Epitaxial Coatings for Coated Conductors

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FY 2006 Funding: 800 K

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Relevance to DOE Superconductivity Program and Technical Targets

"R&D Needs/Knowledge gaps" identified in the DOE Coated Conductor Technology Development Roadmap *and* addressed in this project:

- **Enhancing flux-pinning in superconductors**
- **Growth of epitaxial layers**
- **Understanding of microstructural factors affecting critical current density**
- **Developing better and improved buffer layers**

Specific Objectives/Milestones

- Correlate HTS properties with the level of $BaZrO_3$ incorporated in films as 3D, self-assembled, columnar nanodot arrays.
- Fabricate films of REBCO with and without nanoparticle additions and compare pinning characteristics with YBCO with nanoparticle additions.
- Continue optimizing nanoparticle substrate surface modifications for flux-pinning in thick *in situ* and *ex situ* YBCO on RABiTS, and extend to YBCO on IBAD Templates.
- Conduct detailed measurement and analysis of the wide-range thickness dependence of critical currents in *ex situ* YBCO on RABiTS.
- Continue fundamental studies of epitaxial growth of multifunctional buffers on textured substrates.

Presentation Outline

- **FY 2006 Results**

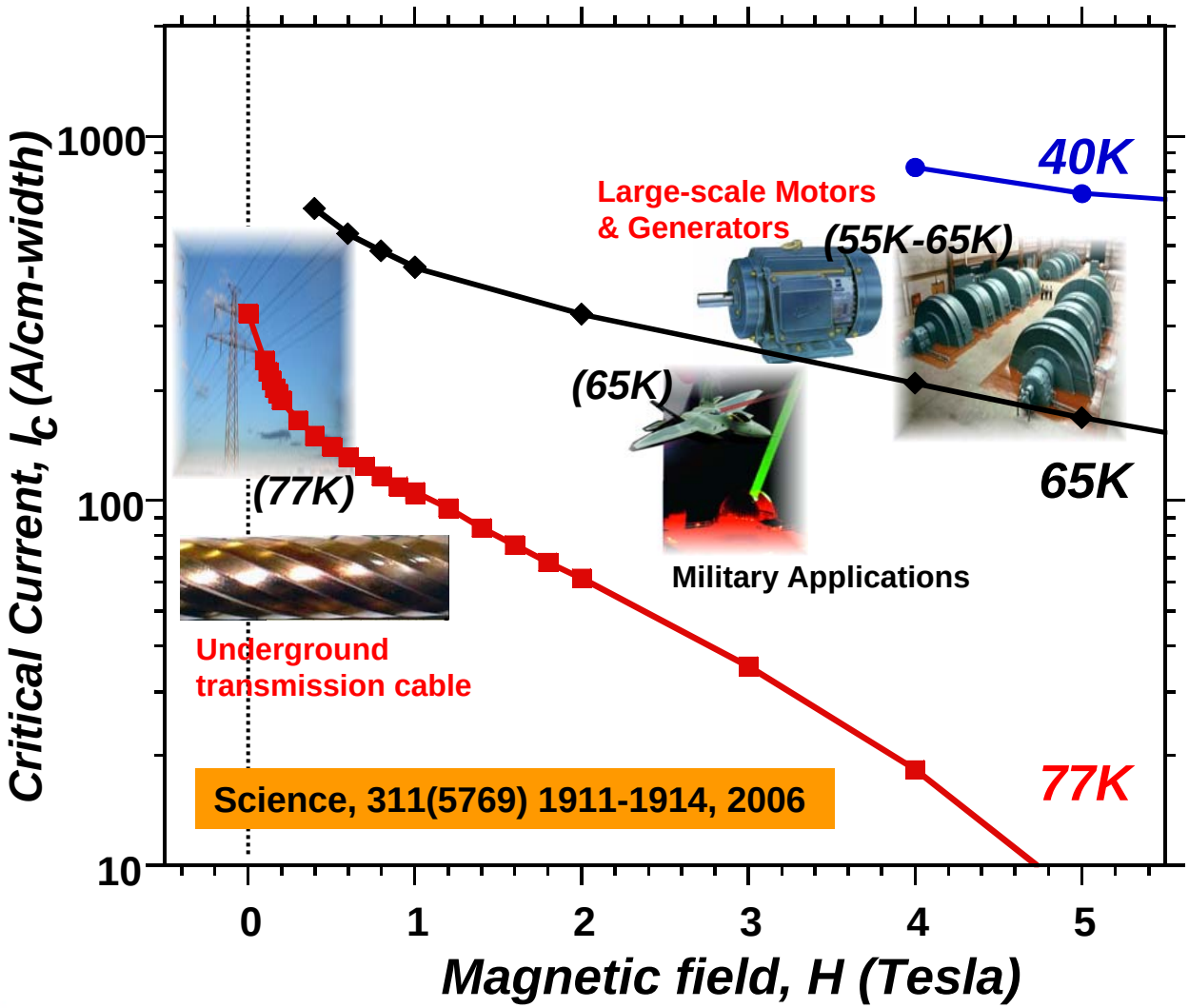
  Flux pinning in YBCO & REBCO films with columns of insulating, self-assembled nanodots (Goyal)

  Systematics of YBCO with BZO nanodots (Christen)
  J_c analysis for Y_2O_3 doped YBCO ex situ vs thickness
  Nanoparticle substrate decoration for J_c enhancements

  Epitaxial growth of buffer layers (Parans)

- **FY 2006 Performance and FY 2007 Plans (Parans)**
- **Research Integration**

FY2005: Performance requirements for many large-scale applications can be met by incorporation of self-assembled insulating nanodots



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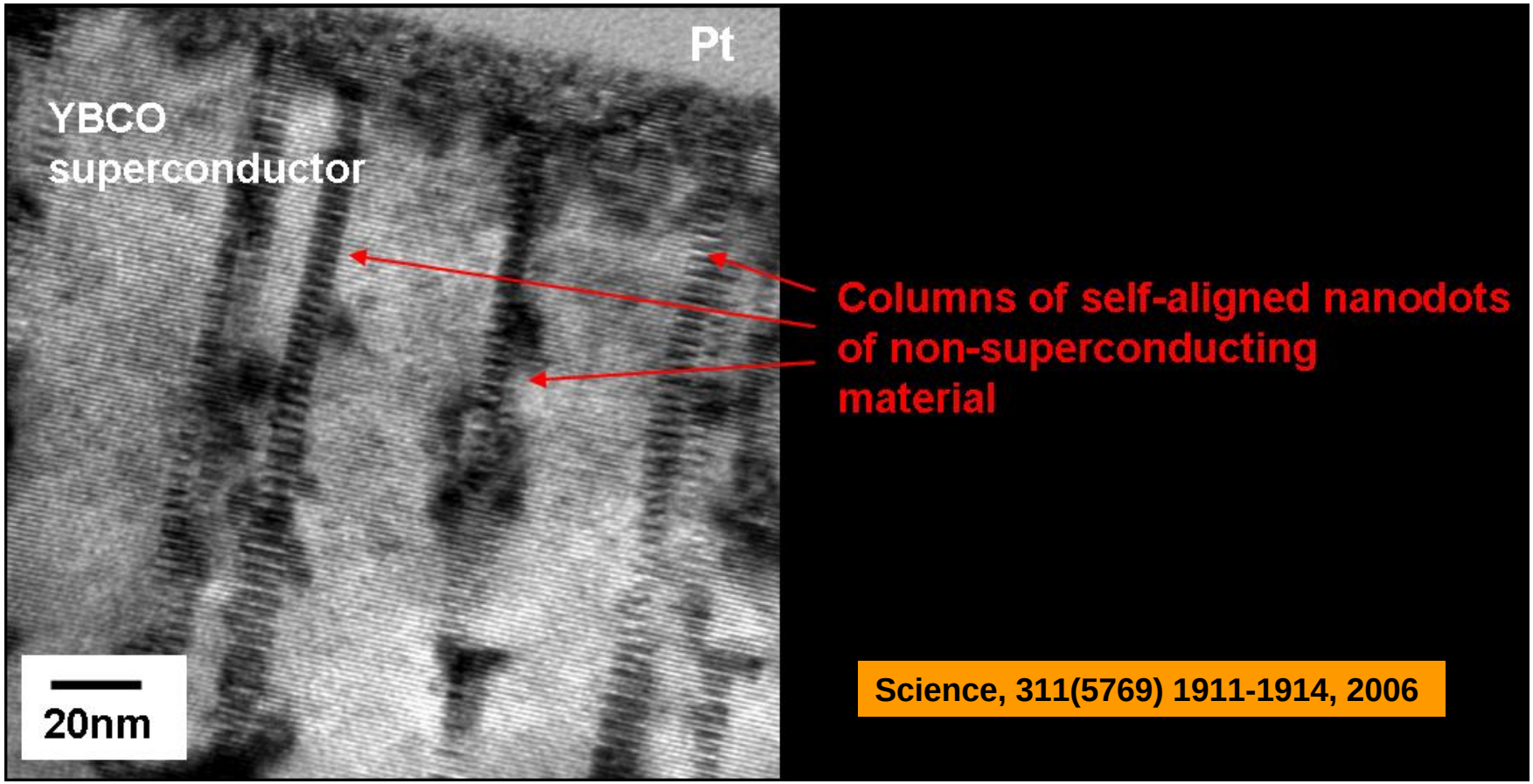
FY2006: Can we improve the performance further by changing the matrix around the columnar defects?



Science, 311(5769) 1911-1914, 2006

What about REBCO films doped columns of self-assembled BZO nandots?

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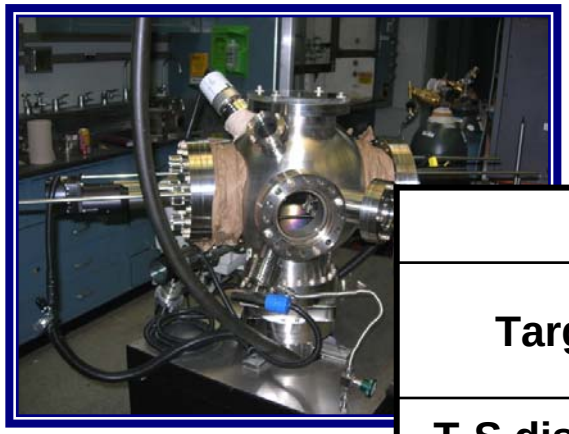
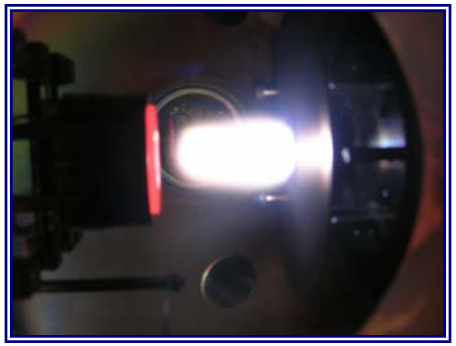
PLD used to incorporate self-aligned nanodots & nanorods

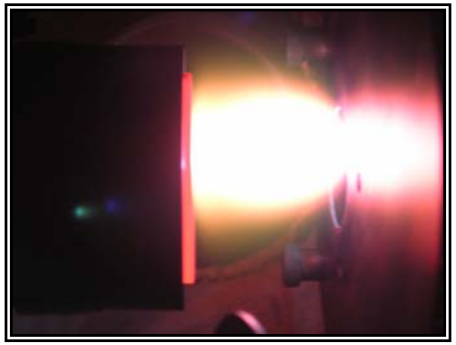


	Condition 1	Condition 2
Target	REBCO and REBCO+ 2 vol.% BZO target	
T-S distance	7 cm	4 cm
T_s	760°C	800°C
Processing gas	1%O_2/Ar	Pure O_2
Pressure	800 mTorr	200 mTorr
Laser energy	90 mJ (1.8 J/cm^2)	
Substrate	**RABiTS:** CeO_2/YSZ/Y_2O_3/NiW **IBAD:** $LaMnO_3$/IBADMgO/Hastelloy	

