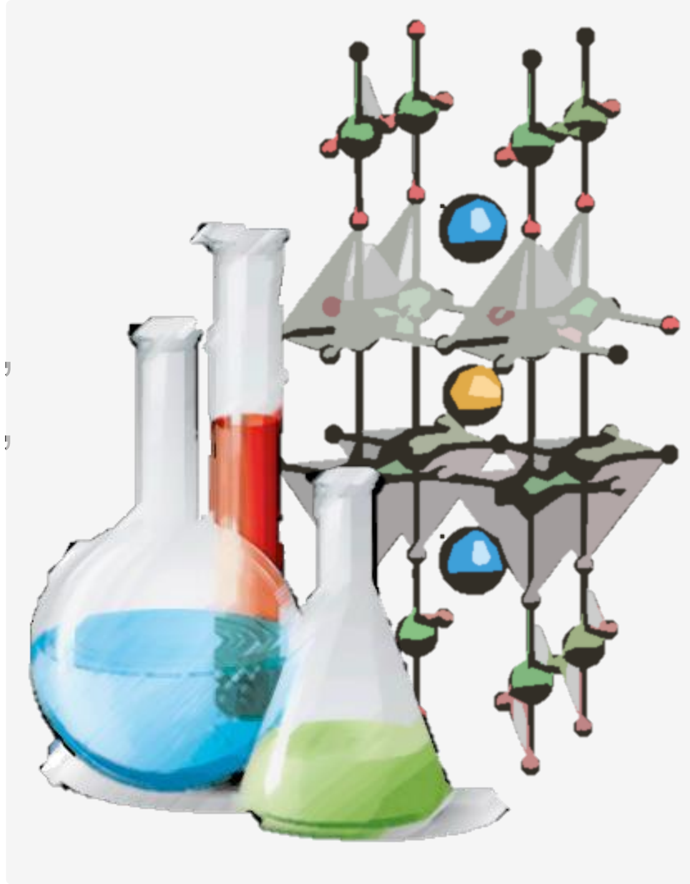


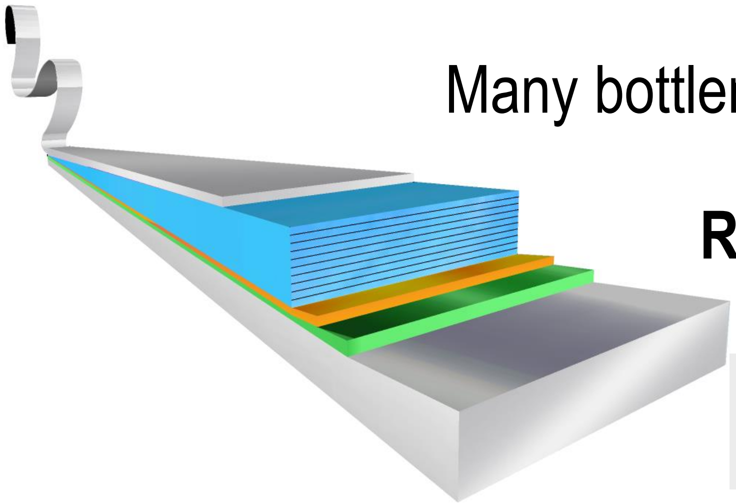
25-yr of Coated Conductor technology development

INSTITUT DE CIÈNCIA DE MATERIALS DE BARCELONA

EXCELENCIA SEVERO OCHOA

CSIC

CONSEJO SUPERIOR DE INVESTIGACIONES CIENTÍFICAS



Many bottleneckts have been overcome

REBCO is a true opportunity at high-T but also at high-H & low-T

I_c (77K, sf) ~ 500 A/cm-w (for 500 m -1 km piece length)
I_c (4.2 K, 20 T) ~ 1000 A/cm-w (with nanocomposites)

CHEAPER ← **Need to decrease cost** → **BETTER**

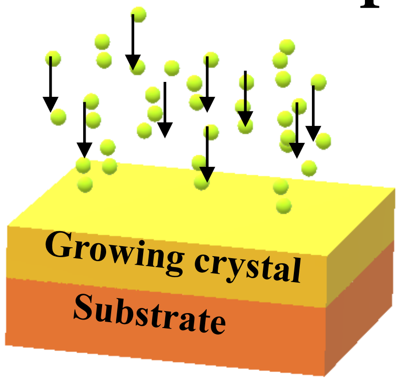
€ / kA m

- **Fast growth**
- **Simple processing and architecture**
- **High yield and low capital investment**

- **High performance J_c(B, T) Nanocomposites**
- **Thick and mechanically robust REBCO films**
- **Thin substrates (J_E)**
- **Customization**

Growth mechanisms for epitaxial REBCO films crystallization

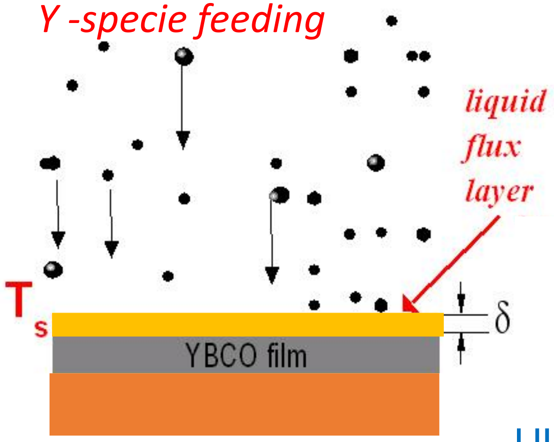
Growth from **vapour** phase



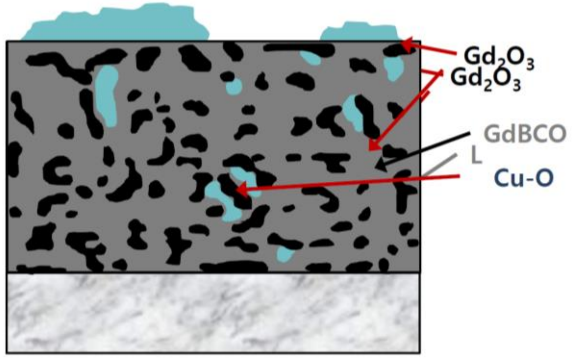
- PLD
- MOCVD
- Evaporation

PLD : 5-12 nm/s (Superox), 14 nm/s (SST)
A-MOCVD:3 nm/s (U. Houston), Evapor.: 4 nm/s (THEVA)

Growth from high temperature supersaturated **equilibrium liquids**



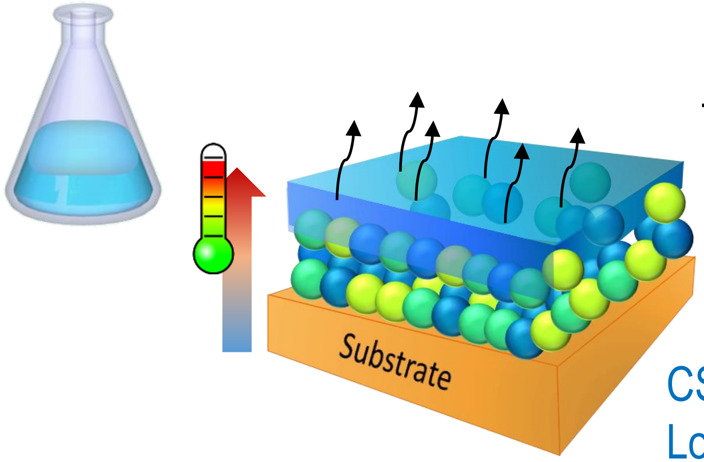
- HLPE, VLS, TPE



-RCE-DR (ex-situ)

HLPE: ~10 nm/s (UCAM)
RCE-DR: > 30 nm/s (SUNAM)

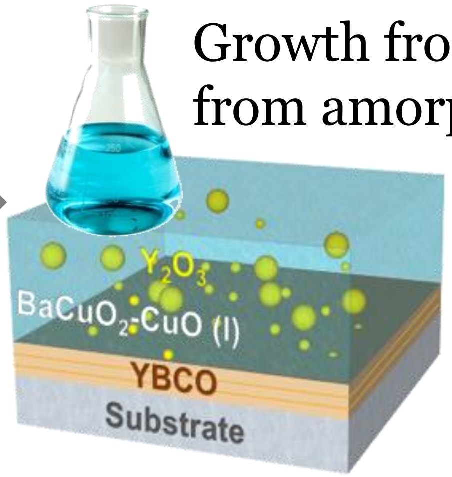
Growth from **amorphous precursor solids**



- CSD (ex-situ)

CSD : ~1 nm/s
Low cost

Growth from **transient liquids** derived from amorphous precursor solids

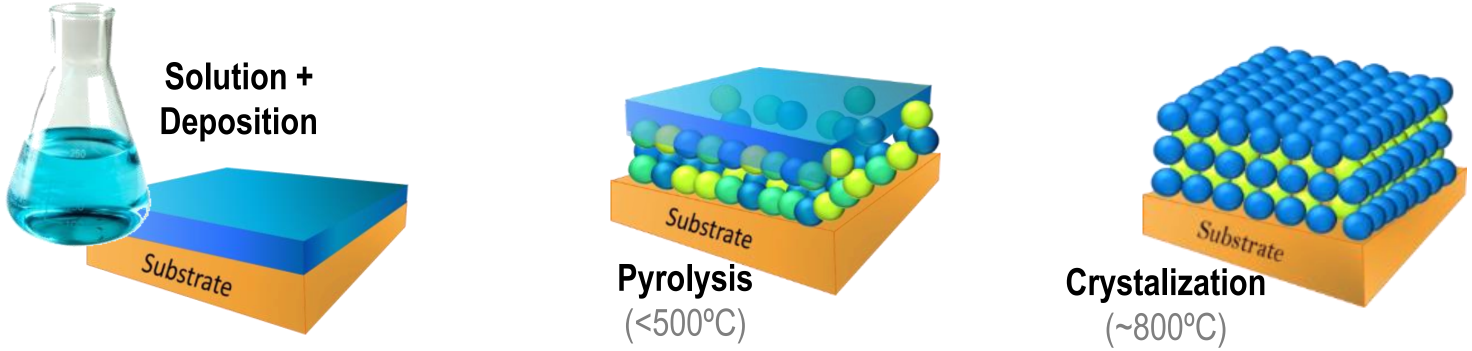


- TLAG-CSD (ICMAB)