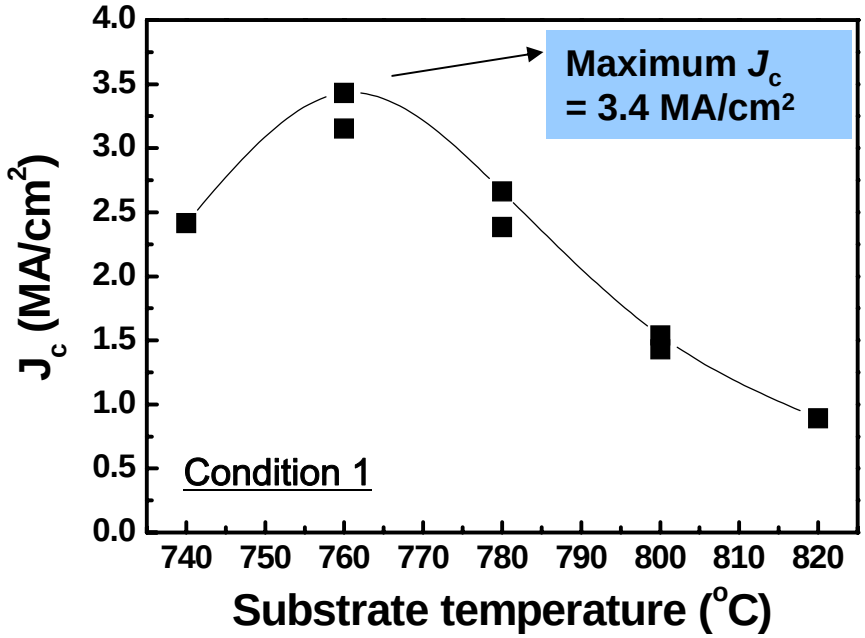
Condition 1

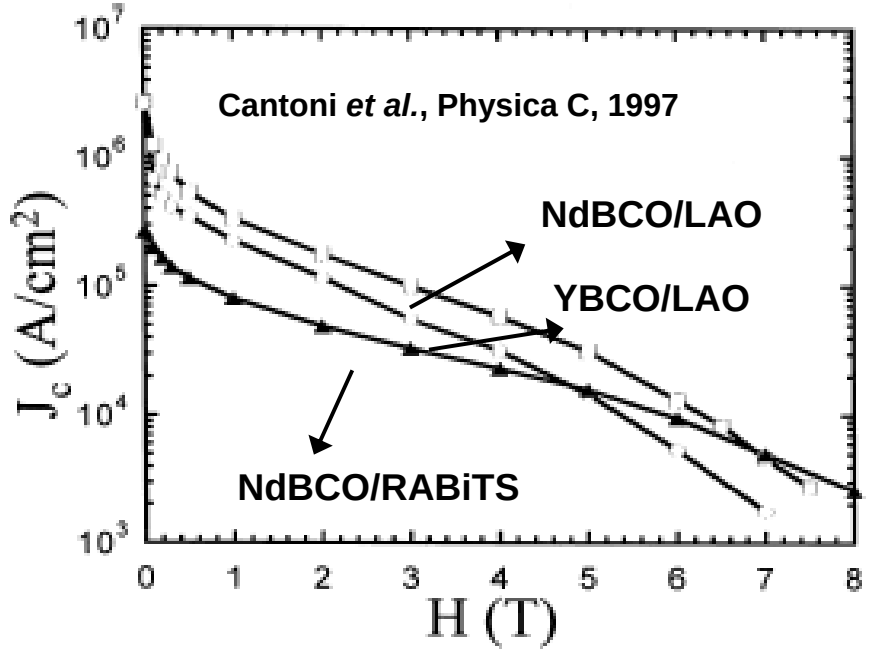


Condition 2



High-J_c observed in thin Nd-123 films for the first time

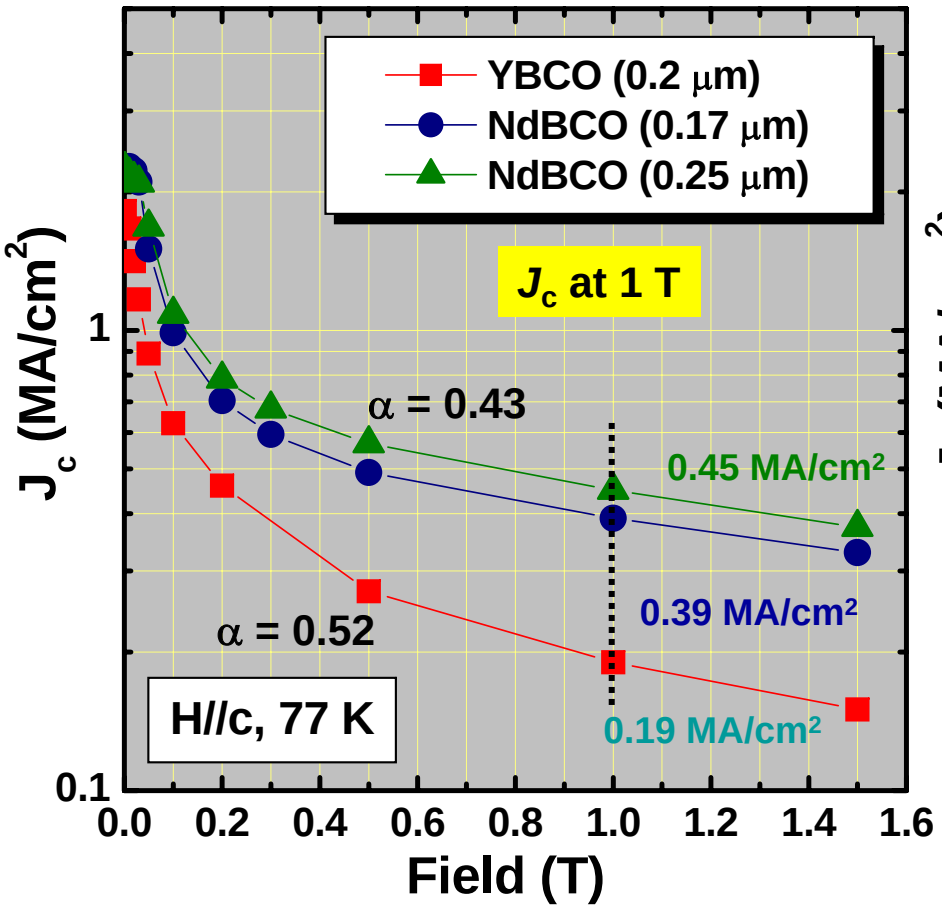


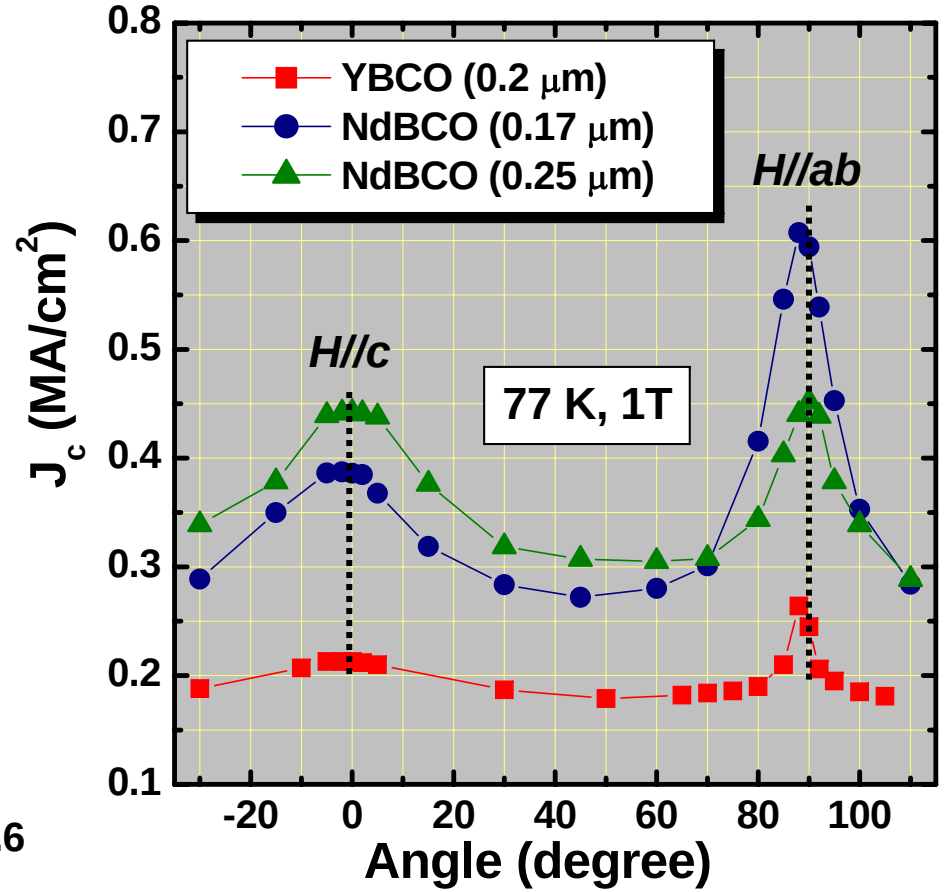


Texture of films is good

Sample I.D.	Thickness (μm)	J_c (MA/cm²)	(006) R.D.	(006) T.D.	(113) Φ-scan
WN08301B	0.13	3.4	3.35	4.71	6.79
WN09091A	0.17	2.2	3.91	4.71	6.94
WN09021A	0.25	2.2	3.68	4.87	6.94

NdBCO films are less sensitive to applied magnetic fields than YBCO films





Angular dependence is better at all angles than YBCO

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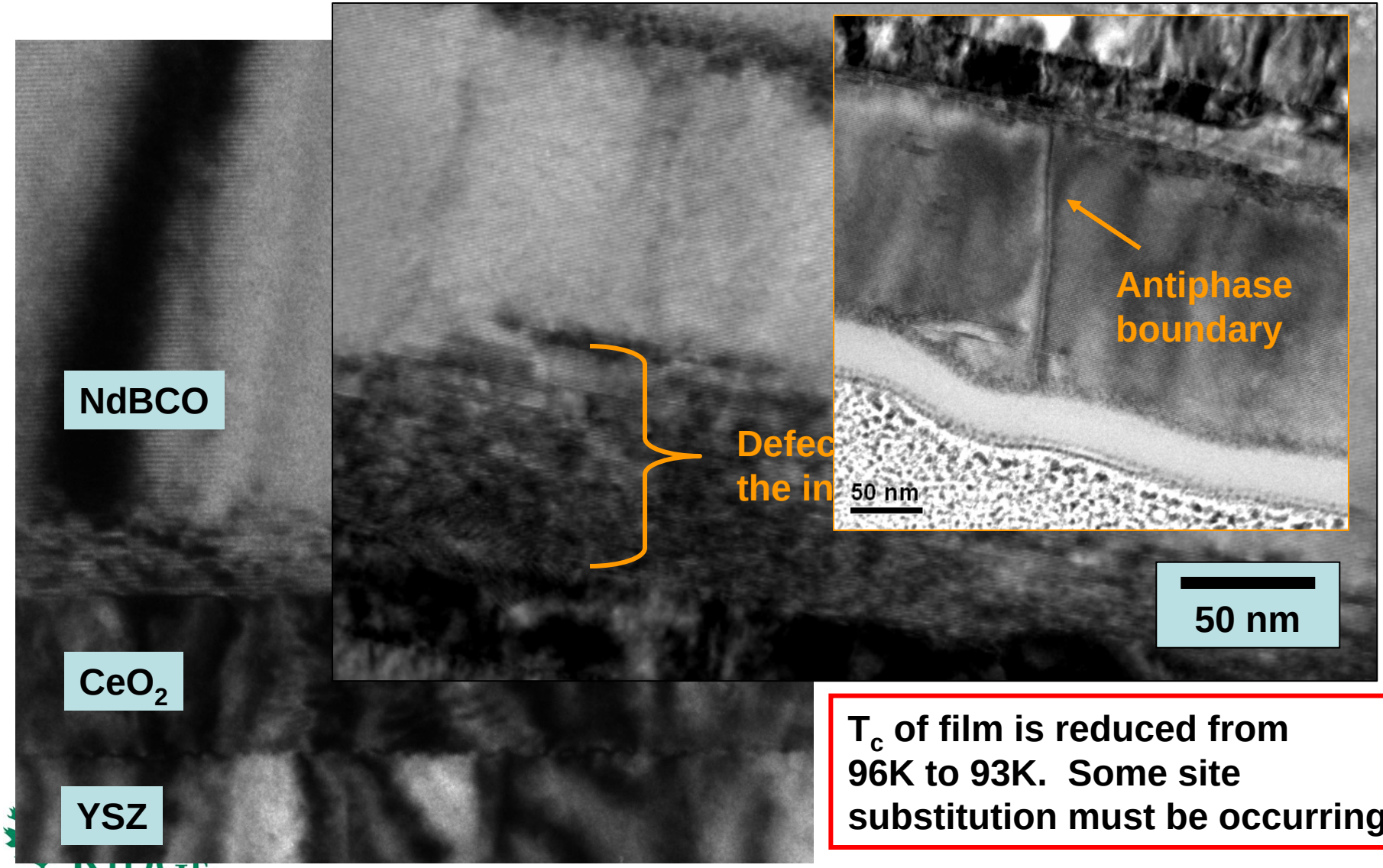
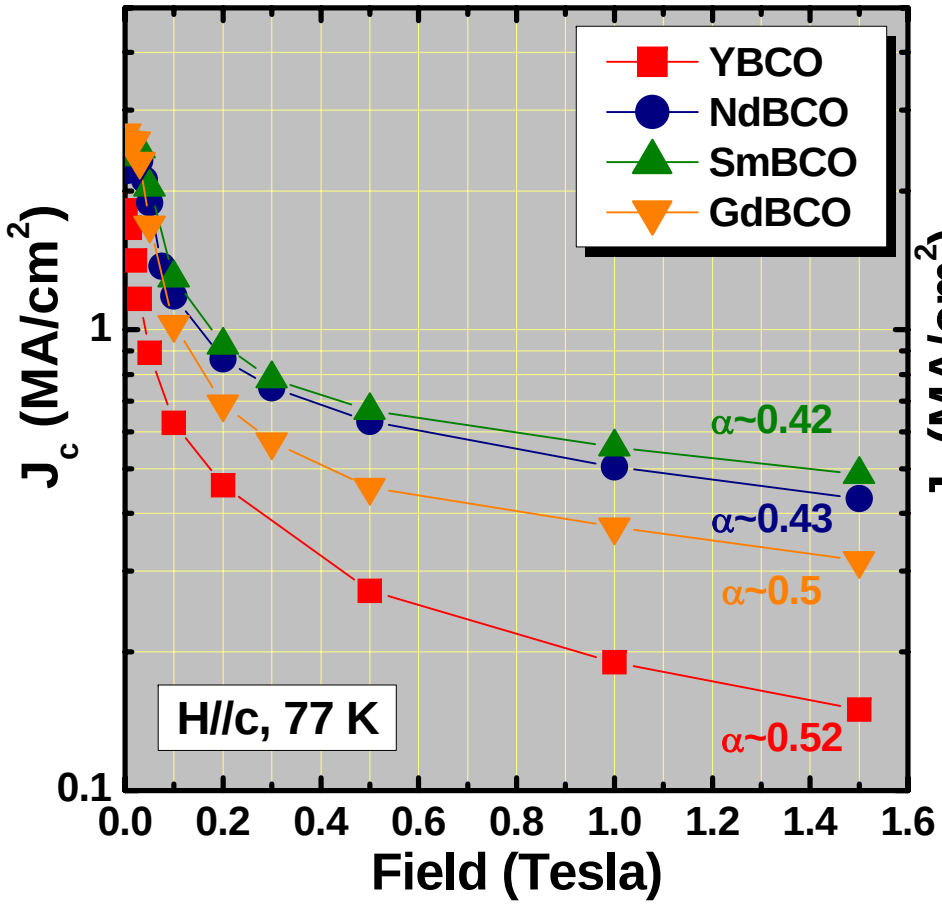
Defects in NdBCO film on RABiTS

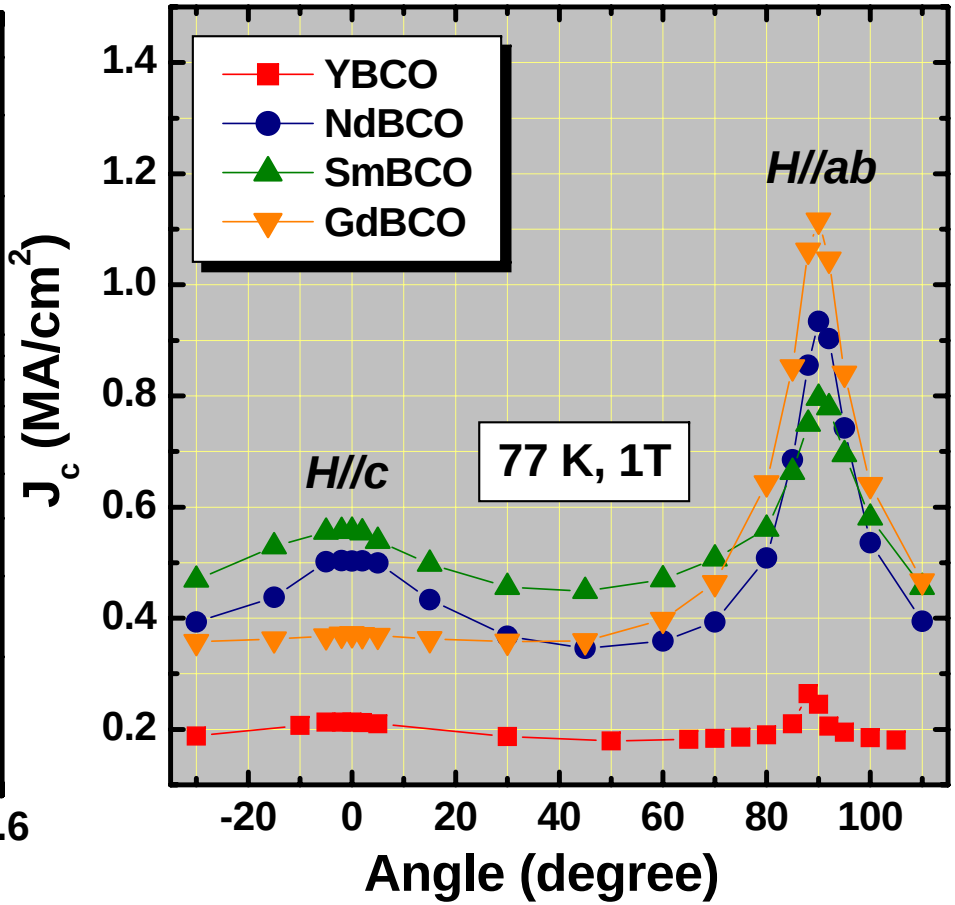


T_c of film is reduced from 96K to 93K. Some site substitution must be occurring!

0.15~0.17 μm thick SmBCO and GdBCO films

All films grown in condition 1





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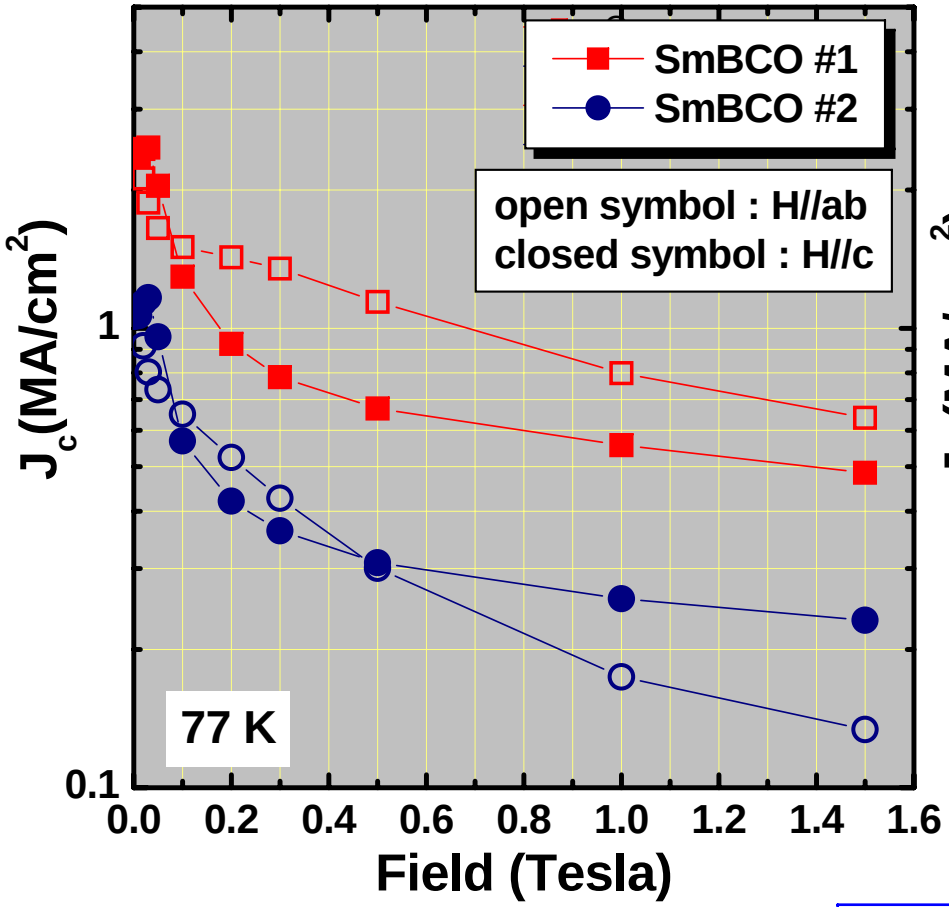
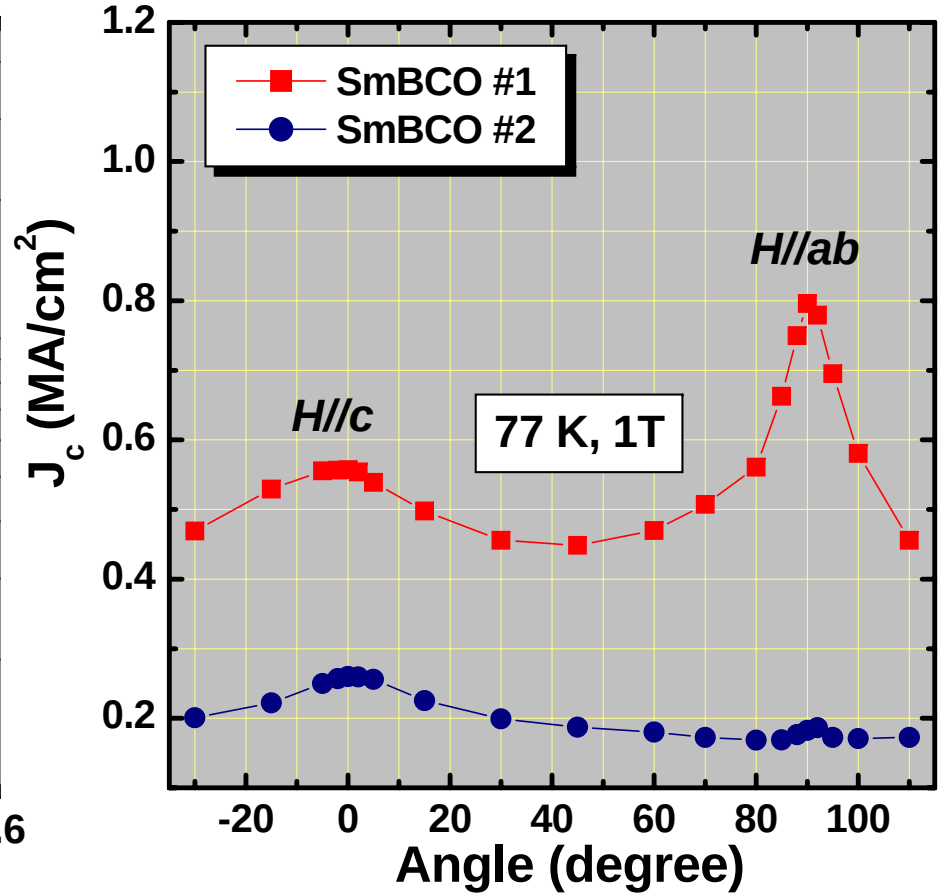
Pinning is very sensitive to growth conditions for Sm-123 films