Growth rates from liquids : ~100 nm/s
Scalable and low cost

L. Soler et al, Nature Communications (2020)

YBCO-CSD Growth methods

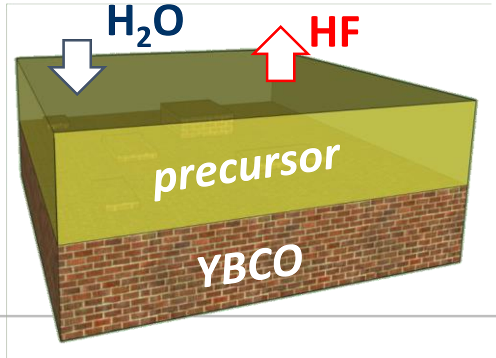


Decoupled processes

TFA- route

Trifluoroacetate based precursors

$Ba_{1-x}Y_xF_{2-y}O_y$ + CuO + Y_2O_3

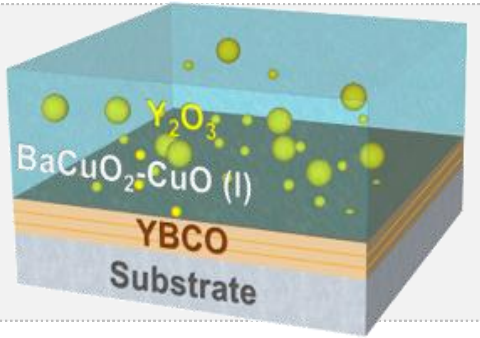


- ❖ *Gas-solid conversion reaction*
- ❖ *Slow growth rates (~1 nm/s)*
- ❖ *Slow HF removal – complex reactor*

TLAG- route

Fluorine Free based precursors

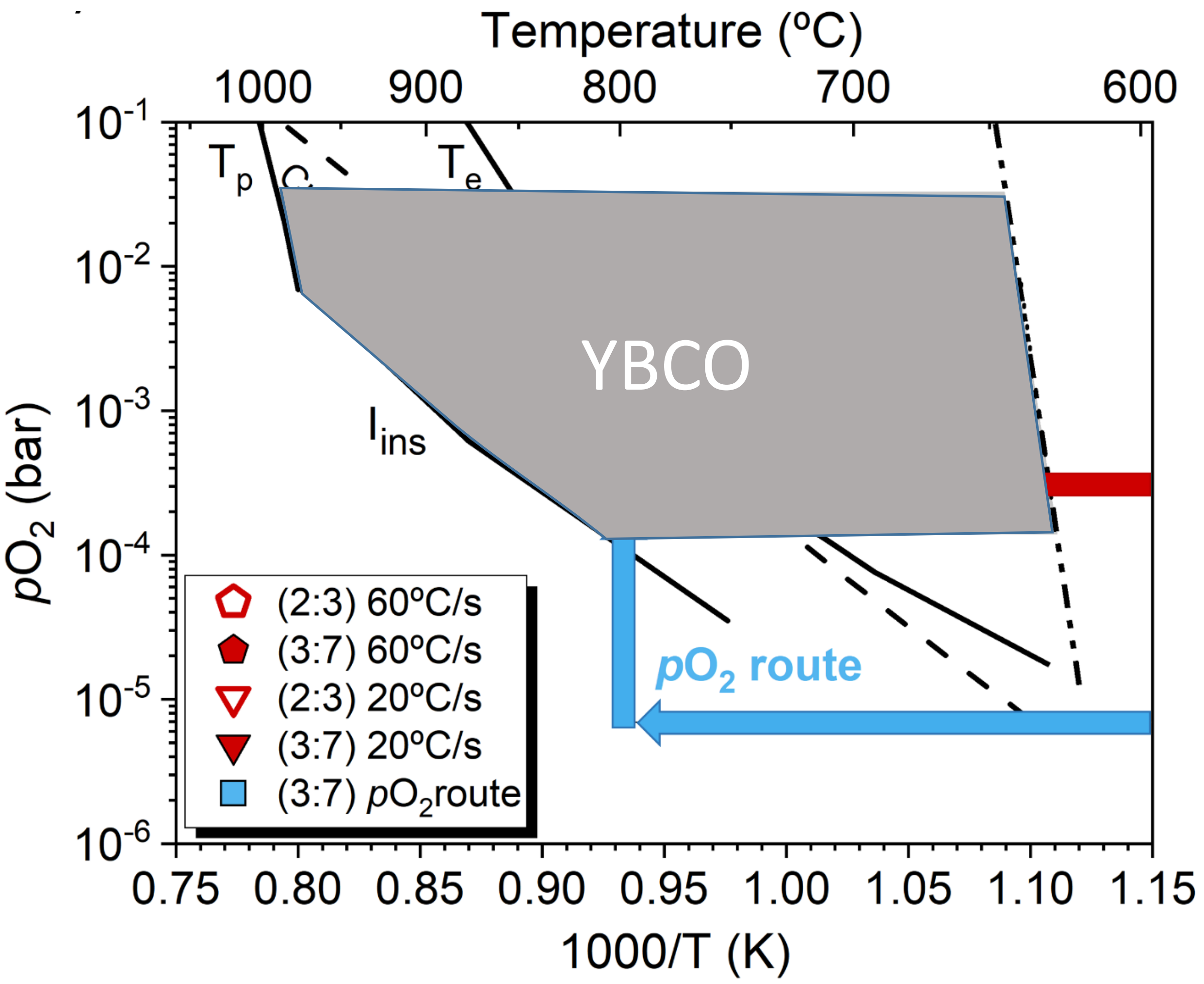
$BaCO_3$ +CuO + Y_2O_3



- ❖ *Liquid-solid conversion reaction*
- ❖ *Fast growth rates-100 nm/s*
- ❖ *Dependent on $BaCO_3$ decomposition*
- ❖ *Simple reactor –environment friendly*

TLAG = Transient Liquid Assisted Growth

Transient Liquid Assisted Growth: TLAG-CSD



$BaCO_3(s)+CuO(s)+Y_2O_3(s) \longrightarrow [Ba\text{-}Cu\text{-}O](l)$
$+Y_2O_3(s) \longrightarrow YBa_2Cu_3O_{6.5}(s)+xCuO\ (s)$

- Transient liquids (Ba-Cu-O) form much faster than the equilibrium solid phase (YBCO)
- No need of equilibrium liquid phases in the phase diagram
- **Kinetically driven non-equilibrium growth process**

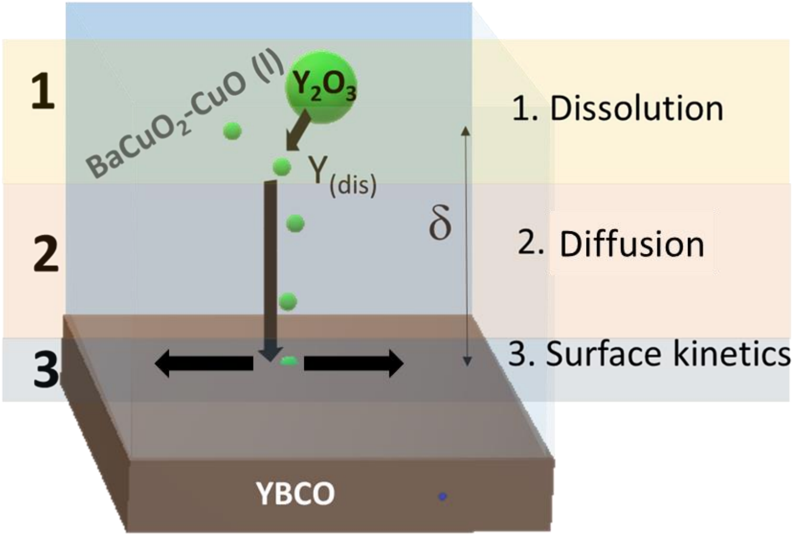
Very wide area of YBCO c-axis nucleation if heating rates > 20ºC/s (RTA furnaces)

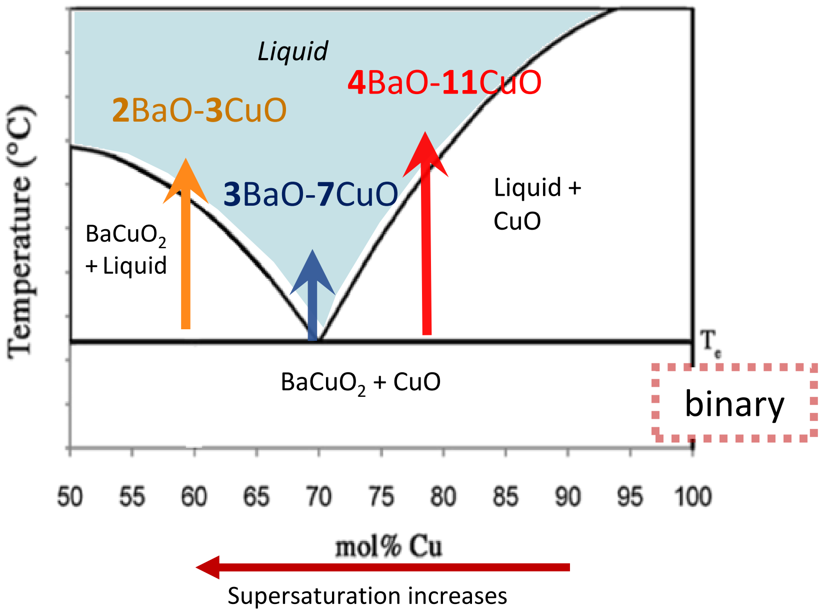
L. Soler et al, Nature Communications (2020)

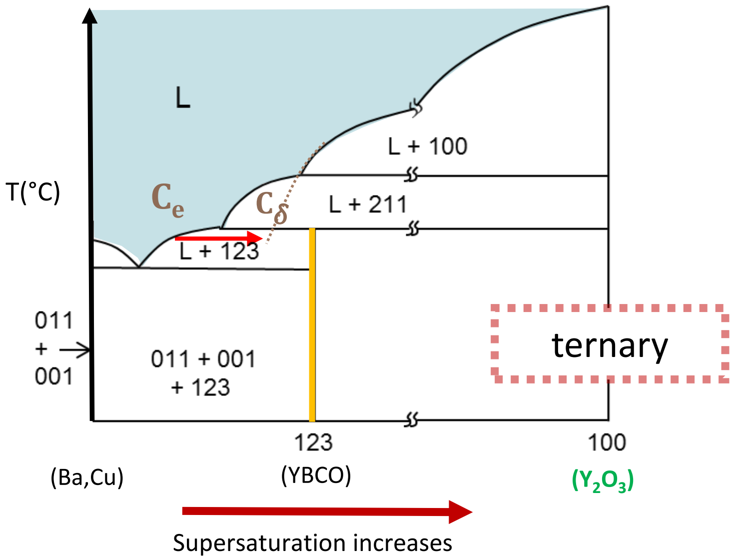
Transient Liquid Assisted Growth: TLAG-CSD

Non-equilibrium process kinetically controlled by supersaturation

$BaCO_3(s)+CuO(s)+Y_2O_3(s) \rightarrow [Ba\text{-}Cu\text{-}O](l) + Y_2O_3(s) \rightarrow YBa_2Cu_3O_{6.5}(s)+xCuO\ (s)$