Pinning properties controlled via PLD processing condition (condition 1 and 2)





SmBCO #1 (0.15 μm) : T_c = 92.2 K
SmBCO #2 (0.32 μm) : T_c = 93.2 K

	(006) R.D.	(006) T.D.	(113) Φ-scan
SmBCO#1	4.73	5.59	7.38
SmBCO#2	4.85	5.87	7.55

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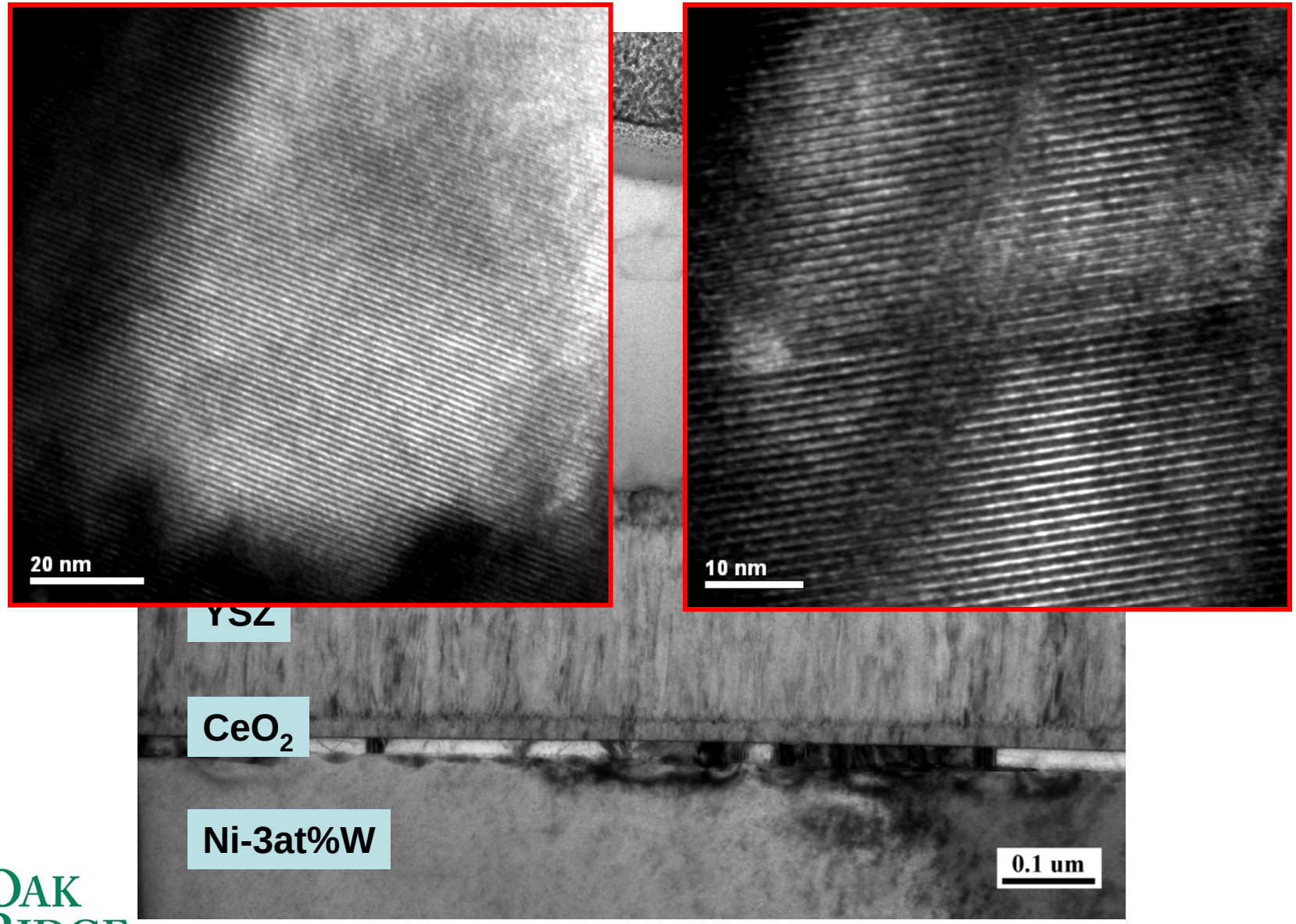
For condition 2, Sm-123 films have few defects



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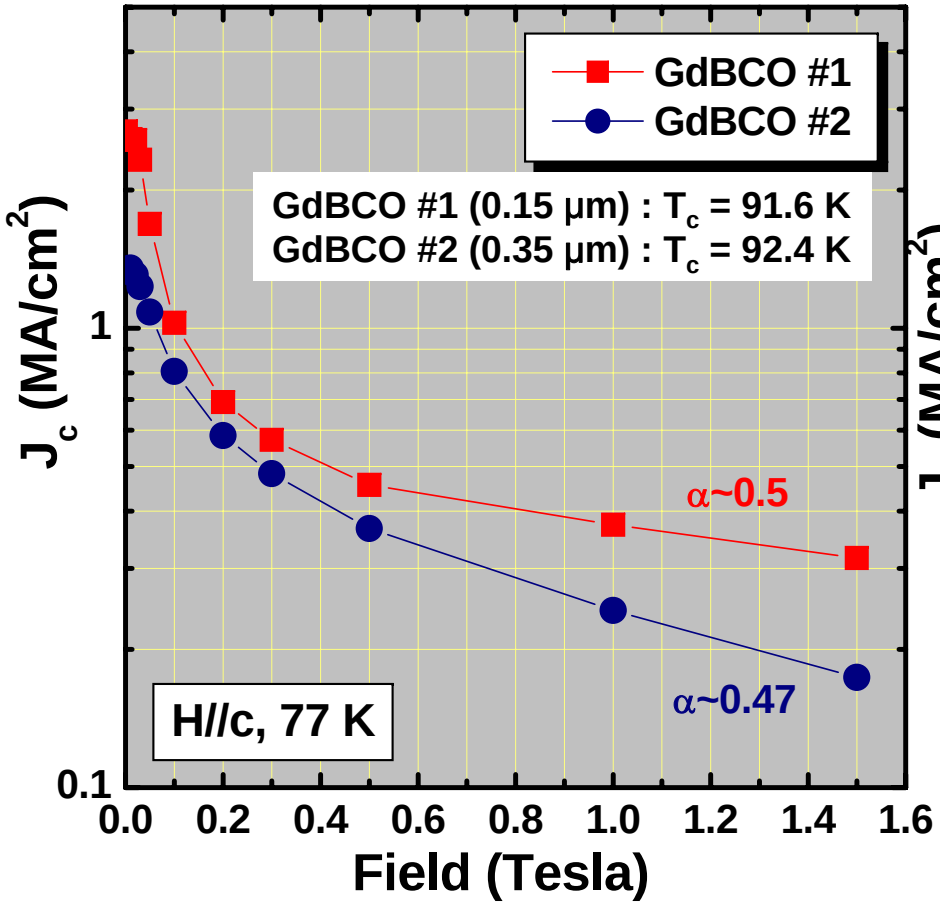
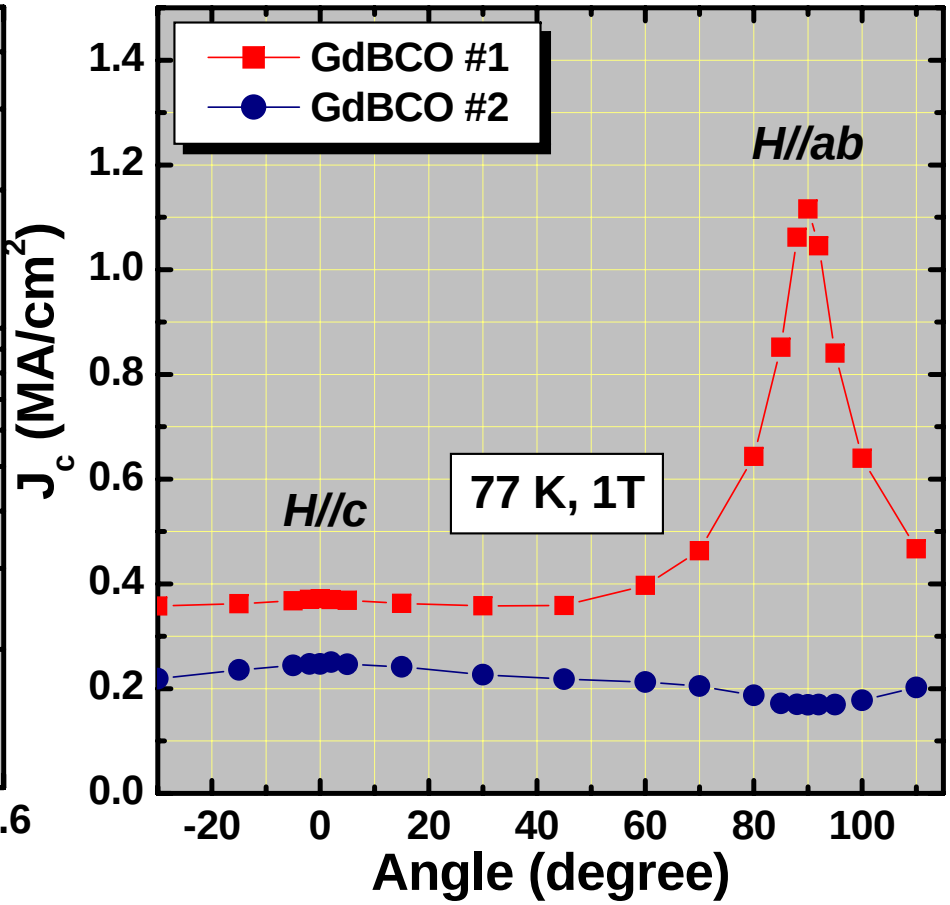
More pronounced effects seen for Gd-123 films

Pinning properties controlled via PLD processing condition (condition 1 and 2)





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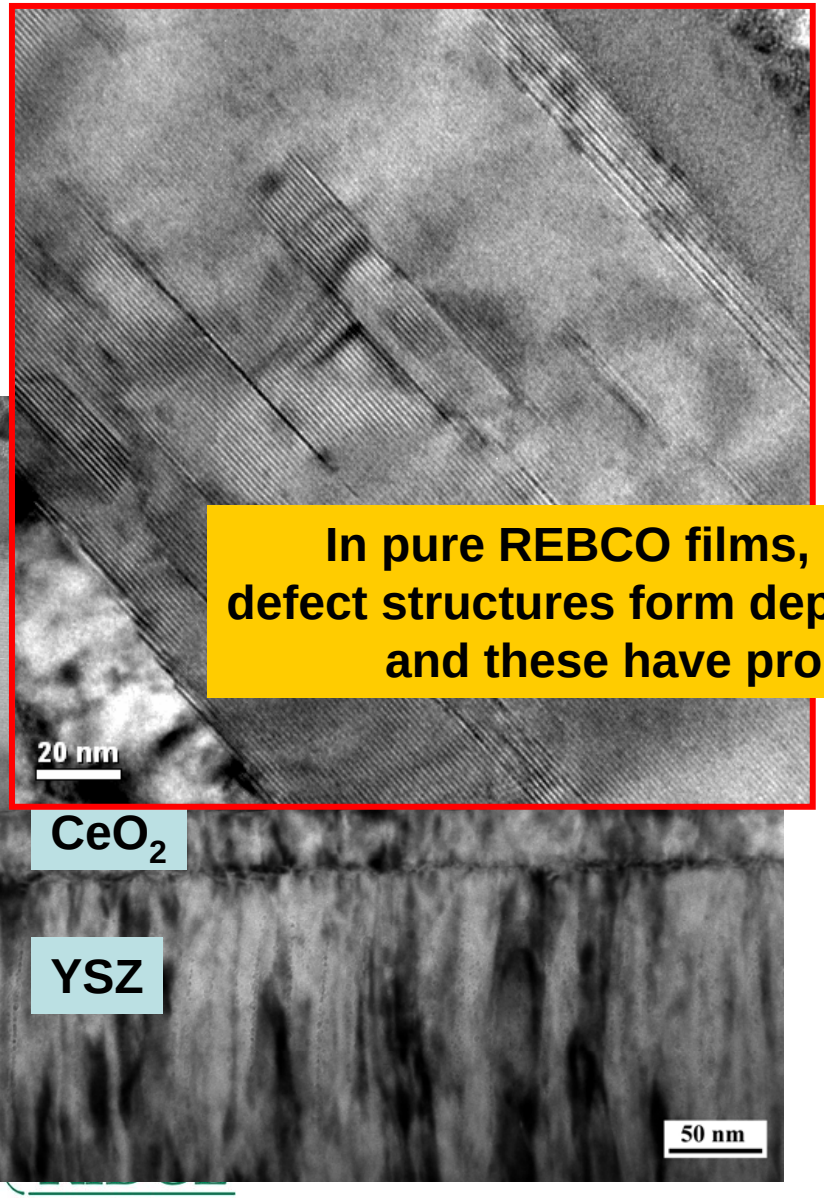
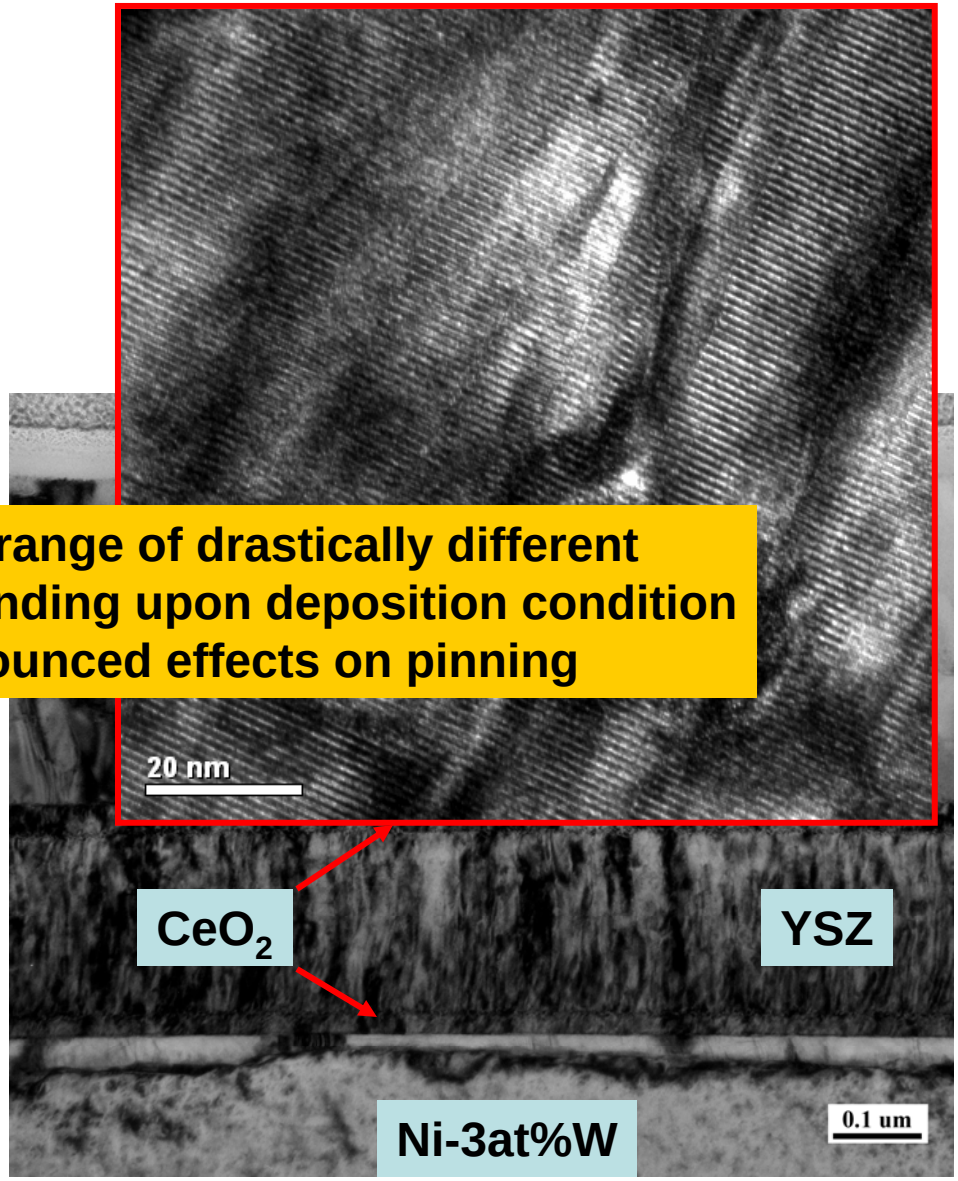
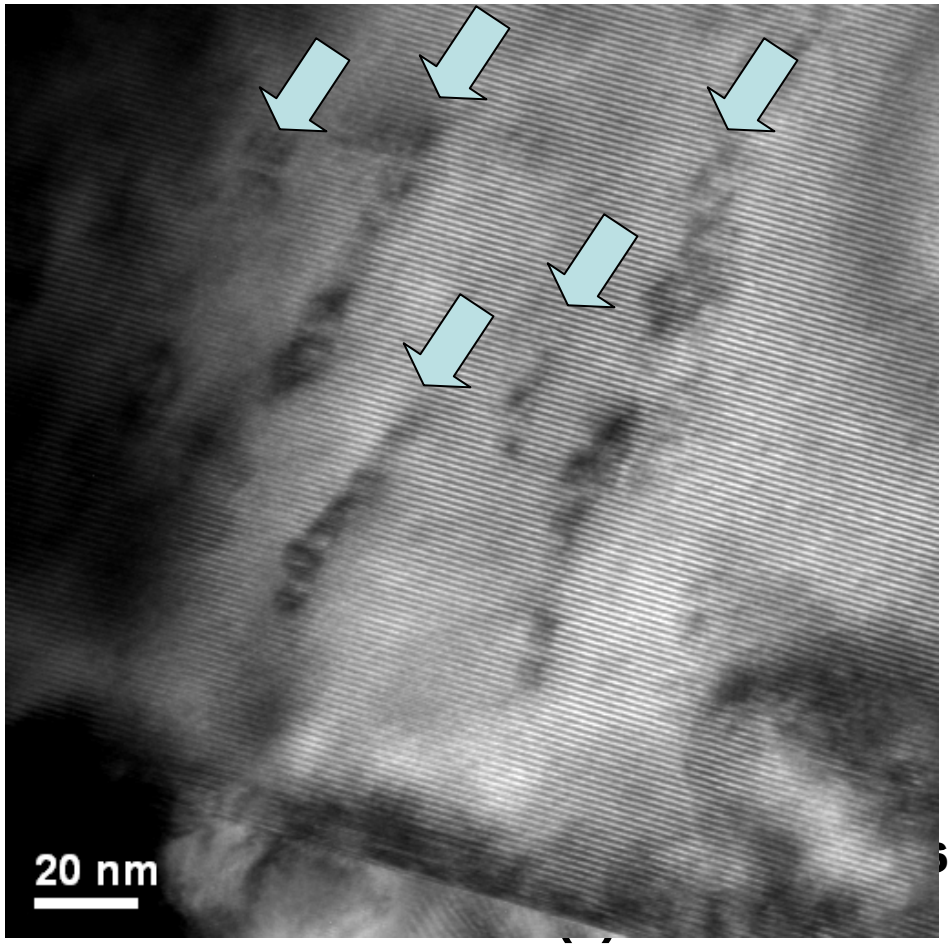
Film with H//ab peak has long stacking faults and film with dip for H//ab, has a highly defected film

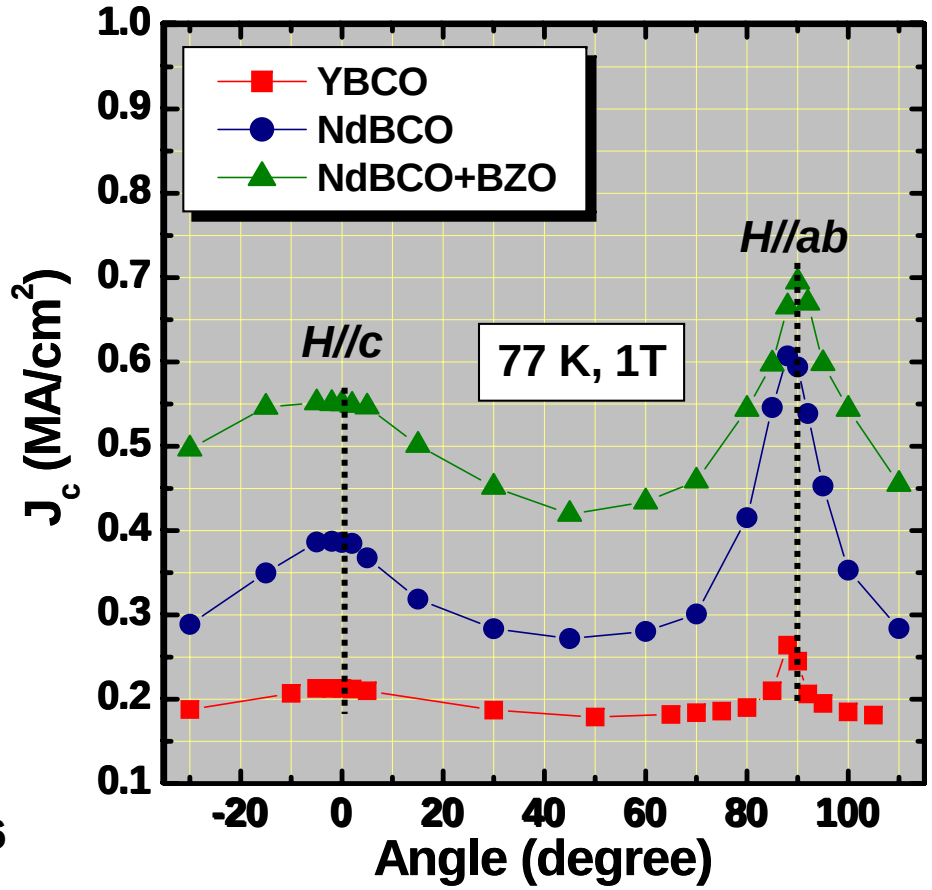




In pure REBCO films, a range of drastically different defect structures form depending upon deposition condition and these have pronounced effects on pinning

Effect of incorporation of columns of self-assembled BZO in 0.2 μm thick Nd-123 films on RABiTS





YBCO: α = 0.52
NdBCO: α = 0.43
NdBCO+BZO: α = 0.37

All deposition from here on: condition 1

For thicker films: New target – BZO very well mixed, denser target of 2” diameter

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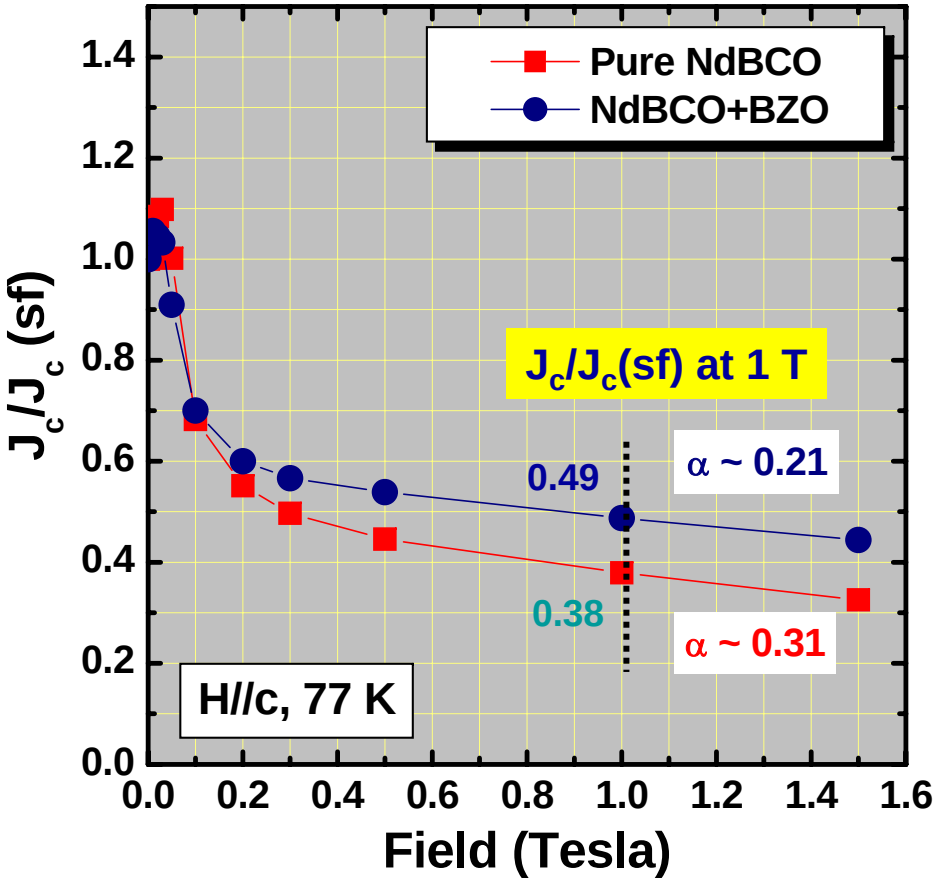
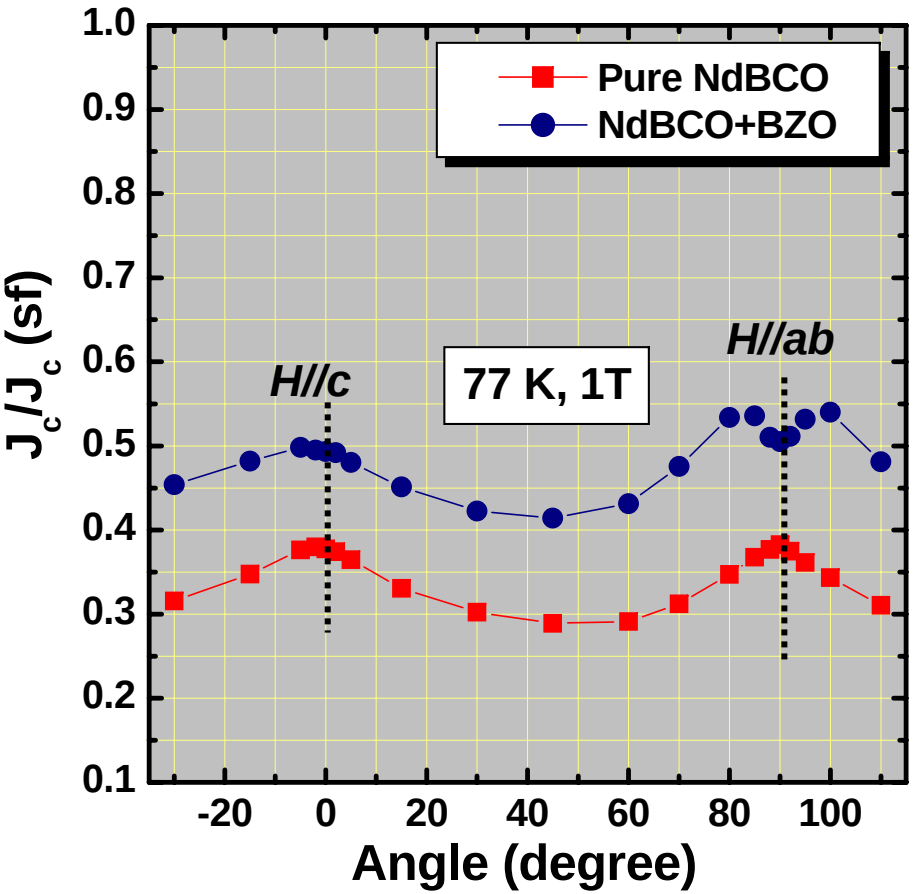
NdBCO (~0.6 μm) with and w/o columnar defects of BZO





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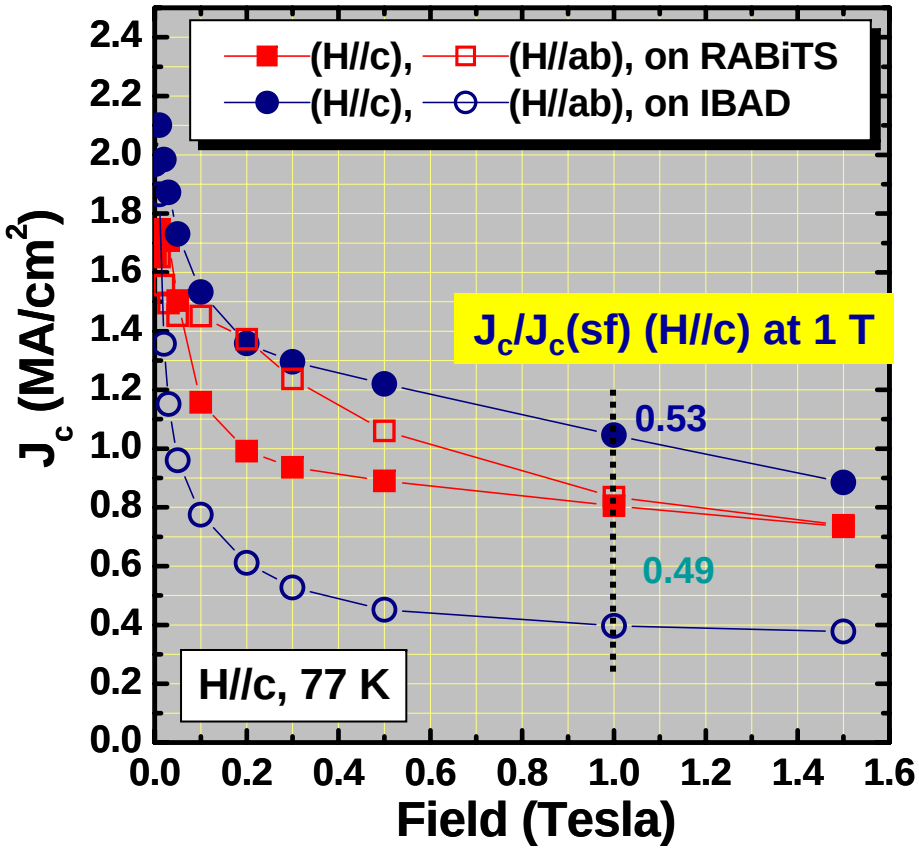
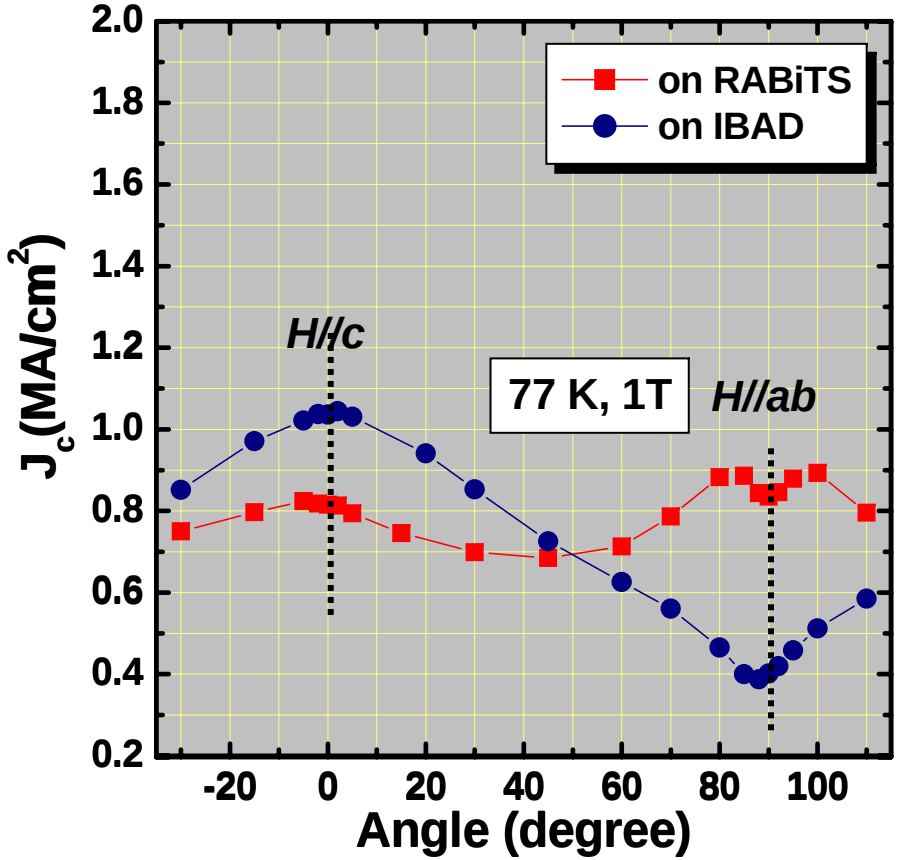
NdBCO+BZO samples : 0.6~0.7 μm thick films