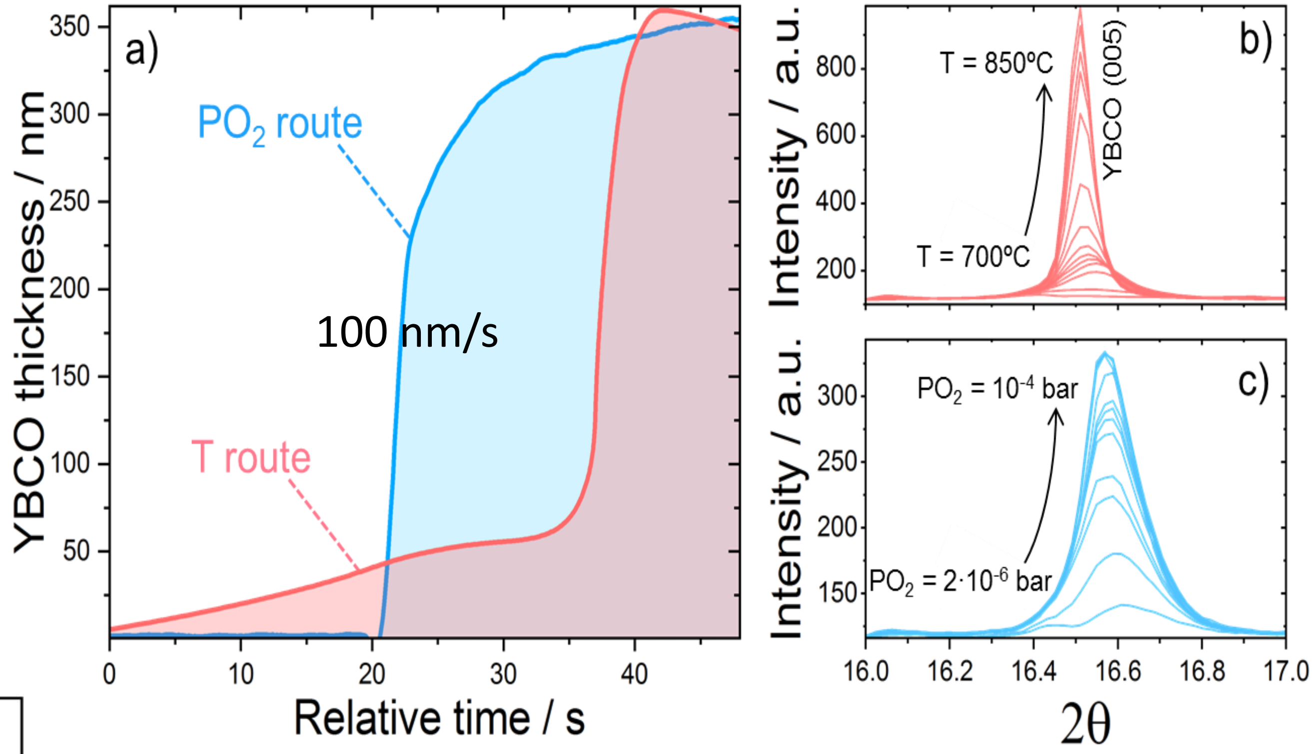
The **driving force** for nucleation is **supersaturation:**

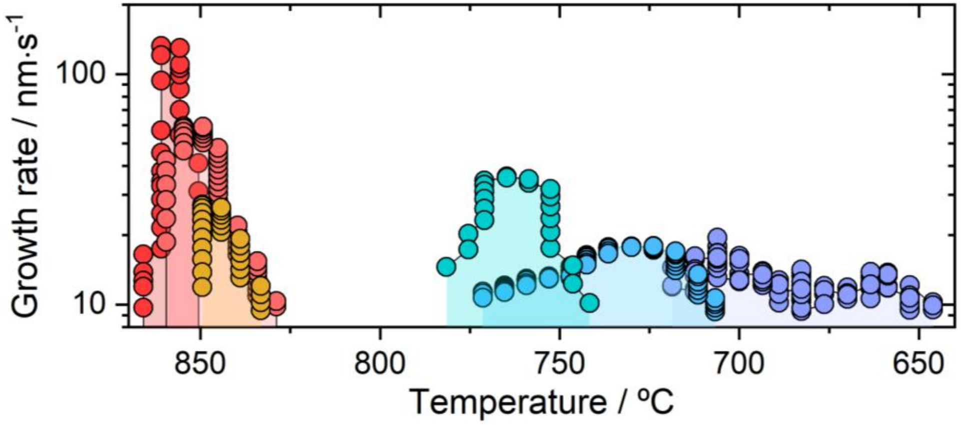
$$\Delta\mu = kTln\frac{C_\delta}{C_e} = kTln(\sigma+1)$$

Ultrafast (non-equilibrium) growth by TLAG-CSD

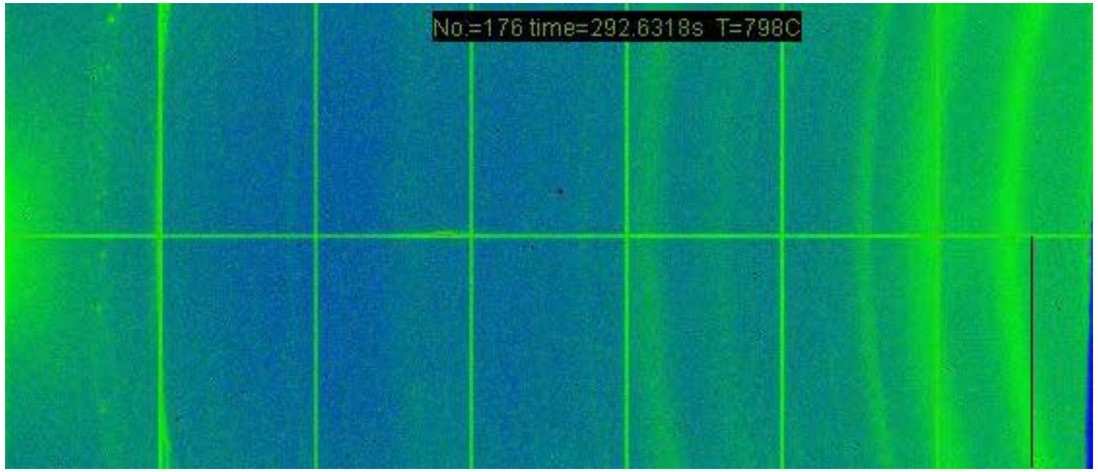
In-situ XRD synchrotron exp.

100 ms acquisition time





High heating ramps → high growth rate



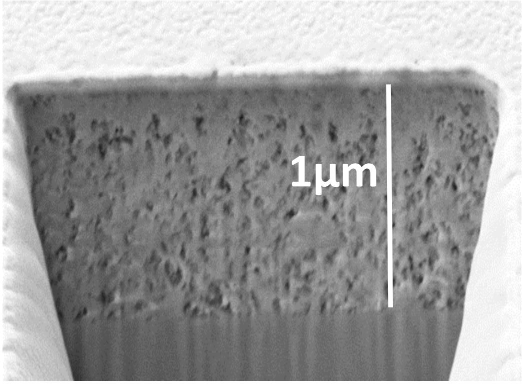
300 nm in 3s

L. Soler et al, Nature Communications 11, 344 (2020)

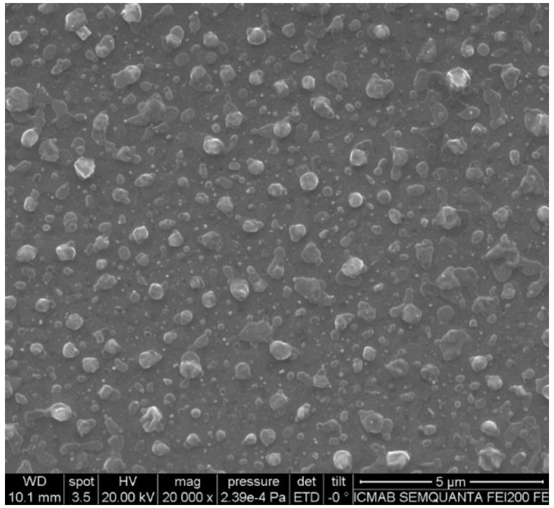
erc
European Research Council
Executive Agency

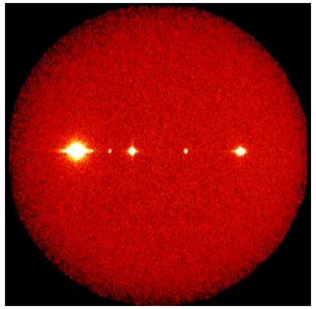
Transient Liquid Assisted Growth (TLAG-CSD) films