	(006) R.D.	(006) T.D.	(113) Φ-scan	True Φ
on RABiTS	4.09	4.45	7.09	6.4
on IBAD	1.69	2.12	4.04	3.2

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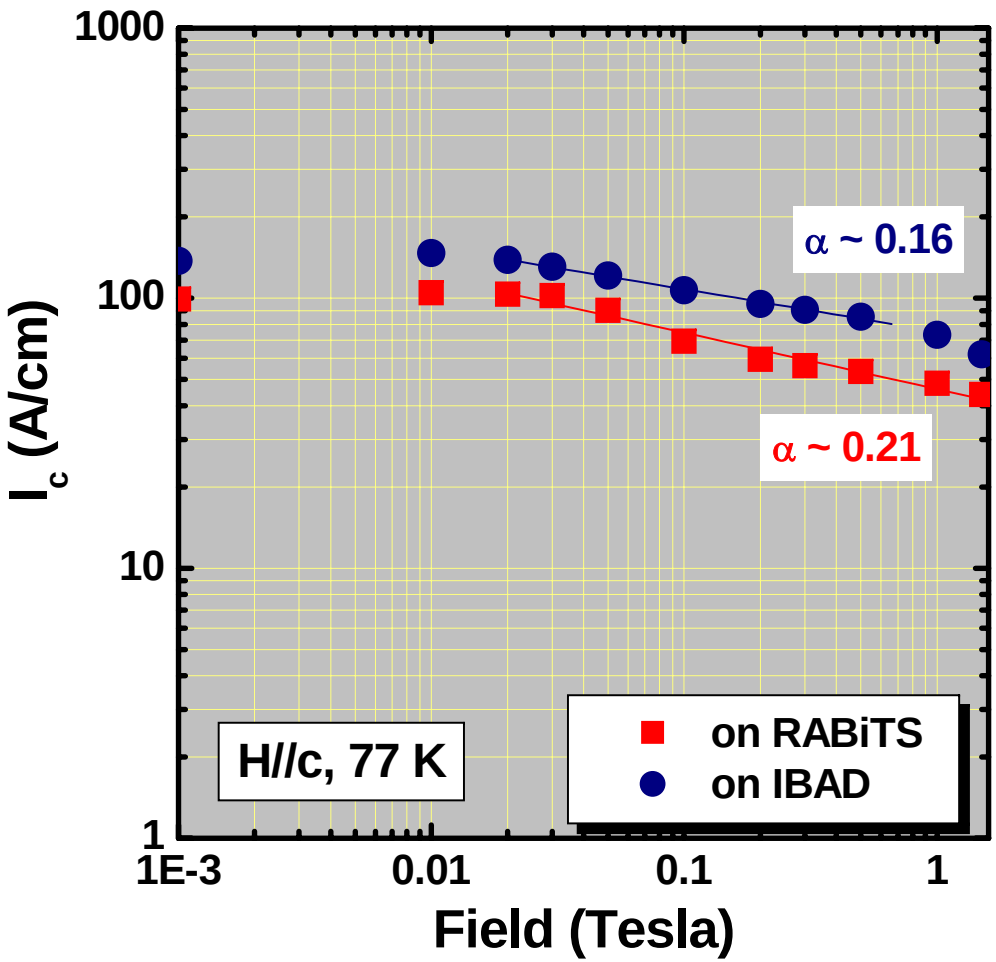
For both films α is much smaller than for YBCO+BZO



For YBCO + BZO: α ~ 0.3

Does this significantly lower α imply a very different defect structure in the film?

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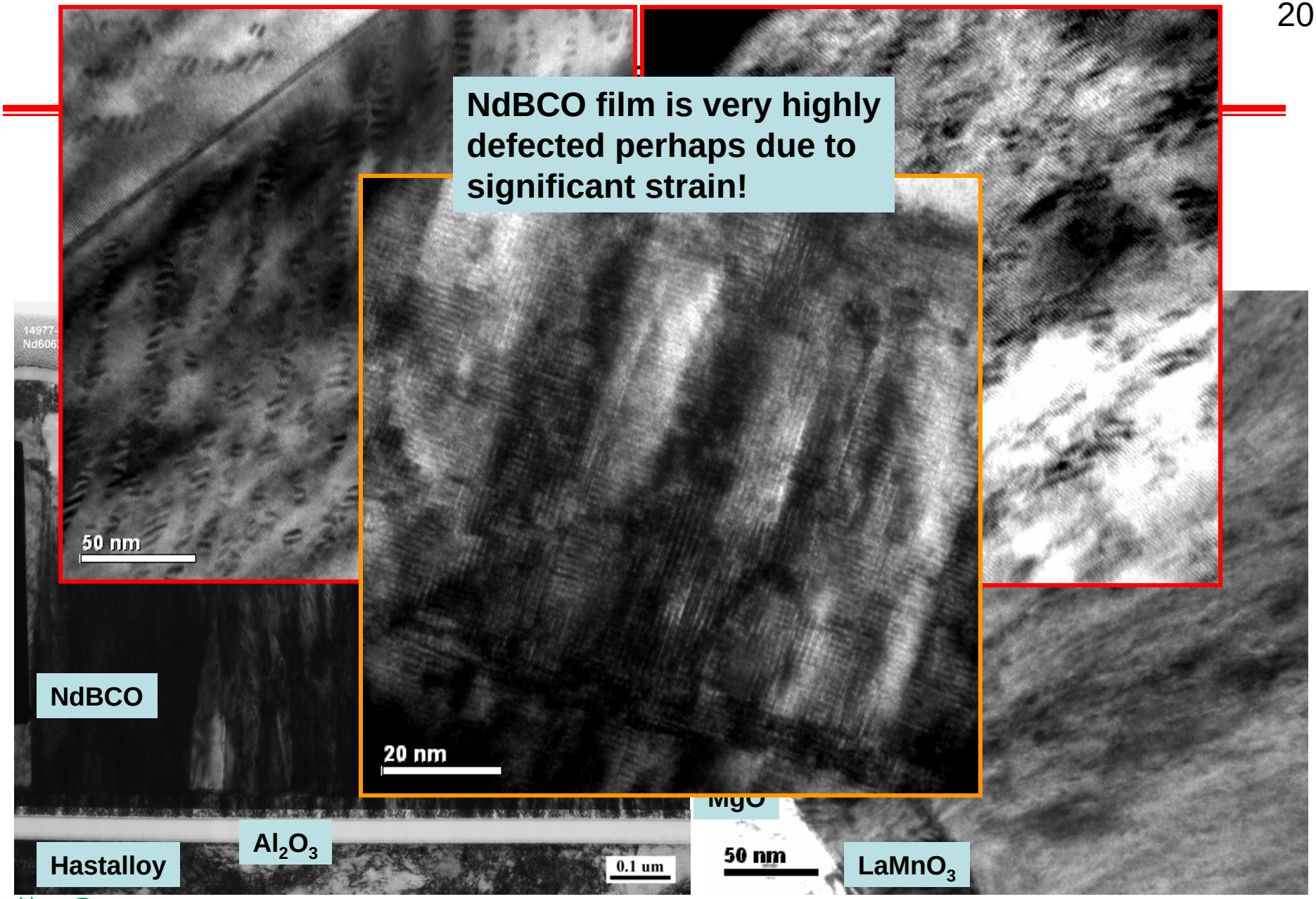
NdBCO film is very highly defected perhaps due to significant strain!



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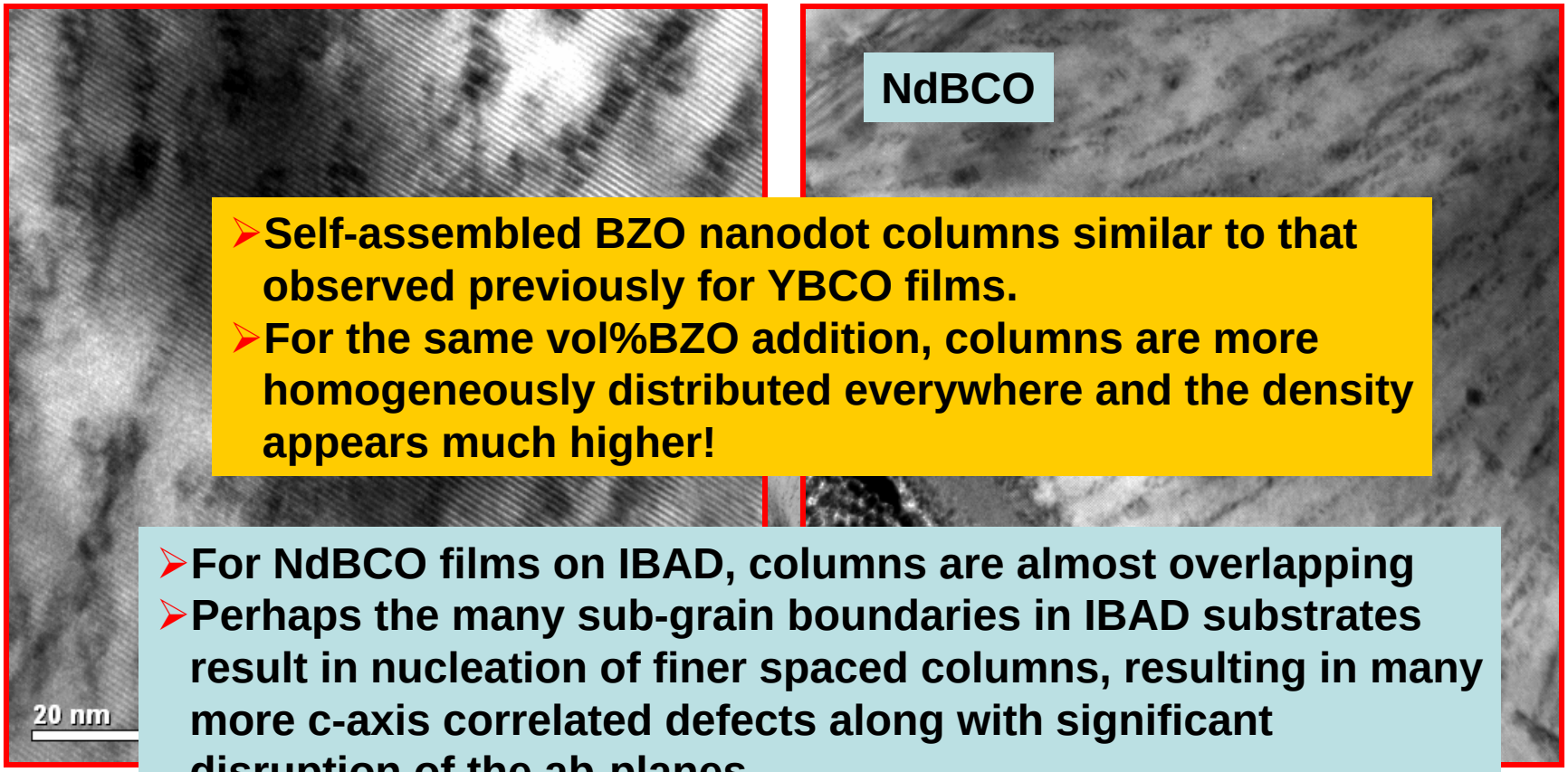
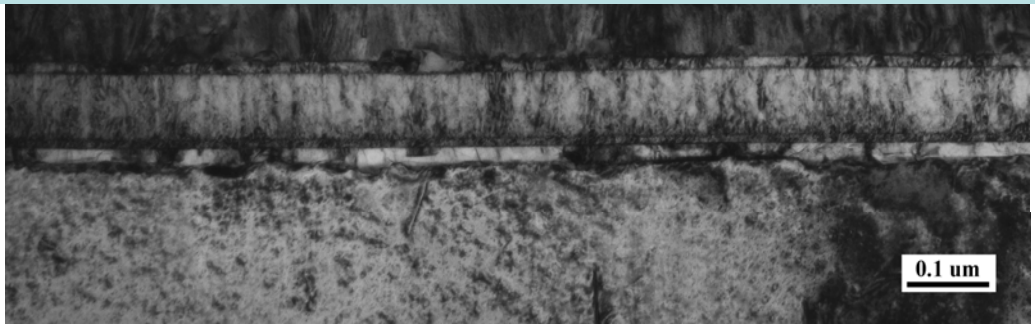
NdBCO film on RABiTS show a finely spaced BZO nanodot columns everywhere in film

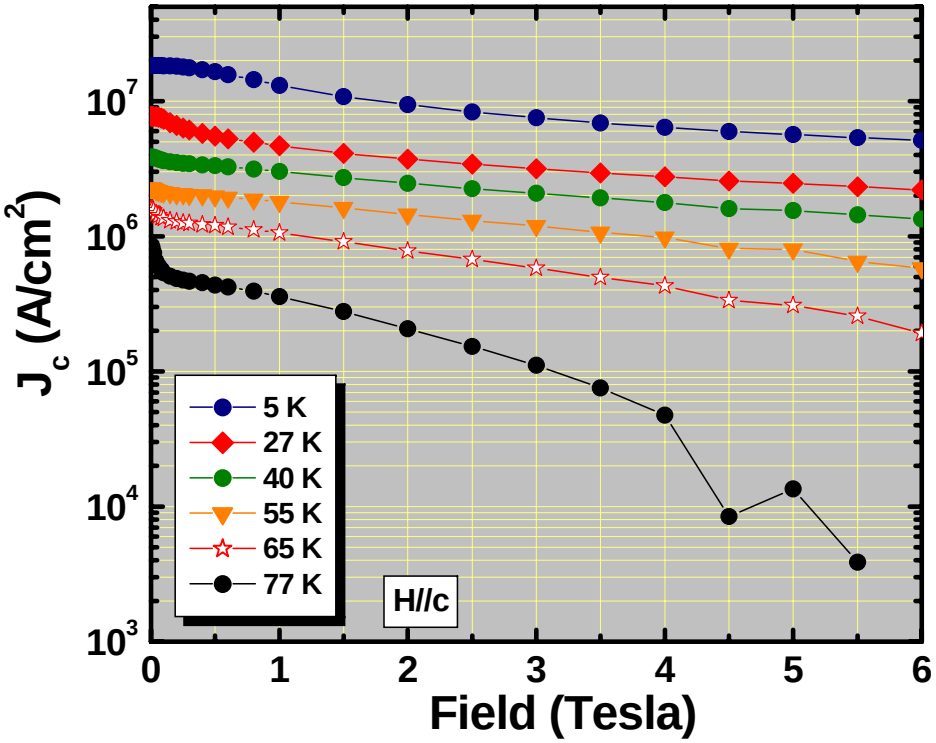


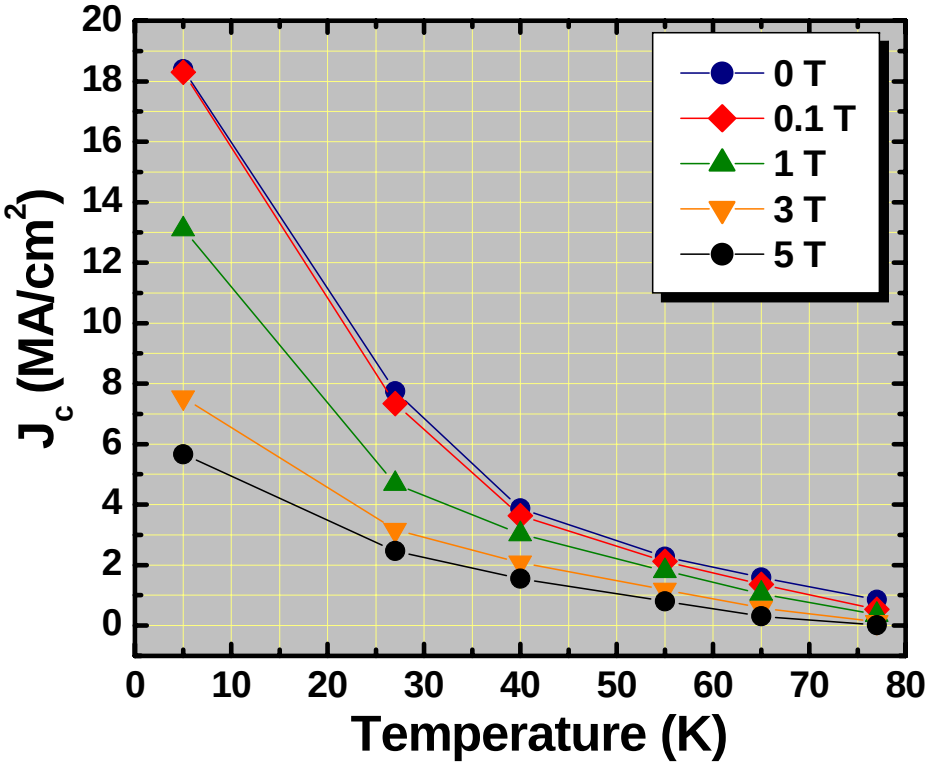
- Self-assembled BZO nanodot columns similar to that observed previously for YBCO films.
- For the same vol%BZO addition, columns are more homogeneously distributed everywhere and the density appears much higher!