CSIC
ICMAB

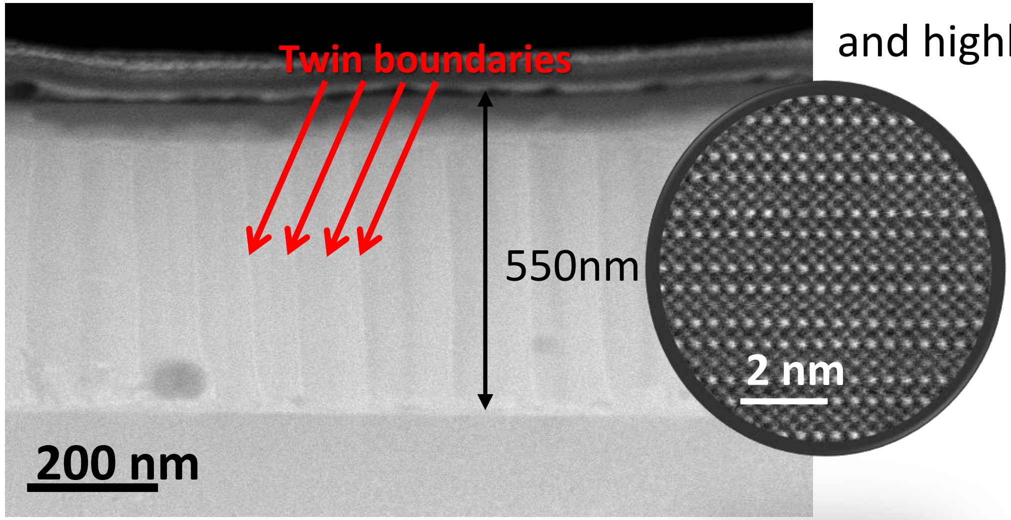


Homogeneous pyrolysis
and compatible with IJP

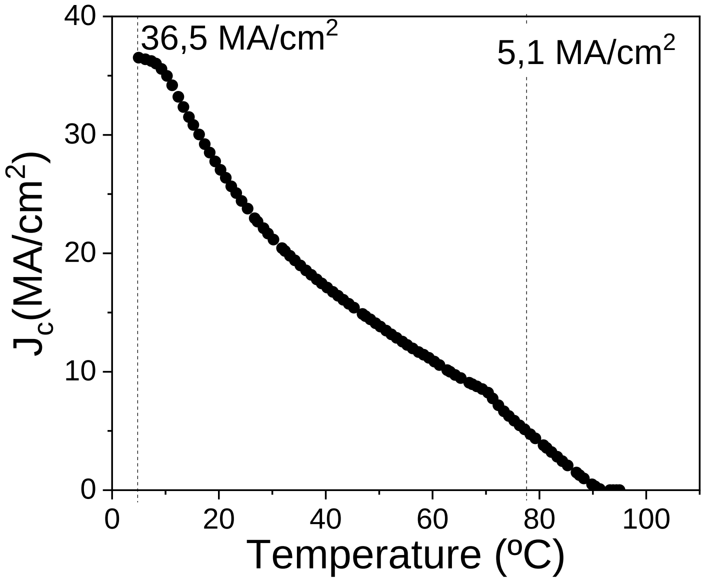




Extremely low porosity
and highly epitaxial



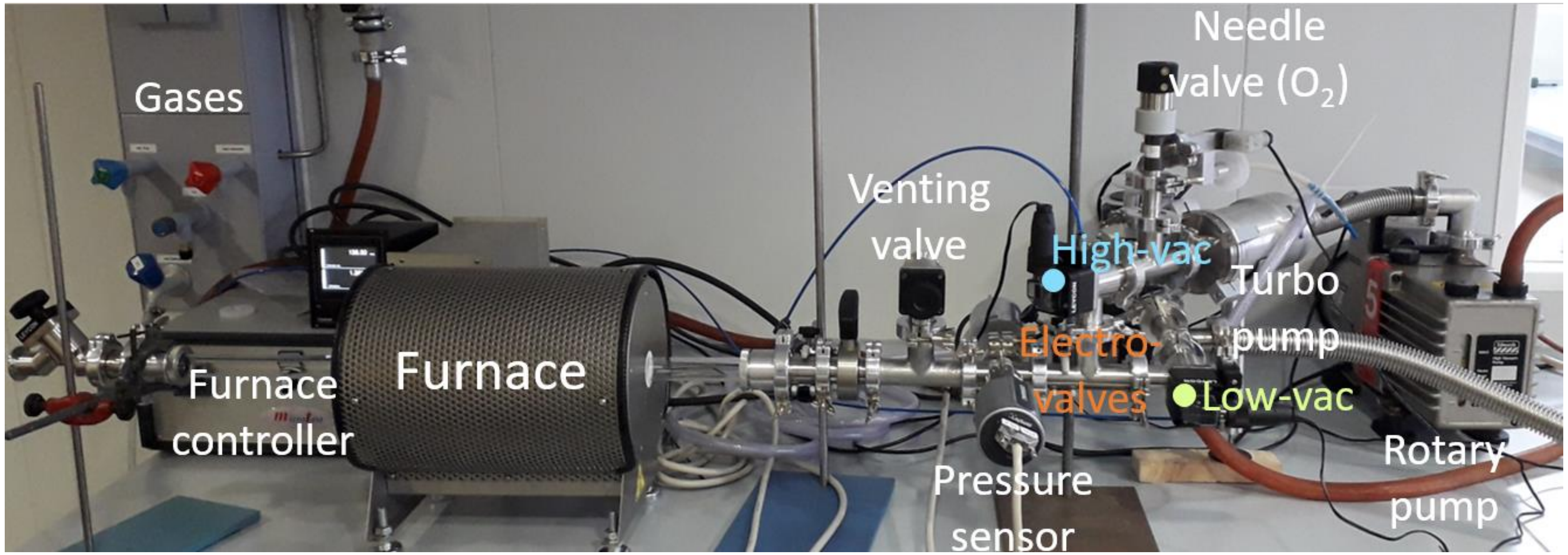
High quality growth



High performance demonstrated
J_c(77K)~ 5 MA/cm^2
T_c ~ 90 K

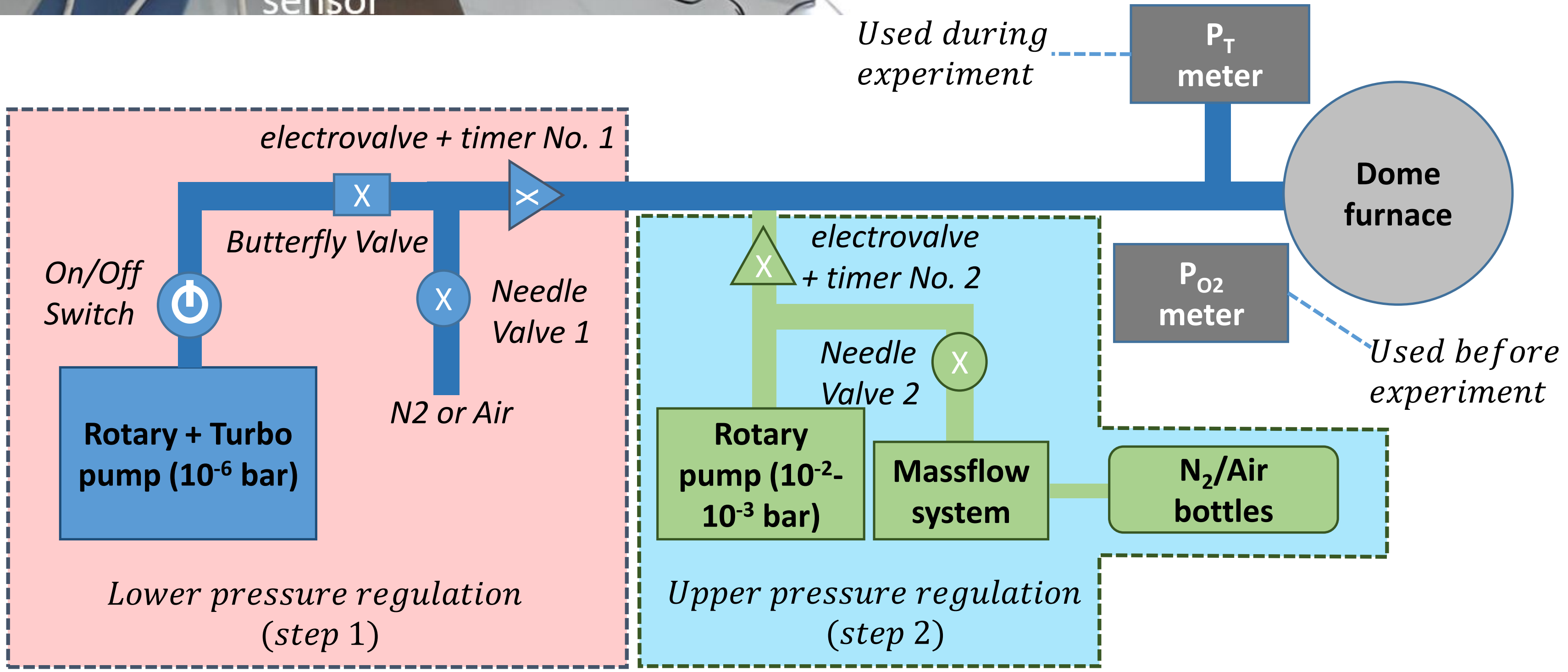
L. Soler et al, Nature Communications 11, 344 (2020)

TECHNICAL set up at ICMAB



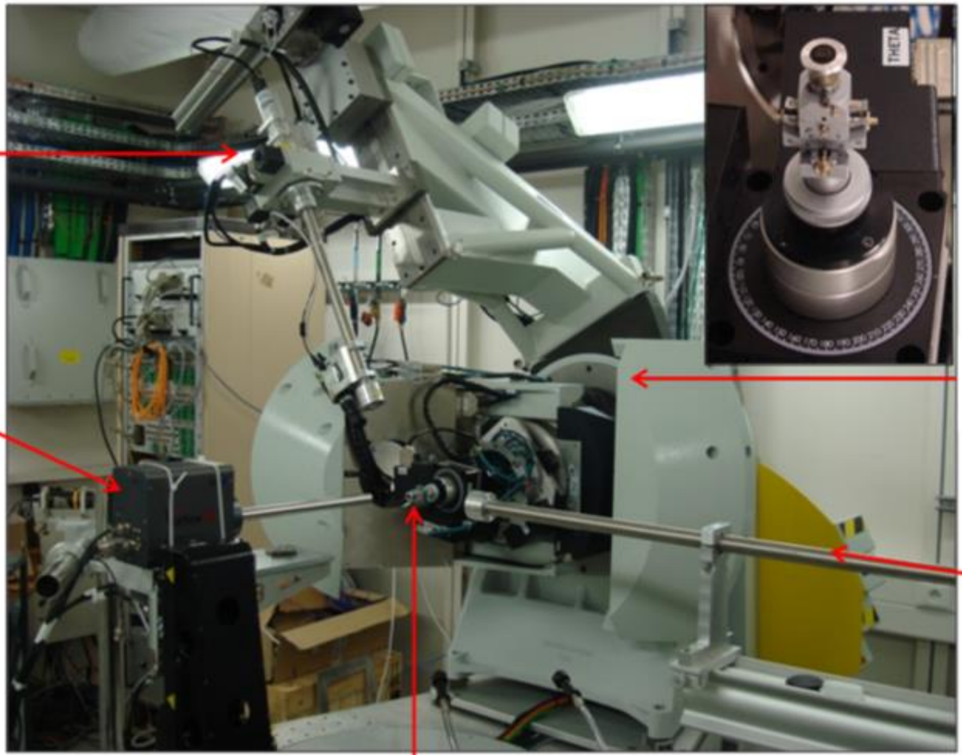
All ancillary equipment was brought to SOLEI except:

- Turbomolecular + rotary pump
- Dome furnace heating stage
- Air/N_2 gas



TECHNICAL set up at SOLEIL

- **XPAD area detector**
  (pixel size = 130 um, 560 x 240 pixels)
- Can be mounted with the long direction horiz. or vertically



"GIXRD & Brag geometry" like

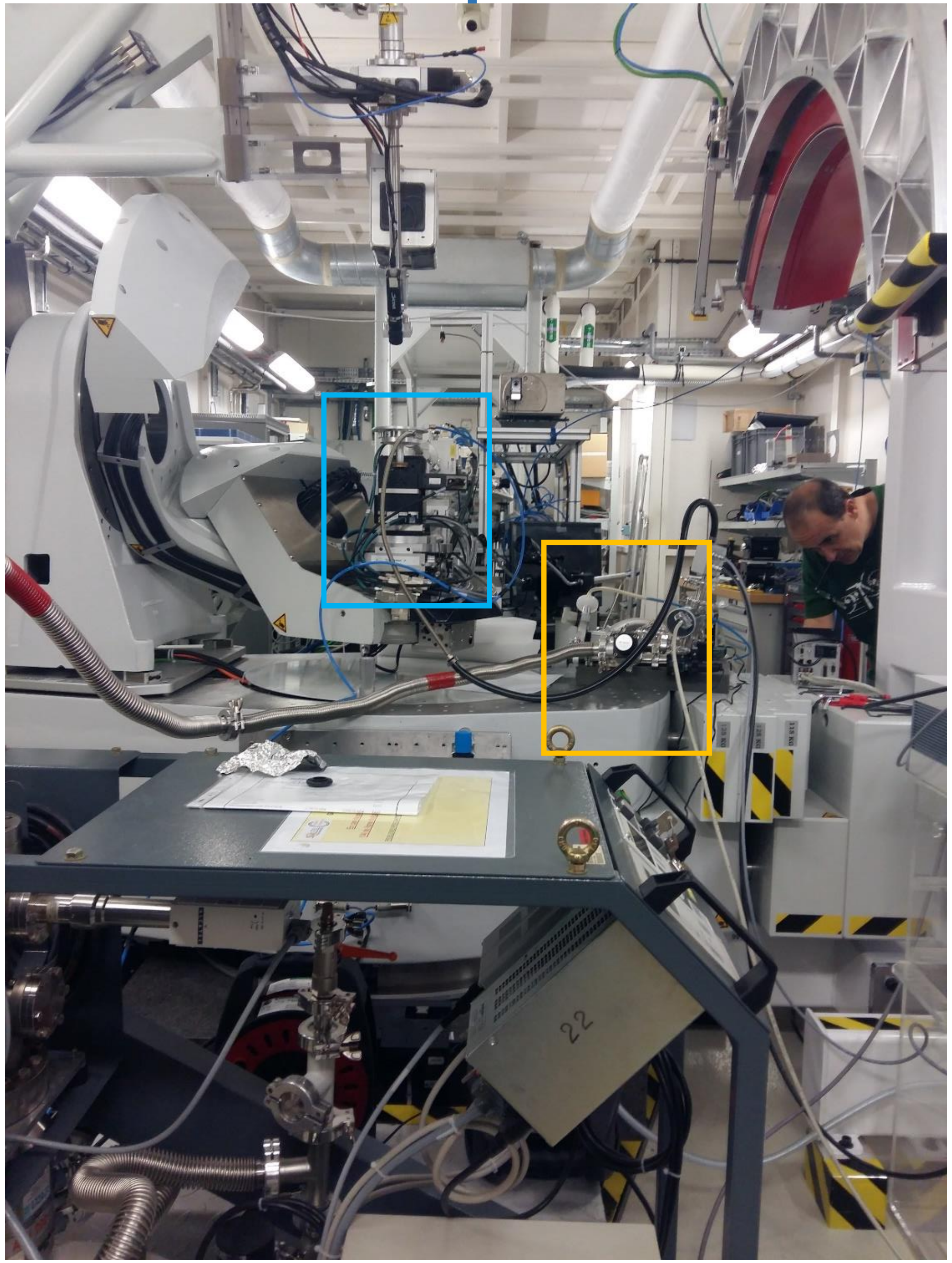
Incoming beam/optics/diffractometer:

- 6+2 circles diffractometer (kappa geometry + analyzer crystal)
- Energy range: E = 3-23 keV adjustable before experiment (18keV used)
- Energy resolution: $\Delta E/E \sim 10^{-4}$
- Beam spot size (H x V) ~ 270 x 230 um (FWHM).
  Beam divergence (H x V) ~ 0.1 x 0.005 deg (width can be reduced on expense of flux, microbeam option down to 10um spotsize available)
- Photon flux: 10^{12} ph/s with double crystal monochromator

Sample ⟺ Detector distance

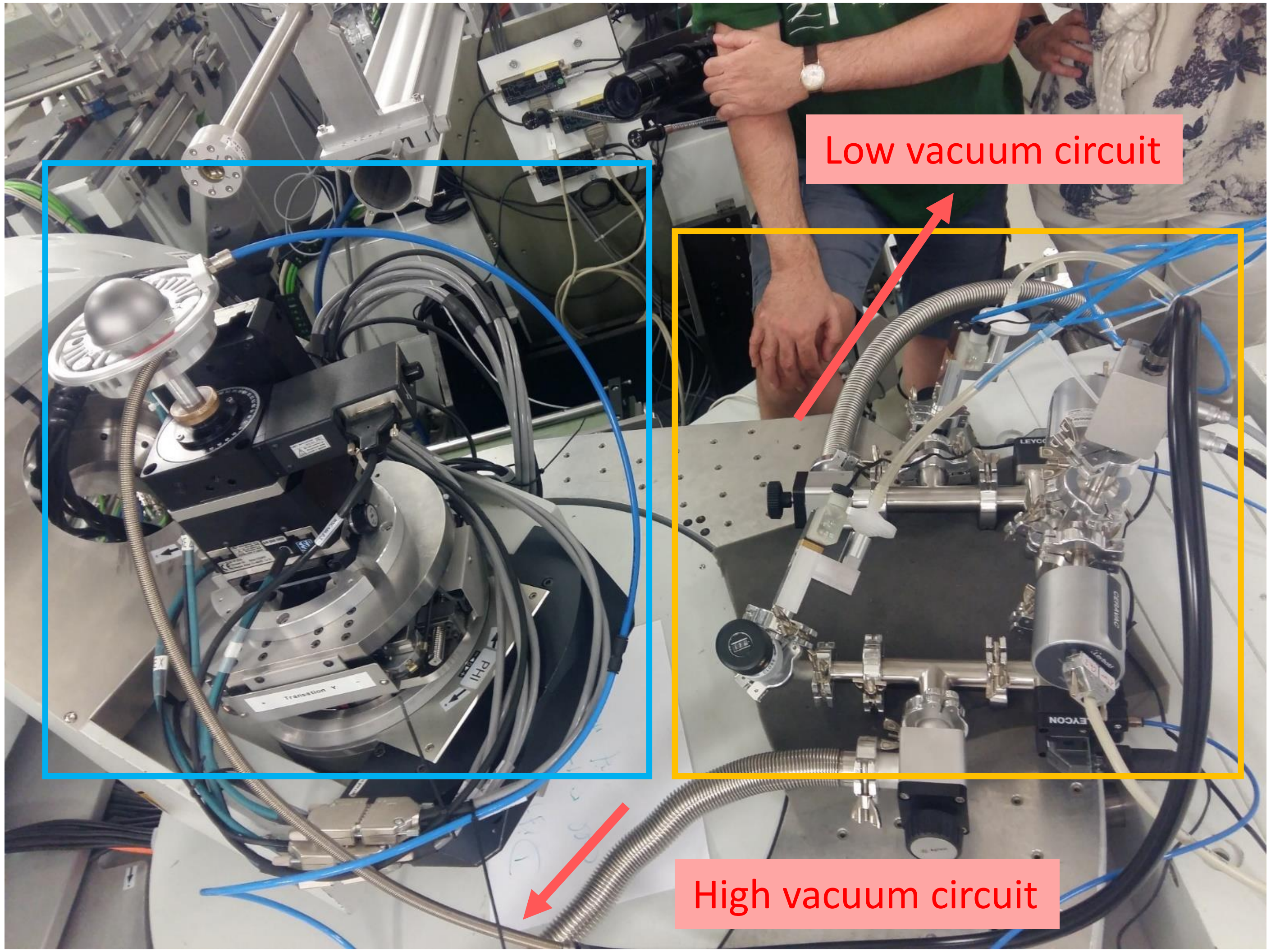
- 400 - 700 mm if point detector remains mounted (allows quick switching between both, but not needed for in-situ experiments)
- 150 - 700 mm without point detector (400 mm used to cover 20-40 deg, converted to CuK_α energy)

TECHNICAL set up at SOLEIL



Gas supply / cooling gas/
Sample stage

Butterfly/needle valves,
massflow/shutter



Gas bottles + 1 massflow outside the hutch

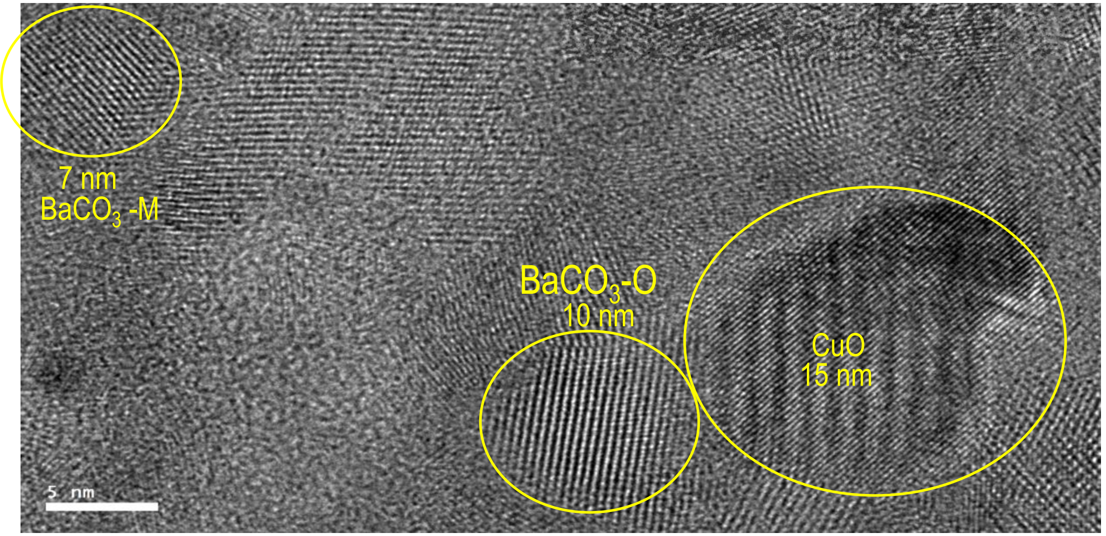
Both pumps inside the hutch

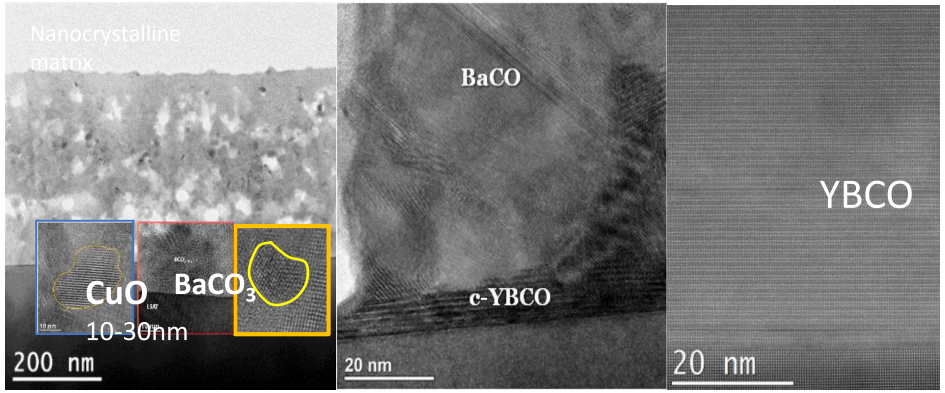
$BaCO_3$ Orthorhombic and Monoclinic decomposition

Y, Ba, Cu propionate salts → CuO + Y_2O_3 + $BaCO_3$ + volatiles

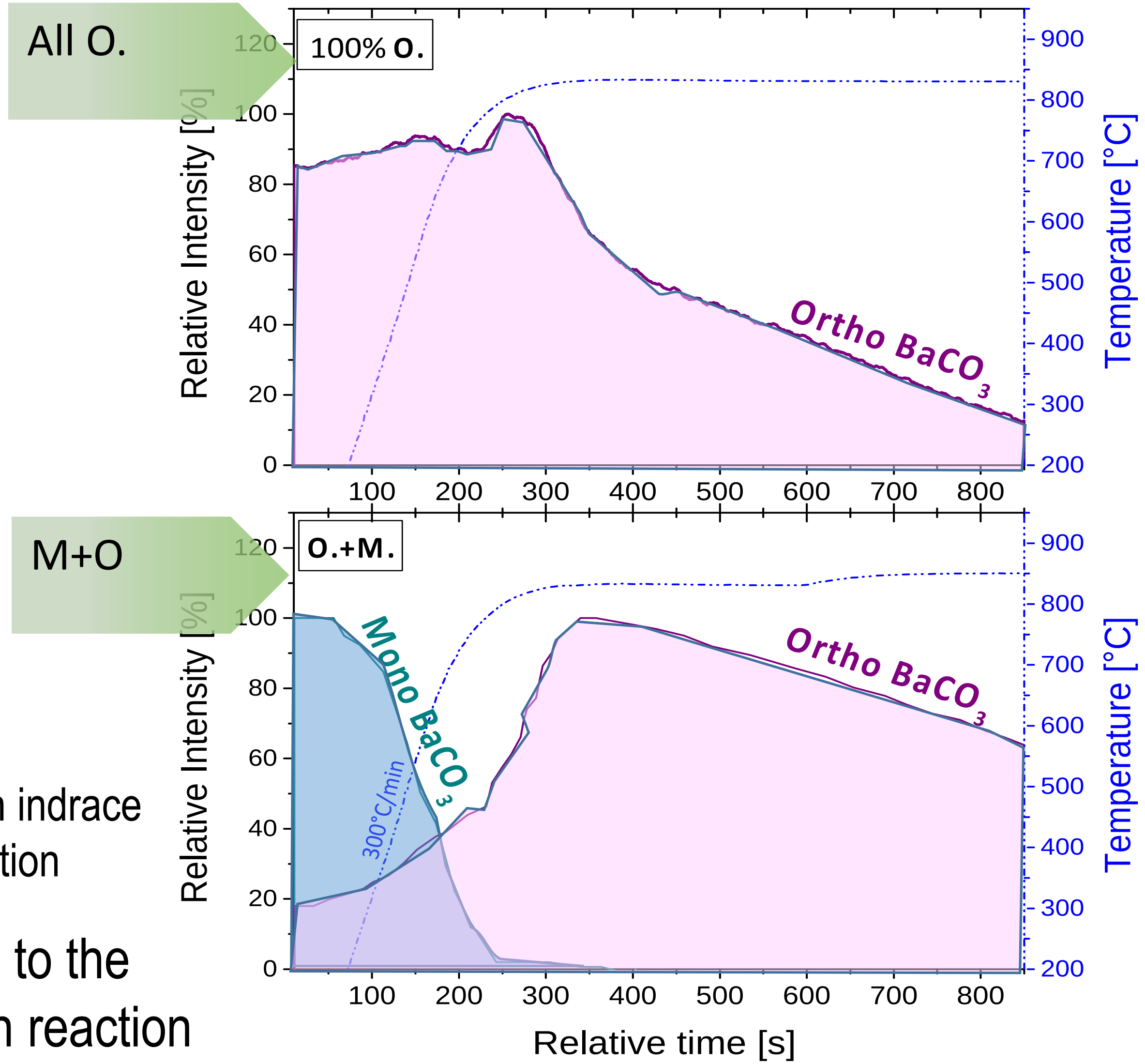
(amorph. and nanocrys. phases)

In-situ XRD synchrotron exp.





BCO is the main indrace for YBCO formation

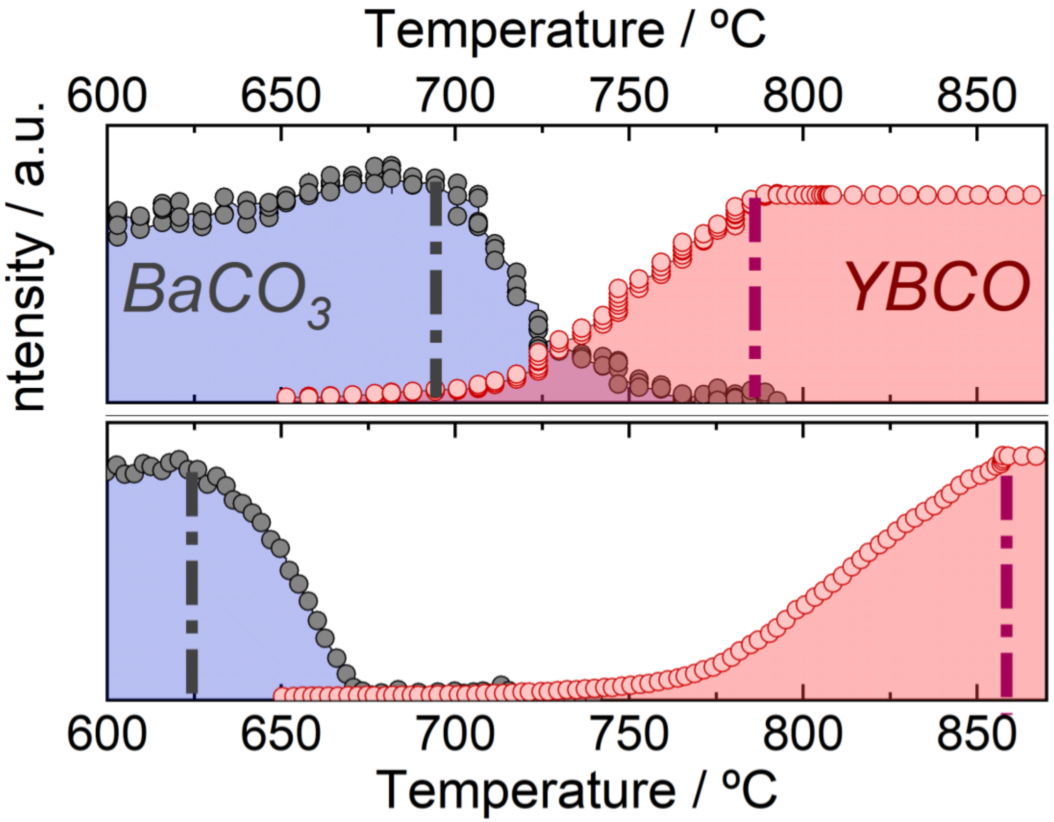


The monoclinic $BaCO_3$ phase transforms to the orthorombic phase prior to decomposition reaction

TLAG-CSD: $BaCO_3$–O decomposition is not an issue

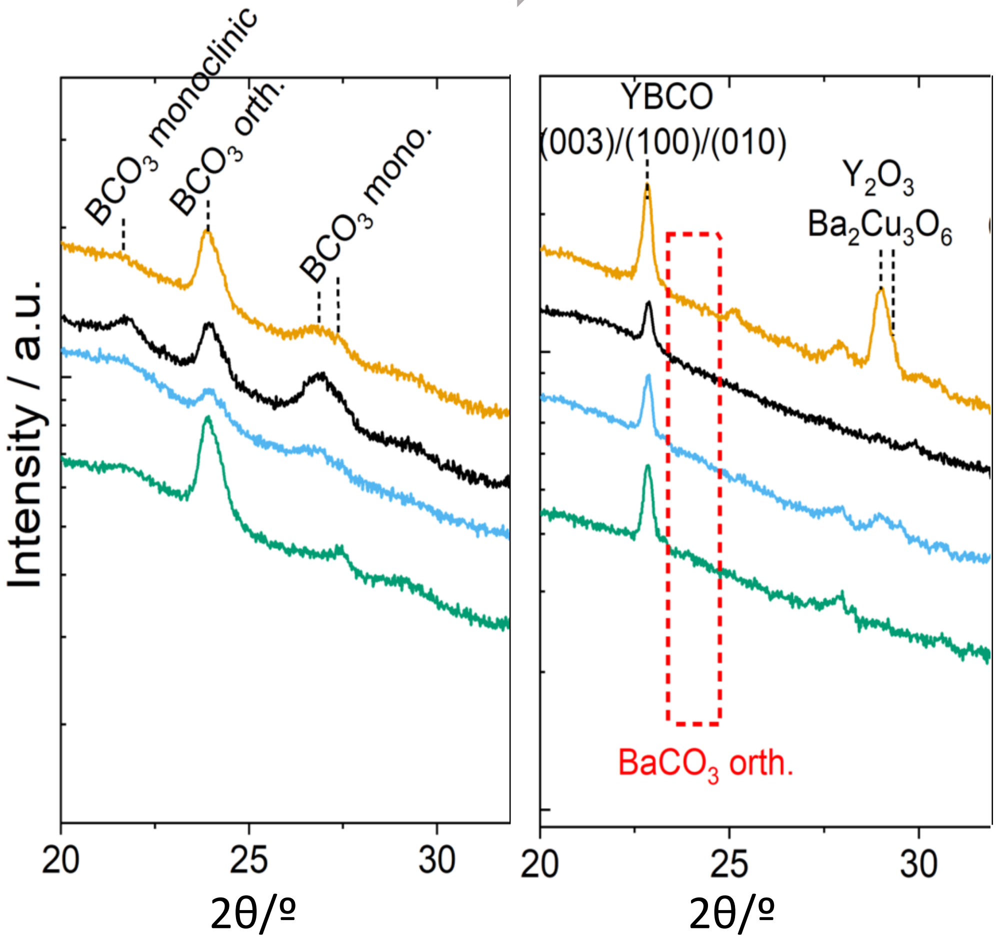
(T, P_{O2}, P_{total}, heating ramp, composition…)

In-situ XRD at Soleil sync.