- For NdBCO films on IBAD, columns are almost overlapping
- Perhaps the many sub-grain boundaries in IBAD substrates result in nucleation of finer spaced columns, resulting in many more c-axis correlated defects along with significant disruption of the ab-planes



For film on IBAD, J_c is very high for H//c at lower measurement temperatures





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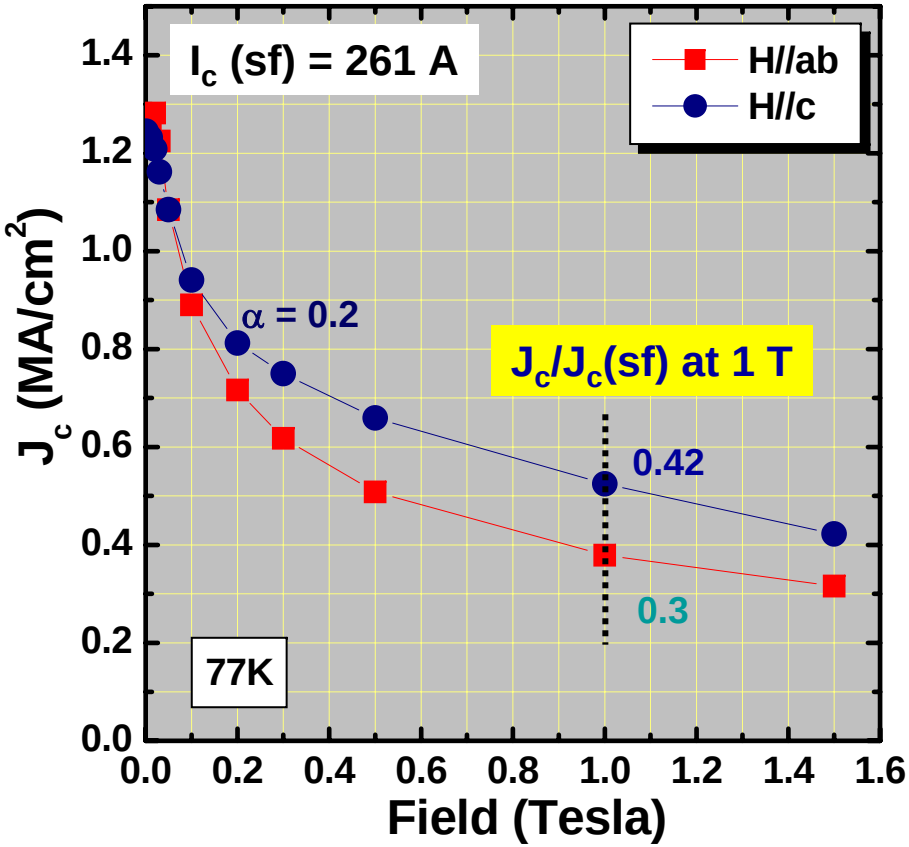
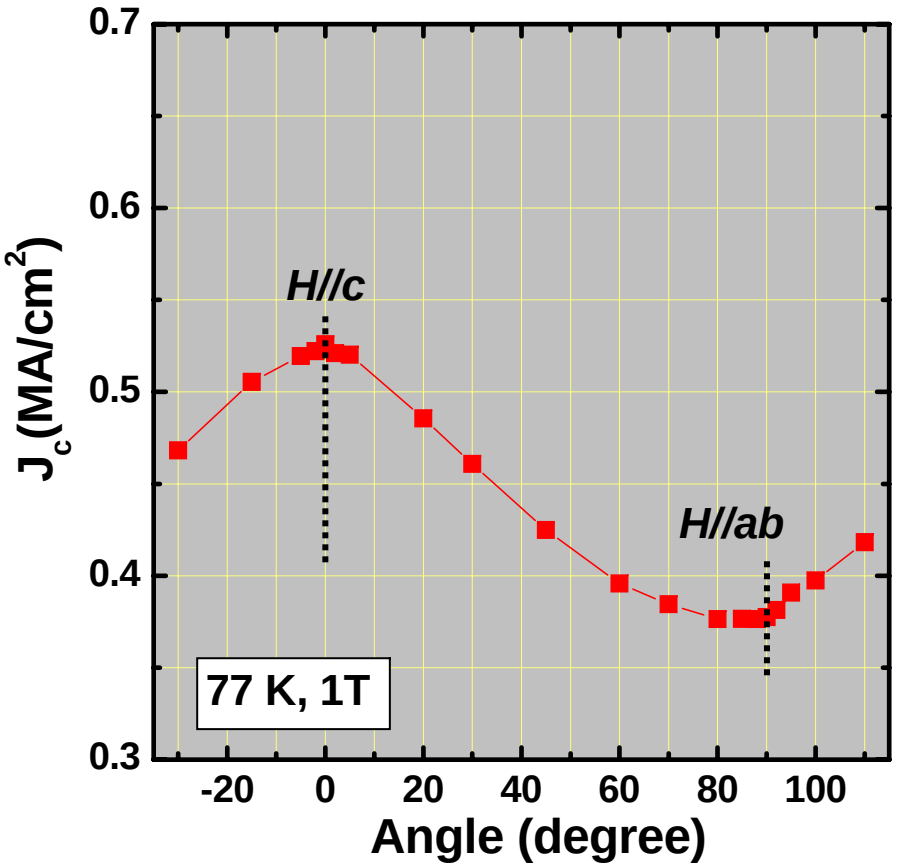
For 2.1 µm thick film on IBAD substrate, similar transport behavior is observed





(006)	(006)	(113) Φ-scan
R.D.	T.D.	
1.66	1.94	3.5

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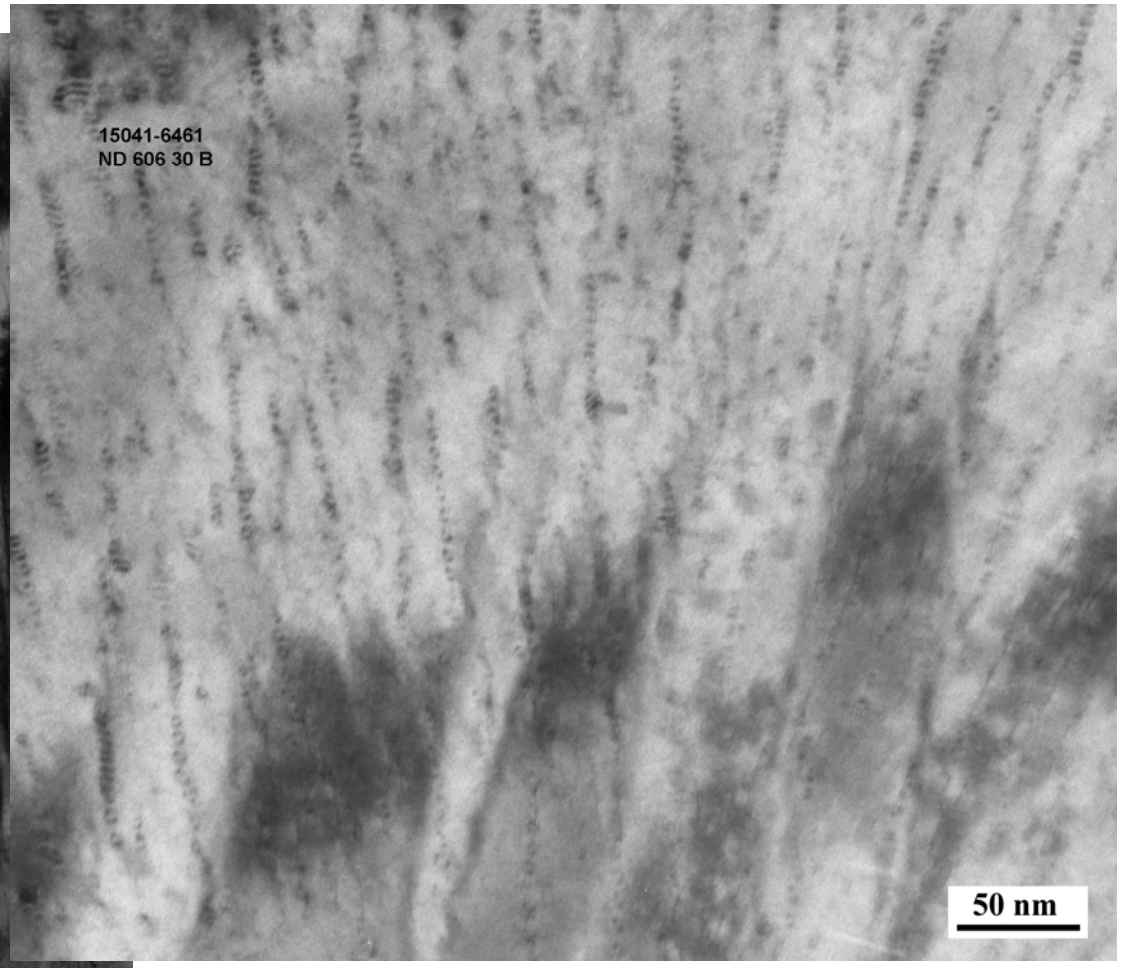
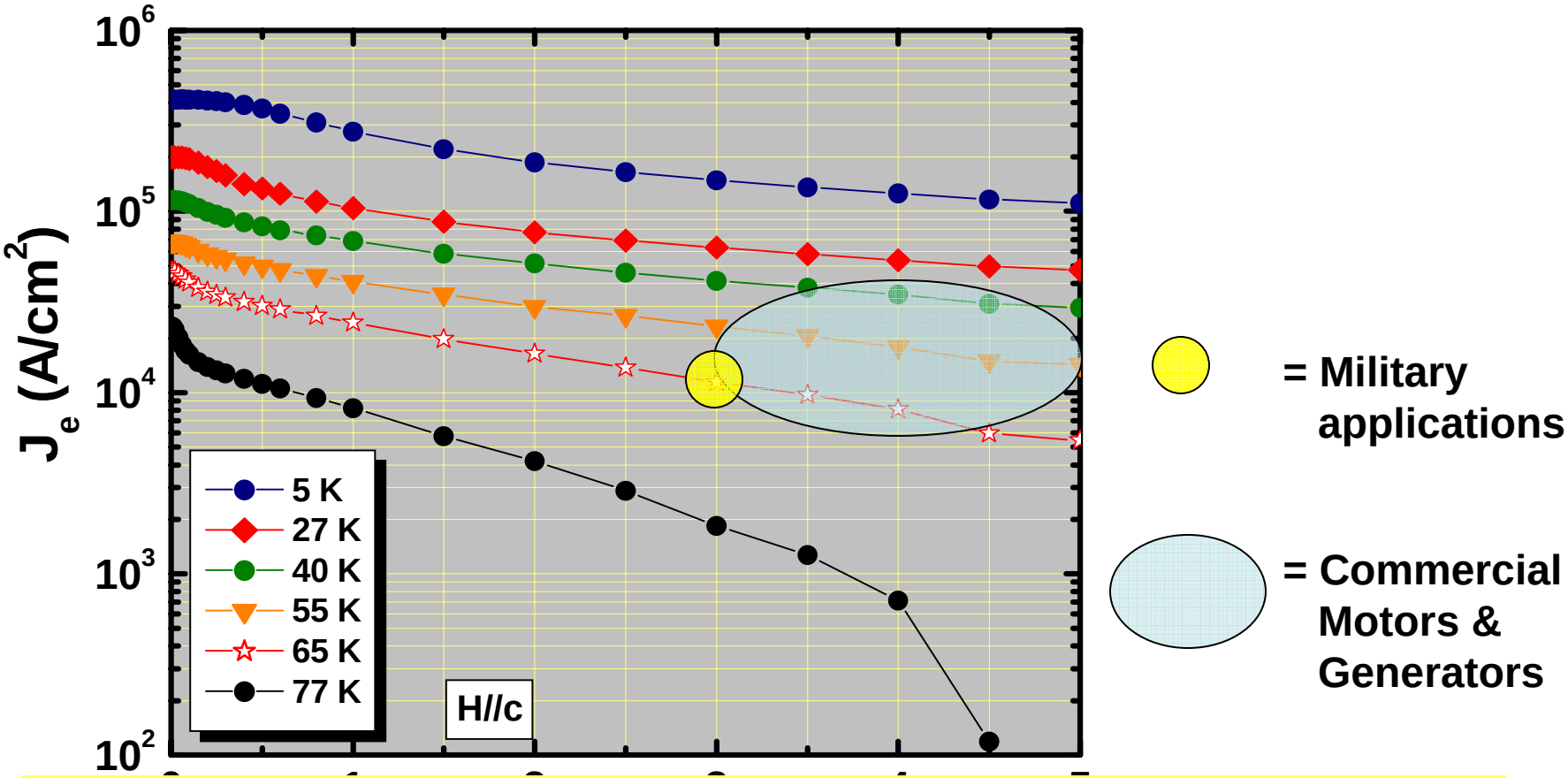
Sub-grain morphology is easily observable in the 2.1 μm thick PLD film on IBAD LMO/MgO/Hastalloy substrates



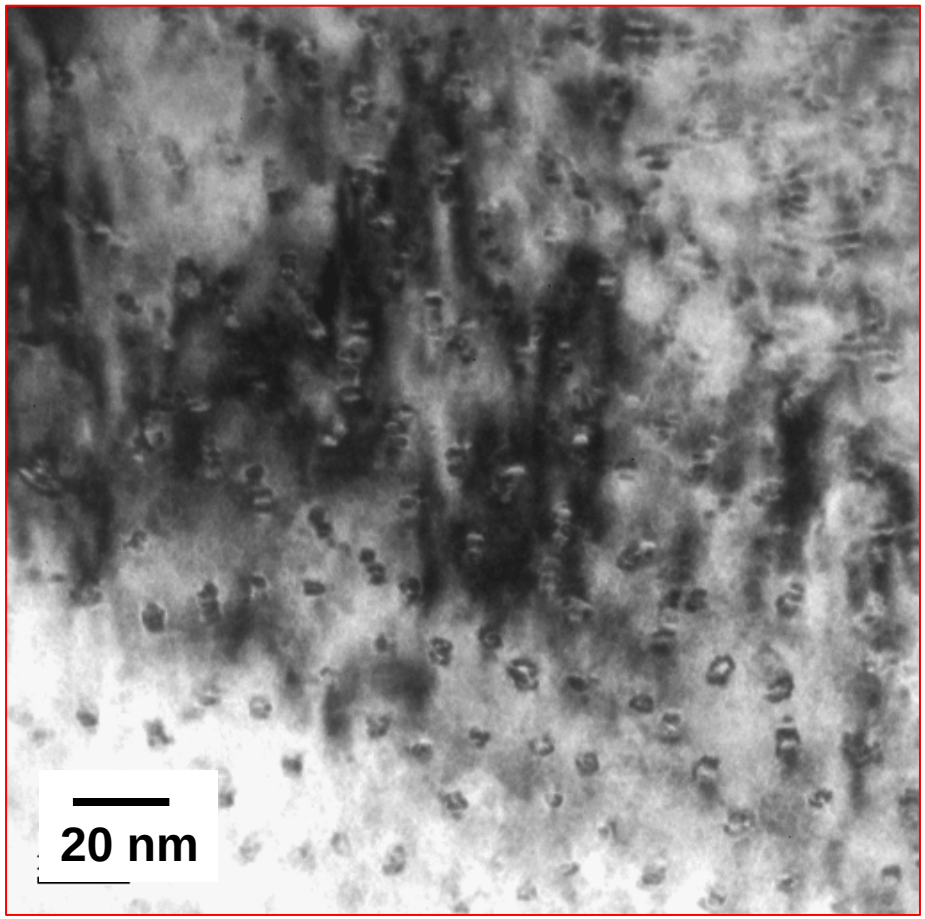
Magnetization I_c for 2.1 µm thick film on IBAD substrate at lower temperatures of interest