Decomposition in seconds at high heating rates

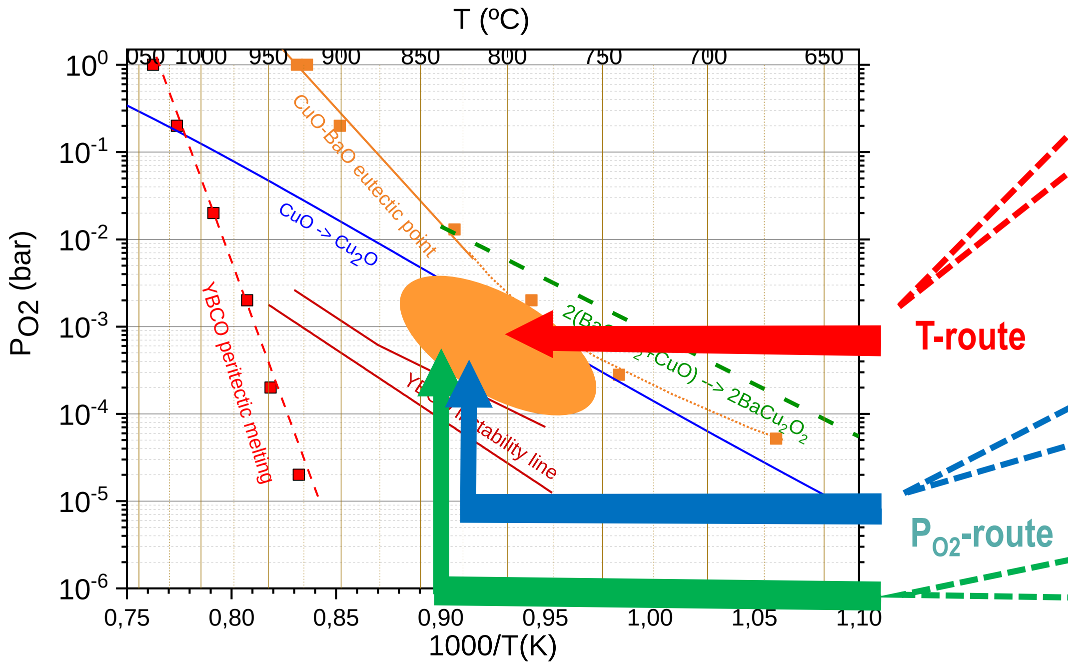
After pyrolysis → *After growth*

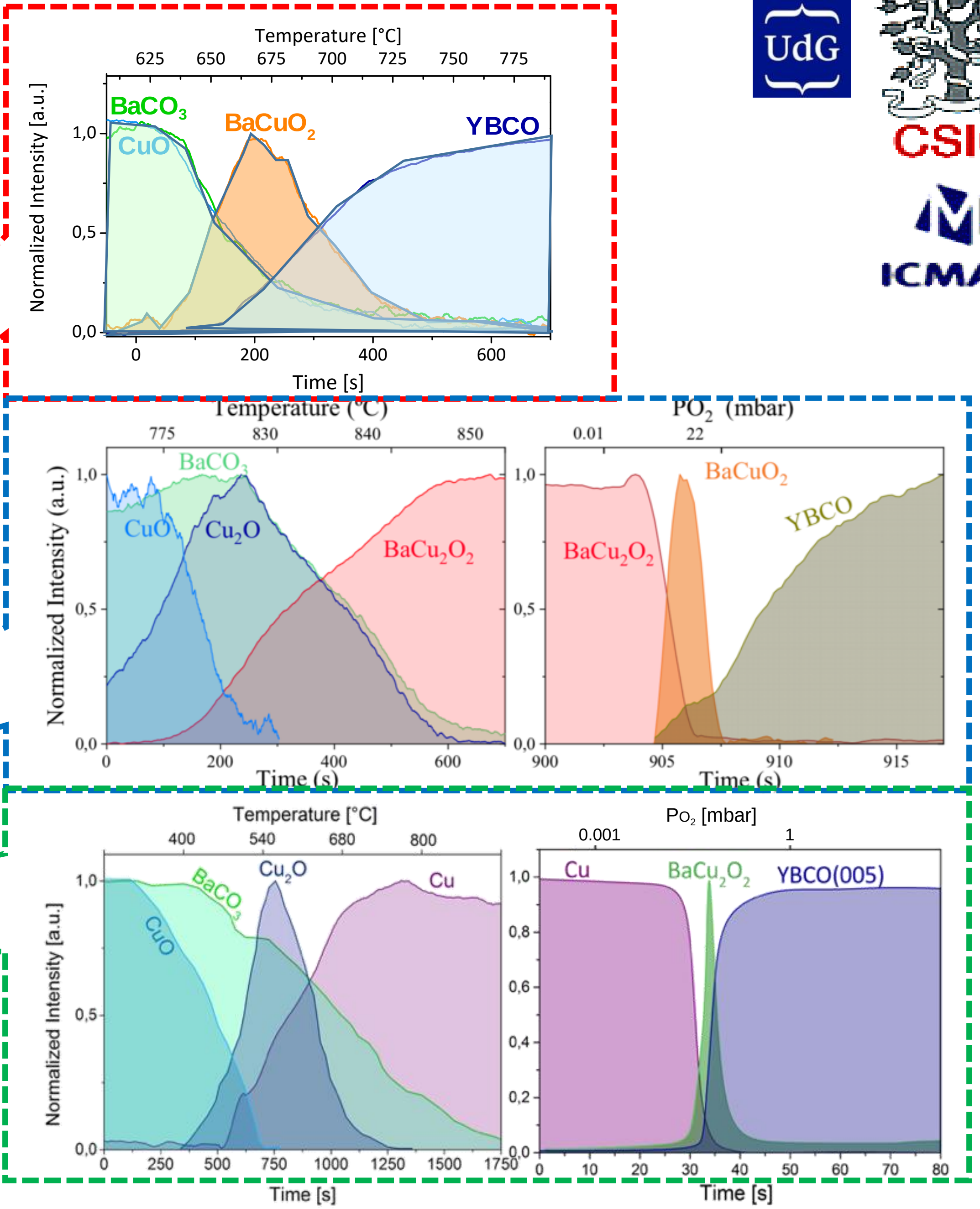


1 μm thick grown layers from diff. routes and conditions

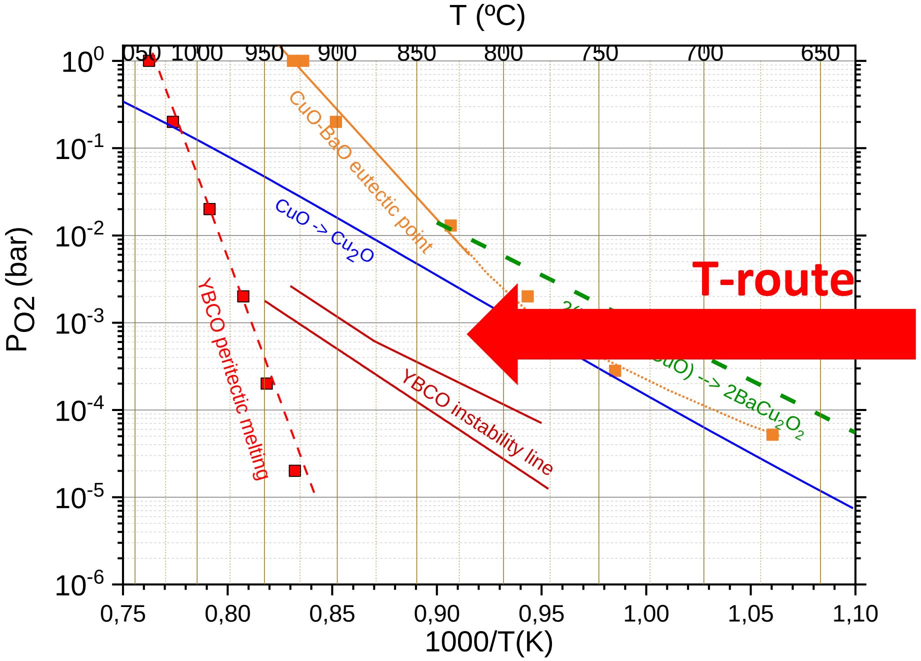
Different Routes for TLAG-CSD

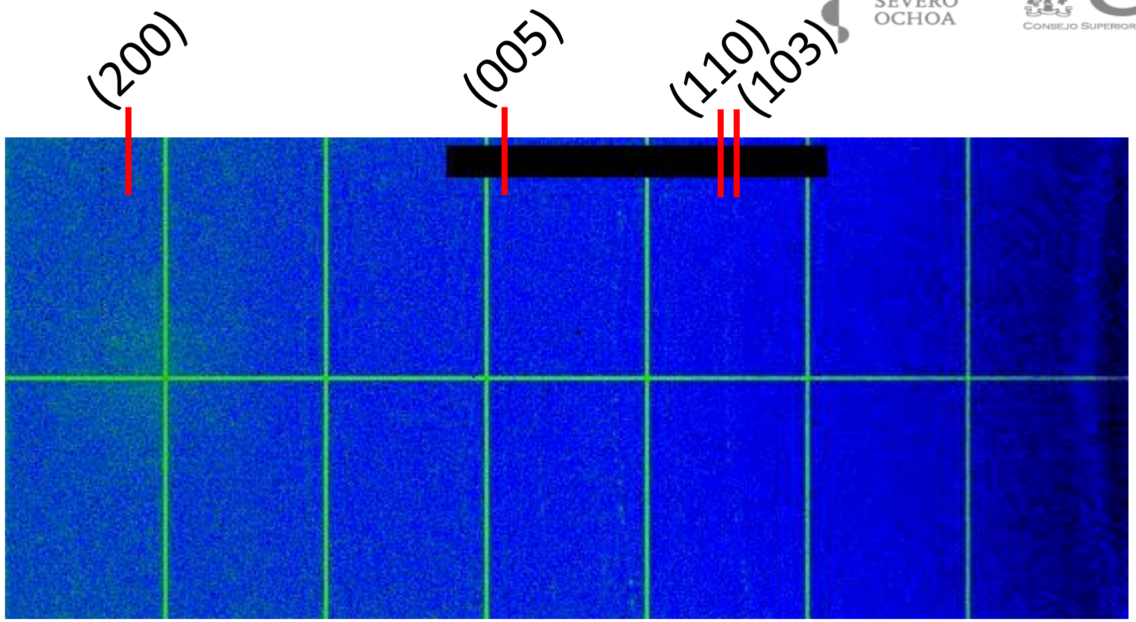
$BaCO_3(s)+CuO(s)+Y_2O_3(s) \longrightarrow [Ba\text{-}Cu\text{-}O](l) + Y_2O_3(s)$
$\longrightarrow YBa_2Cu_3O_{6.5}(s)$

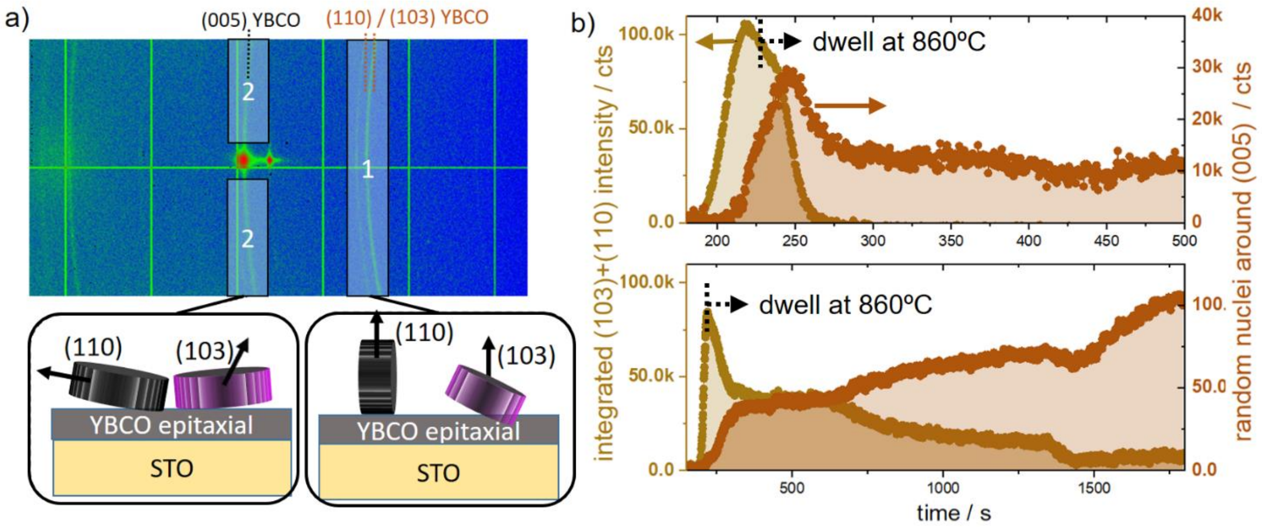




YBCO epitaxy reconstruction in TLAG-CSD







We are able to track a transformation of the random YBCO phase to the epitaxial orientation