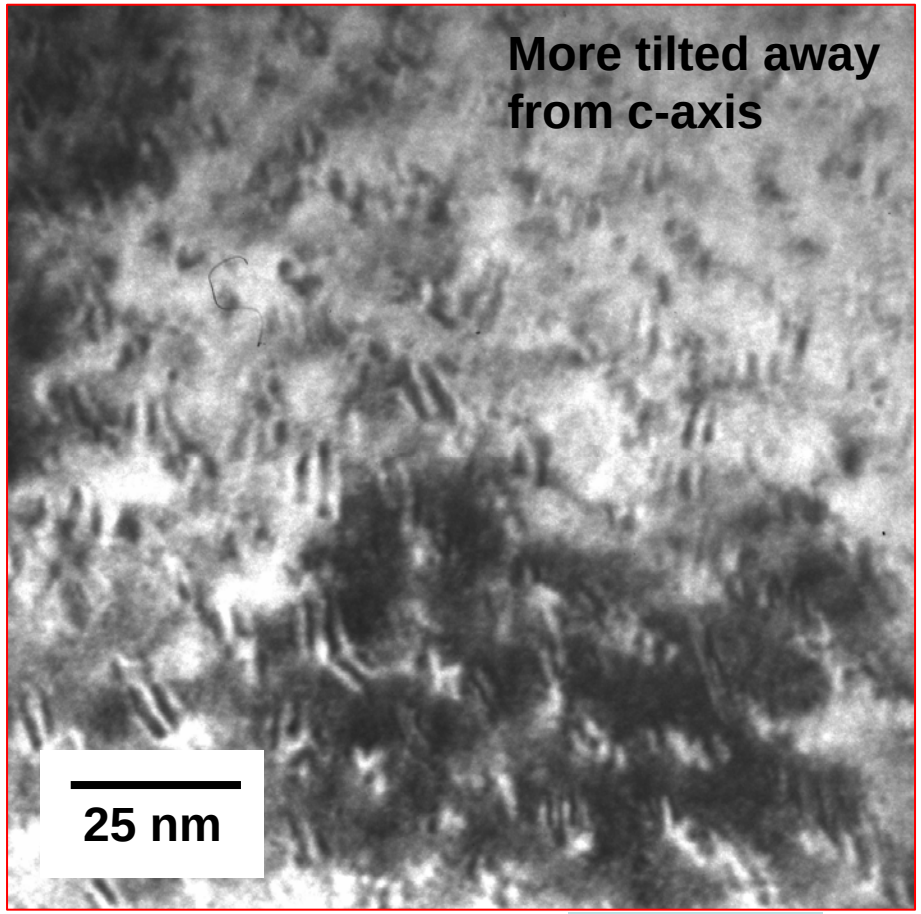
<u>SUMMARY REBCO films</u>: Columns of self-assembled nanodots of BZO within REBCO films result in significant enhancement in the critical current density for H//c.

Why does self-assembly of nanodots occur? What dictates the size of nanodots and the orientation of columns?

2 vol% BZO addition



4 vol% BZO addition

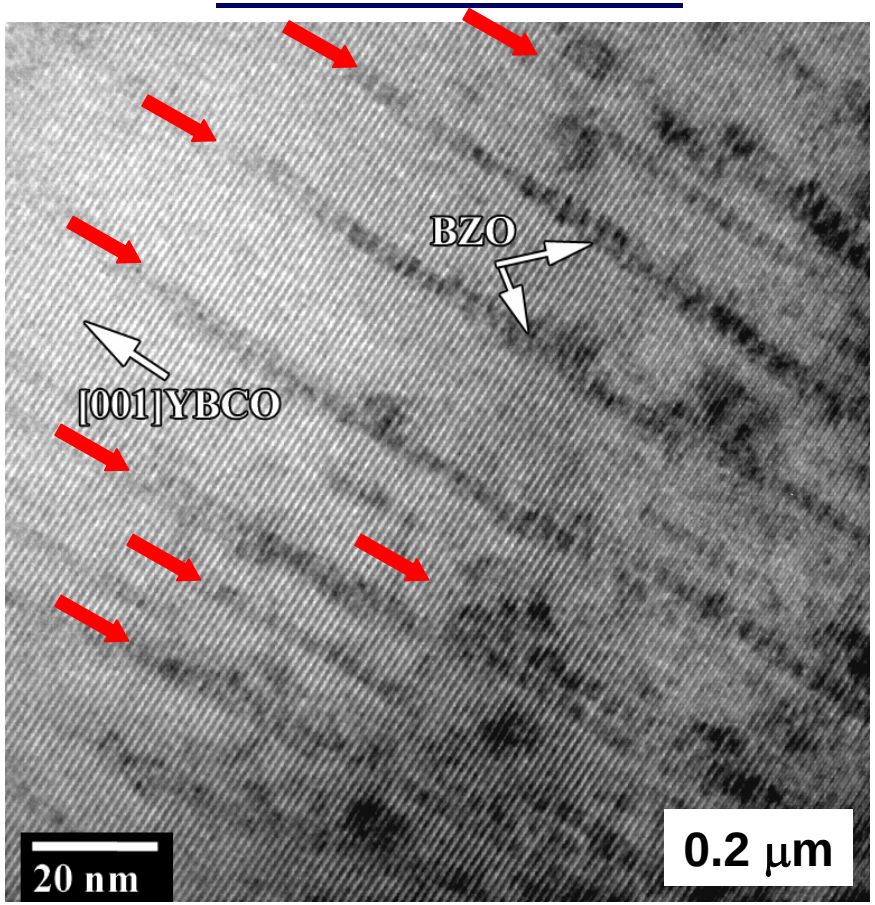


Some increase in column width with vol%BZO

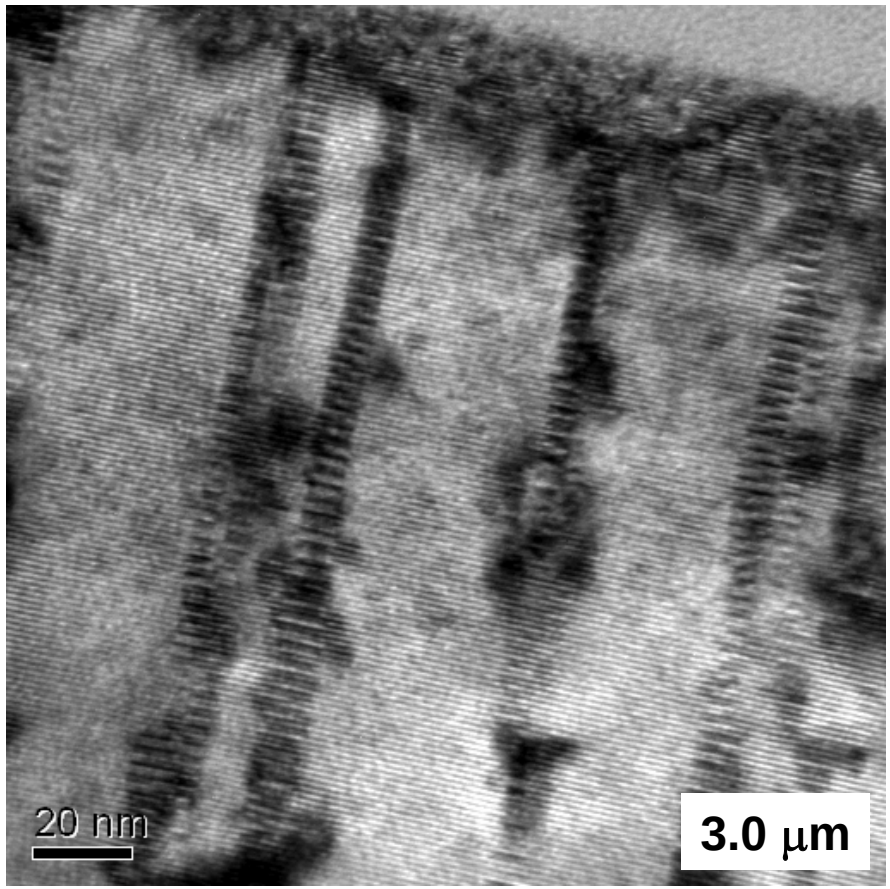
Imaging conditions different

For the same volume fraction of BZO, column width increases with film thickness

2 vol% BZO addition



2 vol% BZO addition



Column size increases with thickness

Different deposition conditions

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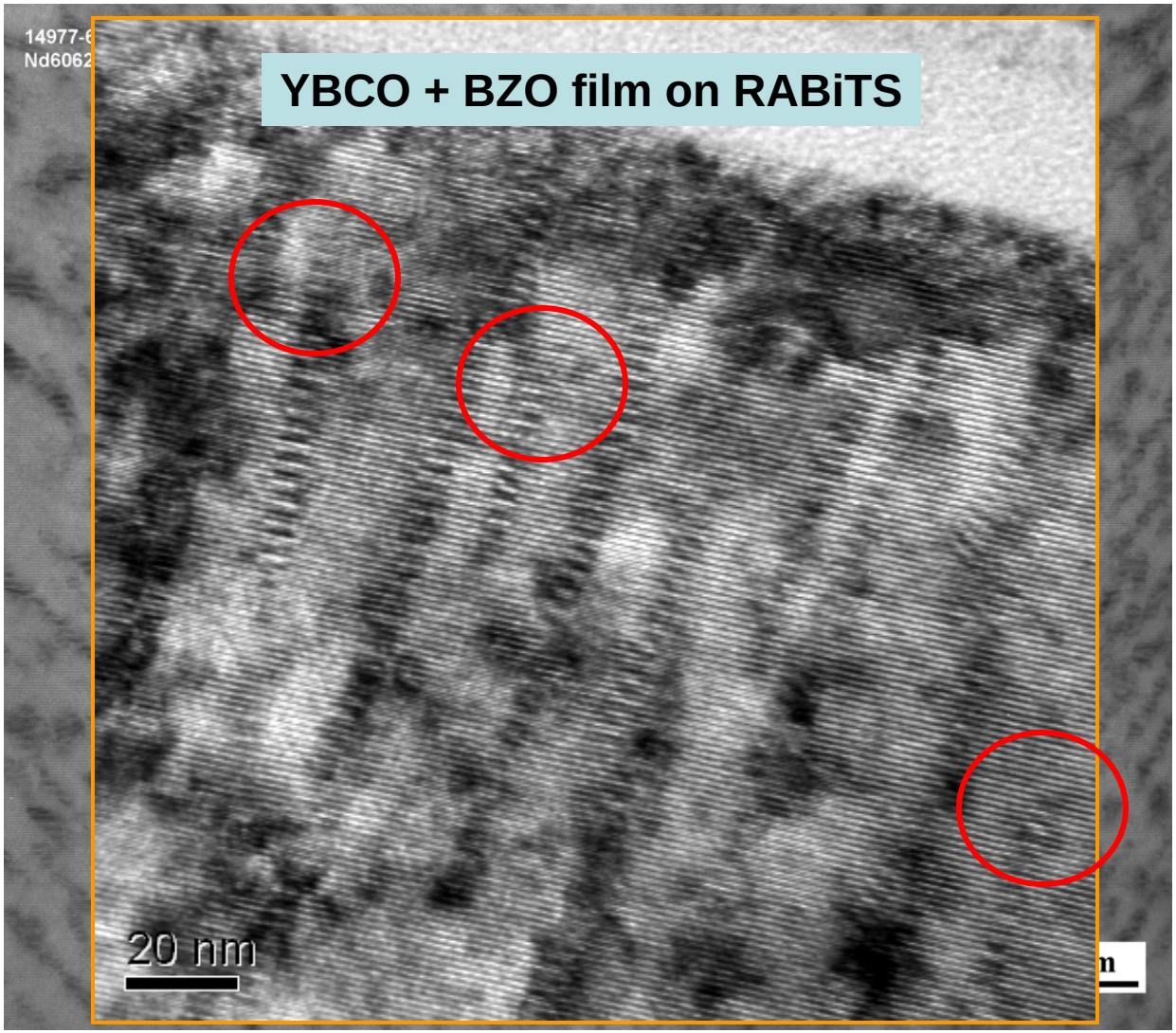
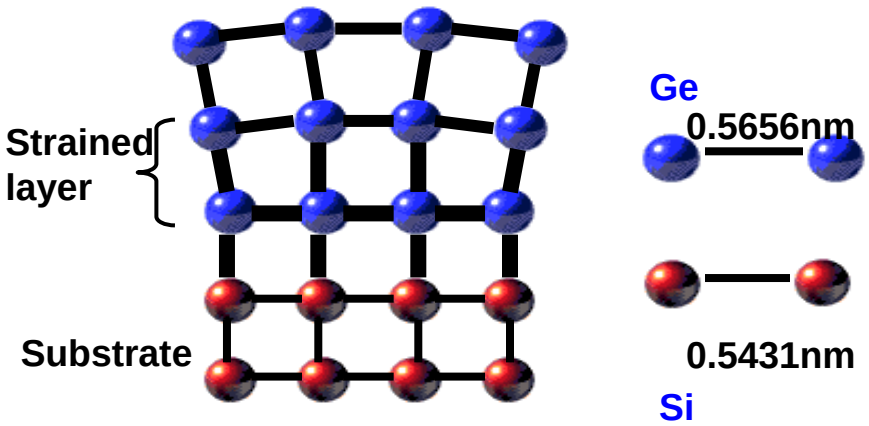
What controls the column termination?



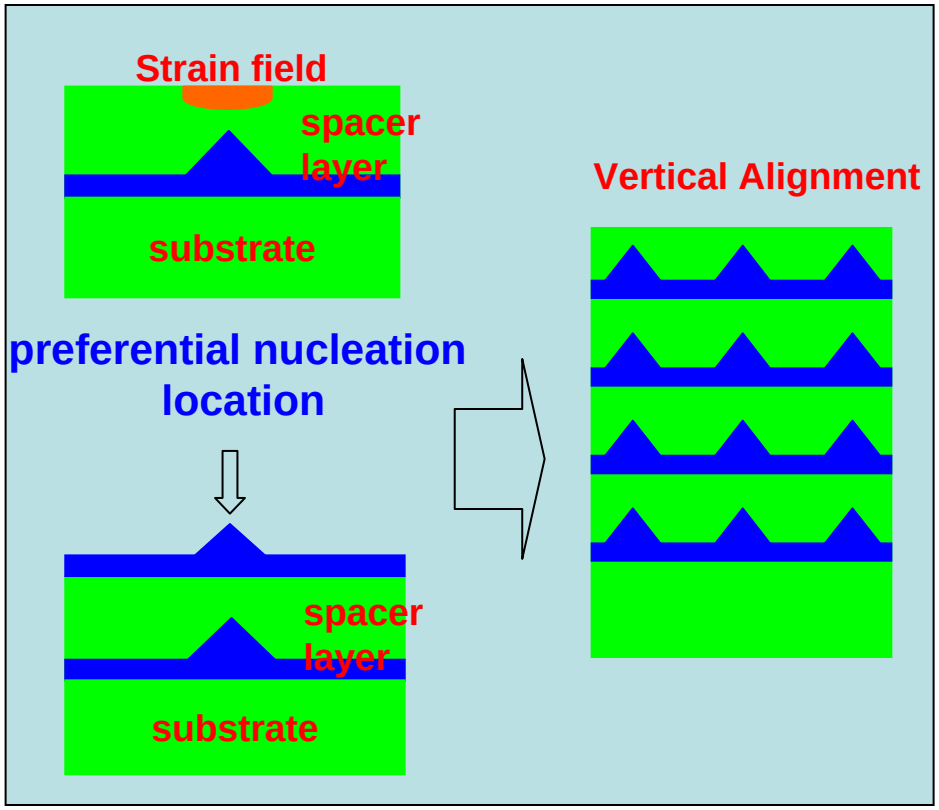
Self-Organization of Coherent Islands in Multilayer Heteroepitaxial Growth

Island formation due to lattice mismatch

Example: Ge/Si



Strain-induced Vertical Alignment of Embedded Quantum Dots in Simple Systems during in-situ multilayer growth