Suggests the presence of a second transient liquid

YBCO Nanocomposites

In-situ evaluation of nanocomposites growth

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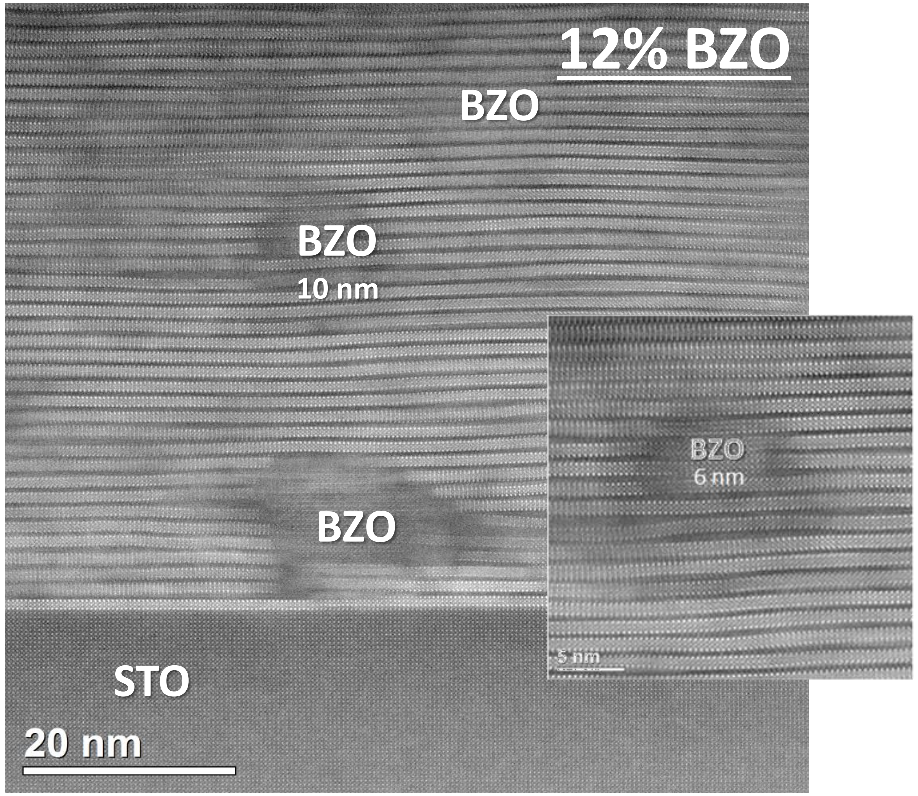
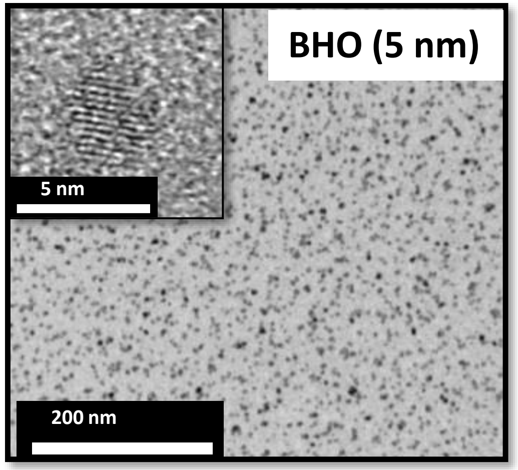
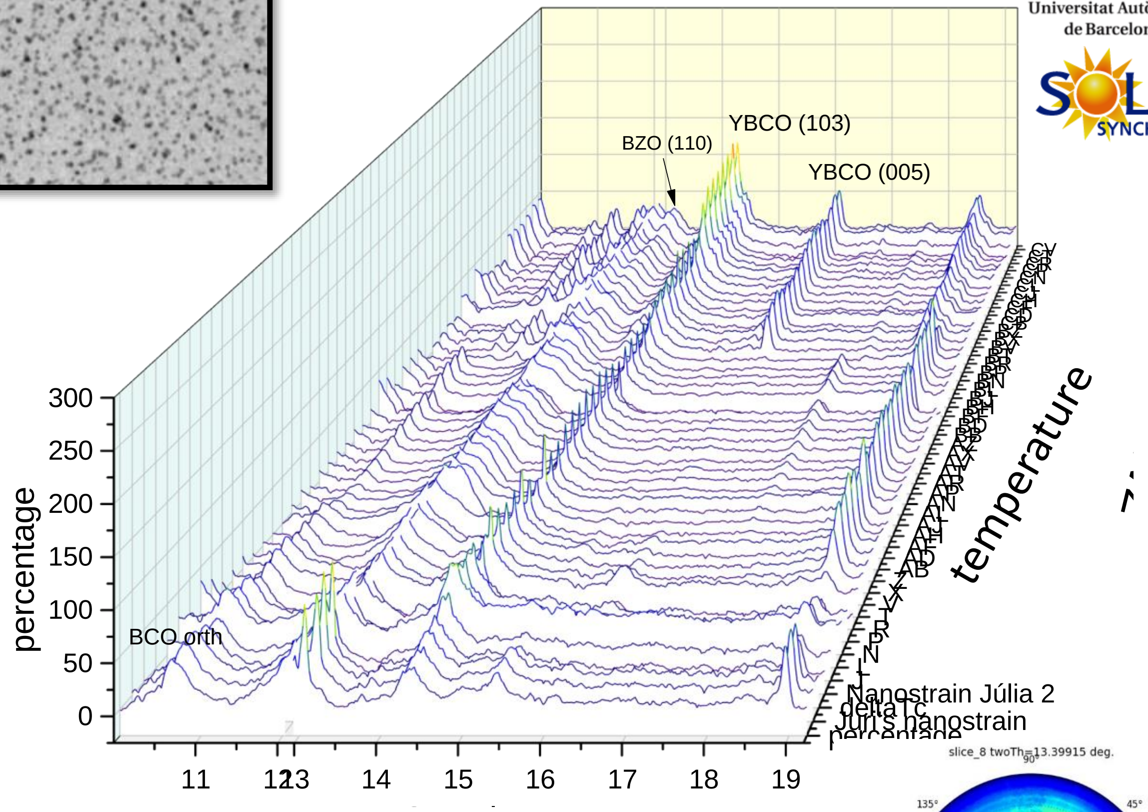
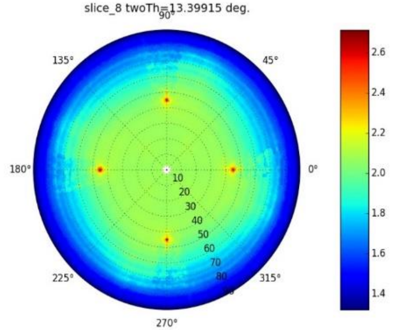
EXCELENCIA SEVERO OCHOA
CSIC
UAB Universitat Autònoma de Barcelona
SOLEIL SYNCHROTRON





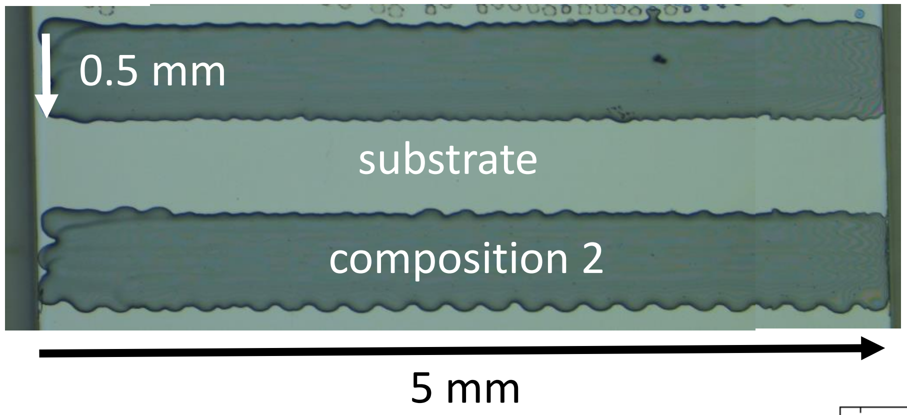




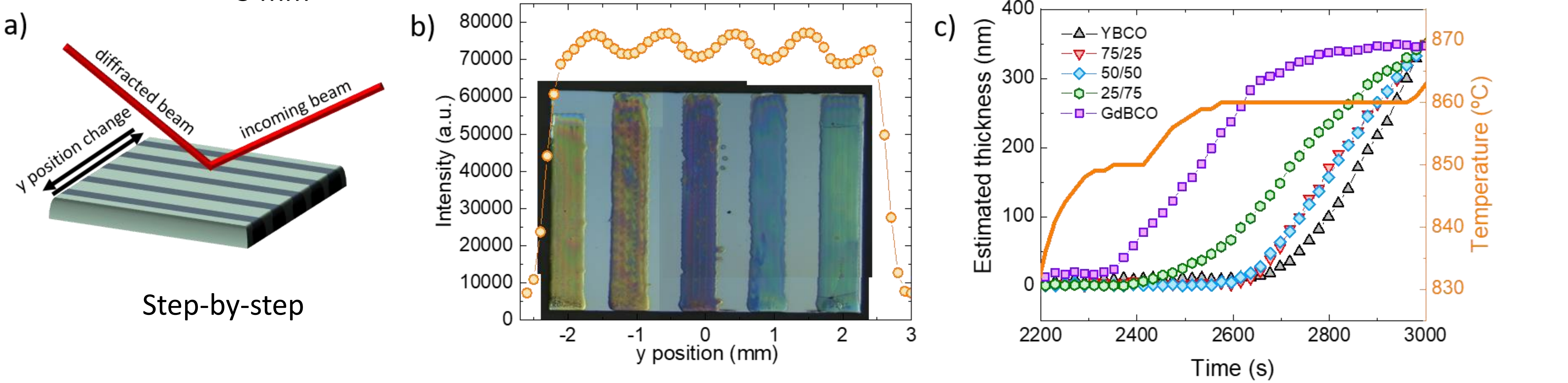
BZO (BHO) nanoparticles grow epitaxial with the YBCO matrix contrary to the other CSD processes

E. Solano et al, Thin solid films 638, 105-113 (2017)
L. Soler et al, Nat Comm. 11, 344 (2020)

Combinatorial Chemistry by Inkjet Printing



Different RE modify the liquid solubility and therefore transient liquid properties



Beam is aligned to determine the stripes' position

In-situ measurements where the nucleation time for the different RE compositions are clearly distinguished

Next is to apply it to compositionanlly-graded films using on-the-fly data acquisition

Conclusions and Proposal

- TLAG-CSD is a non-equilibrium ultrafast growth process able to grow YBCO films using CSD methods where in-situ synchrotron XRD experiments are being very powerful to understand the kinetic mechanisms of the non-equilibrium growth process
- We need to undertake a systematic study of the ultrafast growth depending on liquid solubility (RE), supersaturation (BaO-CuO composition), transient liquid phase, nanocomposites can be pursued
- We propose to build a permanent off-line setup at ALBA compatible with a beamline
- Beam BL11 (NCD) was identified. First meeting was held.
- Need to implement a fast furnace to an existing vacuum chamber
- The ancillary equipment to control pressures can be provided by ICMAB
- Opens the possibility to design an efficient high-throughput platform for advanced materials processing based on combinatorial chemistry approaches