Strain-Mediated Phase Ordering

J. Zhong
ORNL

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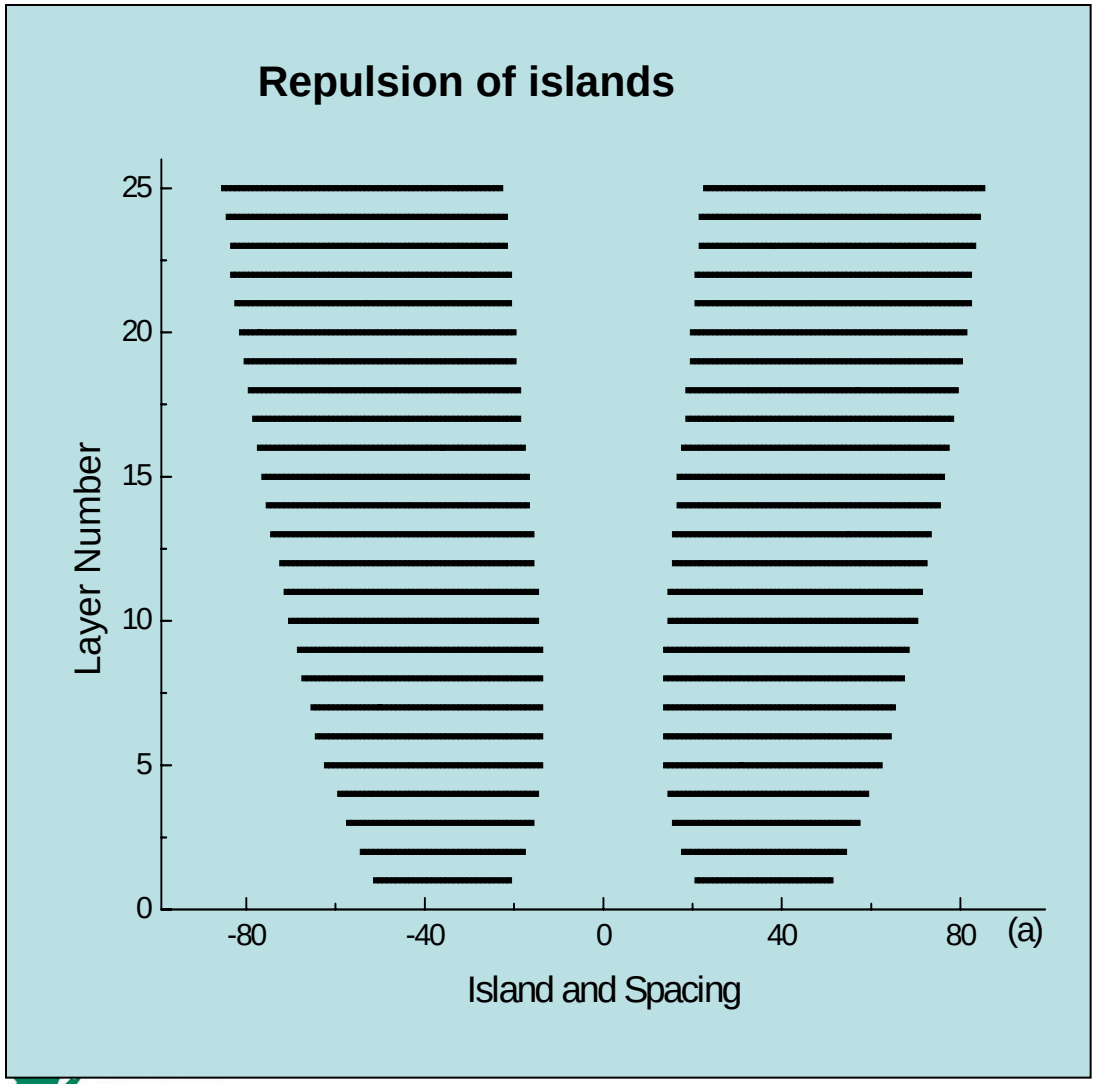
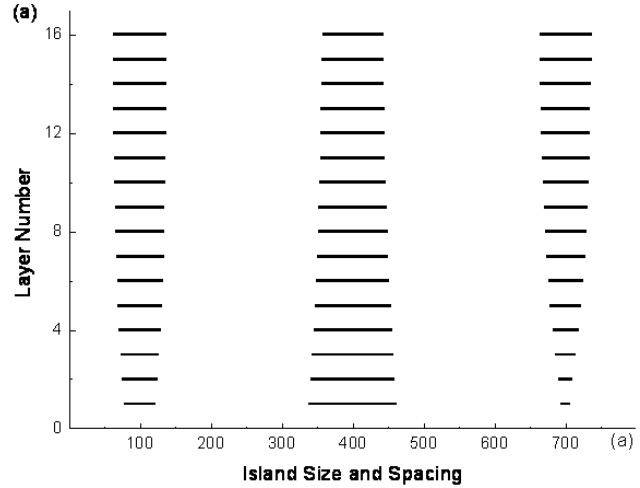
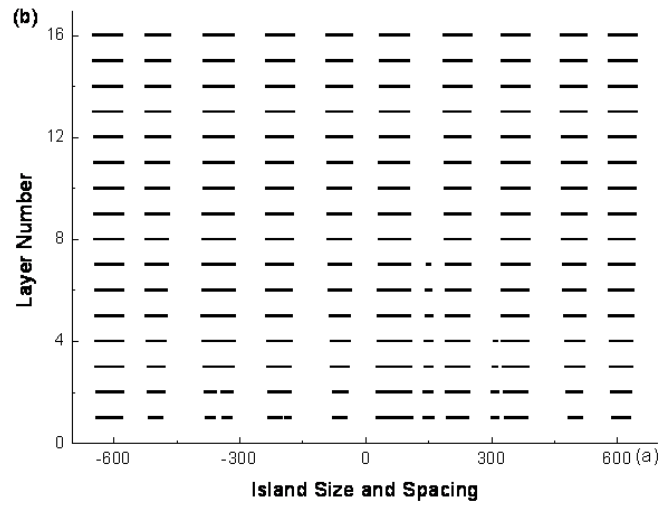
Interaction of Strain Fields and Self-Organization of Coherent Islands in Multilayer Heteroepitaxial Growth





Sparse distribution: vertical ordering



Dense distribution:
vertical and lateral ordering

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Si-Ge: Phase Ordering

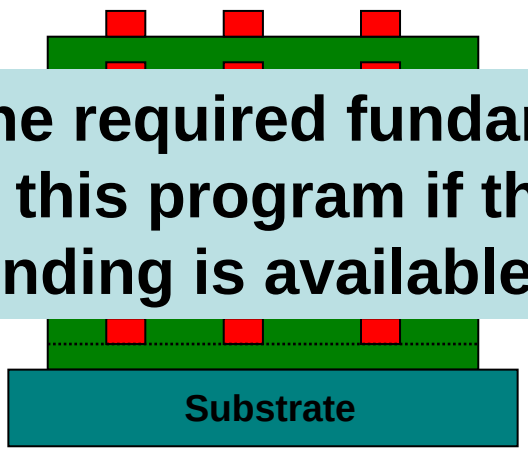
BZO-YBCO: Simultaneous Phase Separation & Phase Ordering

Multi-targets

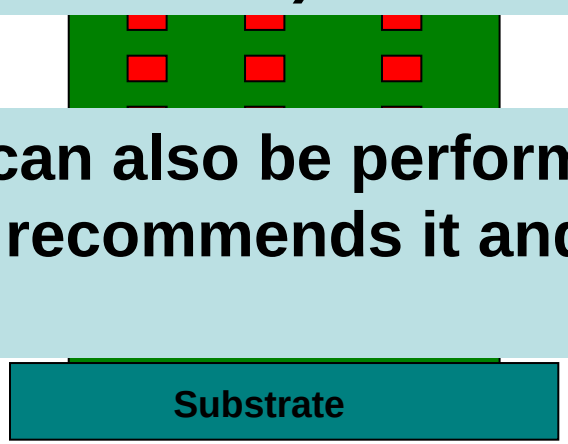
Single target

We have an internal research proposal to understand self-assembly via SPSO using atomistic and continuum models and also accounting for kinetics via Monte-Carlo simulations (with Malcolm Stocks, J. Zhong and Y. Gao).

The required fundamental work can also be performed in this program if the committee recommends it and the funding is available



Stress-mediated ordering in multilayer sequential deposition



Stress-mediated ordering in simultaneous deposition

Presentation Outline

- **FY 2006 Results**

  Flux pinning in YBCO & REBCO films with columns of insulating, self-assembled nanodots **(Goyal)**

  Systematics of BZO doped YBCO **(Christen)**
  J_c analysis for Y_2O_3 doped YBCO ex situ vs thickness
  Nanoparticle substrate decoration for J_c enhancements

  Epitaxial growth of buffer layers **(Parans)**

- **FY 2006 Performance and FY 2007 Plans** **(Parans)**
- **Research Integration**

Goal: Correlate YBCO/RABiTS properties with $BaZrO_3$ doping levels

Objectives and Approach:

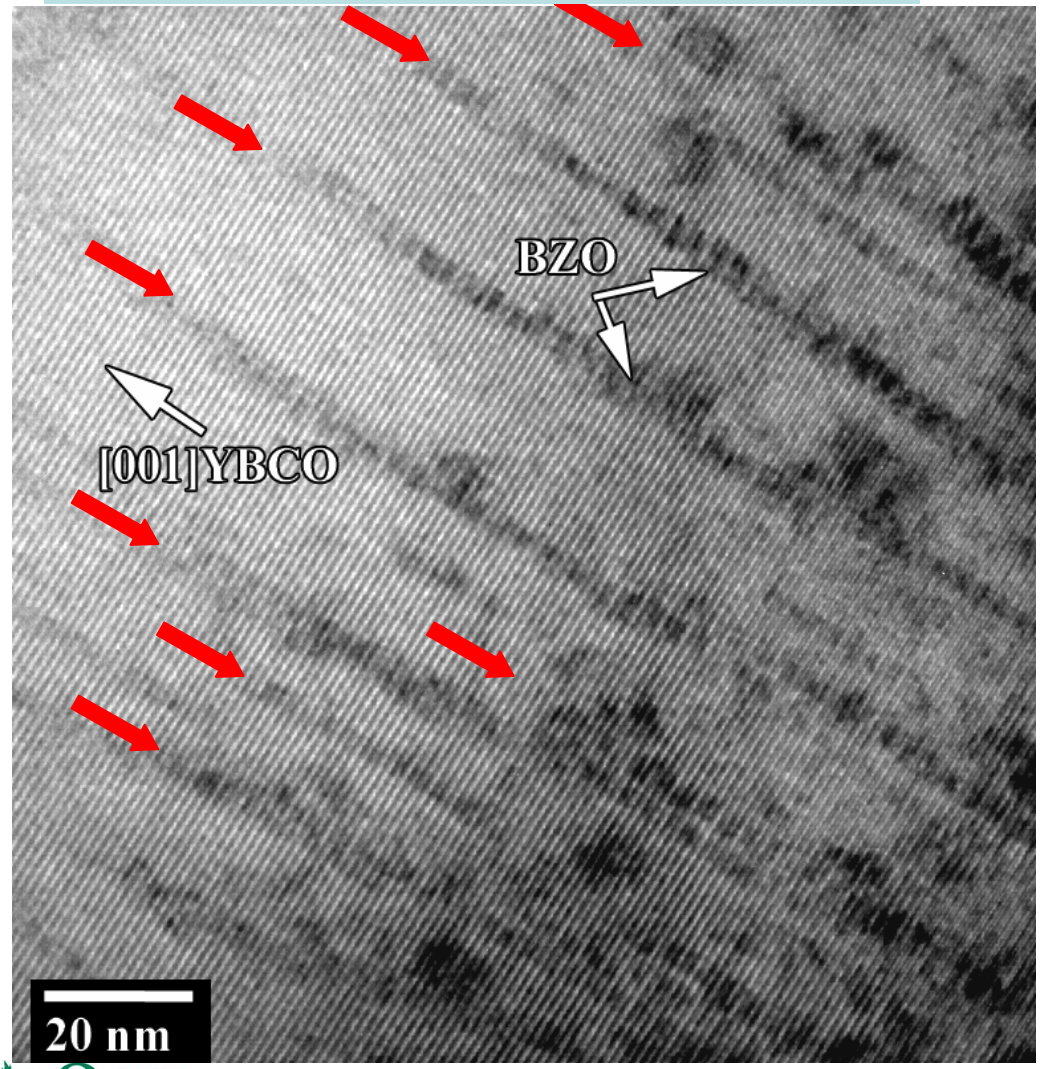
- **Determine systematic effects of BZO concentration on superconducting properties T_c, $J_c(H,T)$**
  - **For series of films, volume fraction x of BZO varied up to 8%**

- **Analyze results in terms with recent theoretical models for effects of columnar defects**
  - **BZO nanoparticles stacks are approximate columnar defects**
  - **Account for effects on both pinning and equilibrium properties**

- **From this analysis, generate predictive guidance for optimization of J_c of coated conductors**

Self-assembly during PLD of $BaZrO_3$ in YBCO

Cross-section TEM of YBCO/RABiTS



PLD from YBCO-2%BZO target

Nanodots and nanorods self-assemble during growth

Has been extended to short samples of 2G wire

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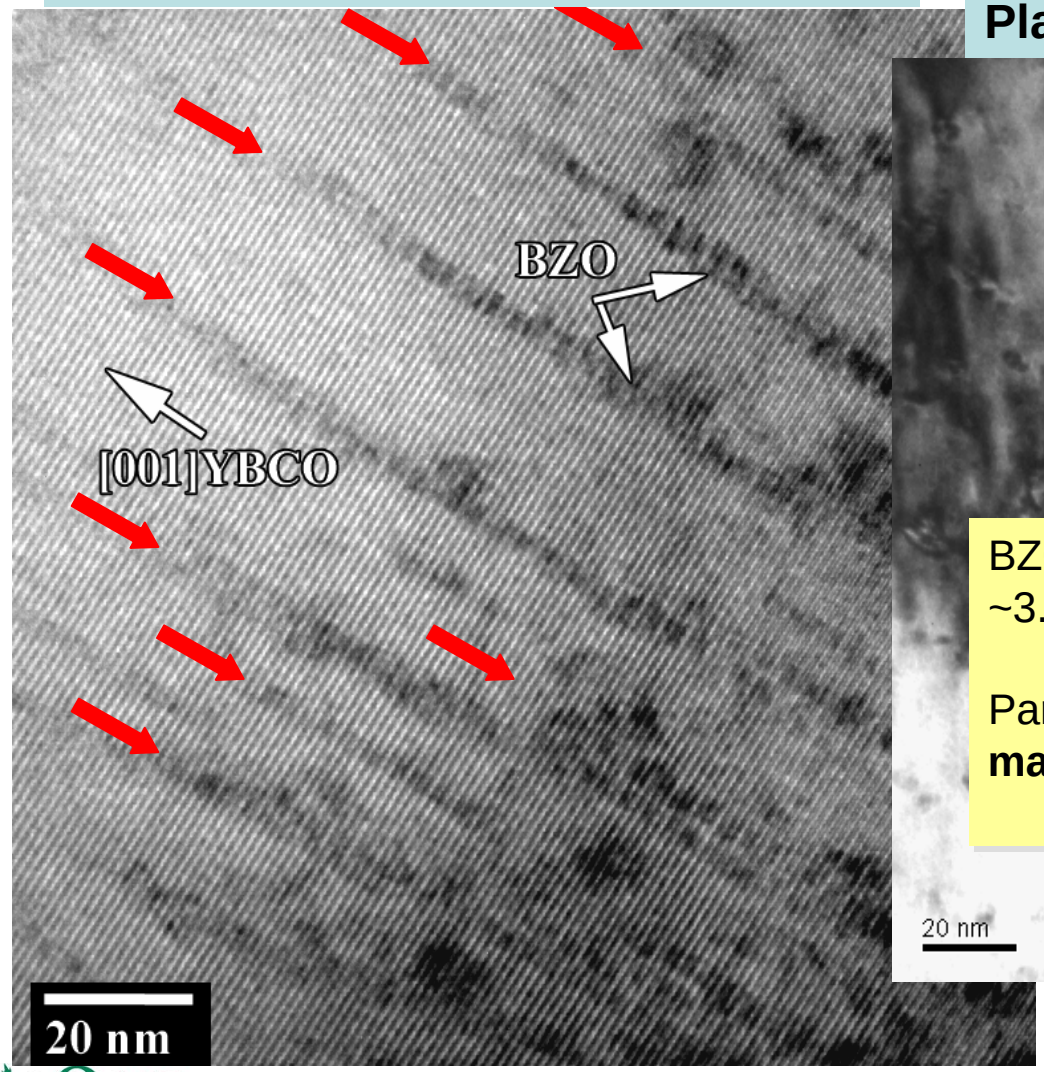
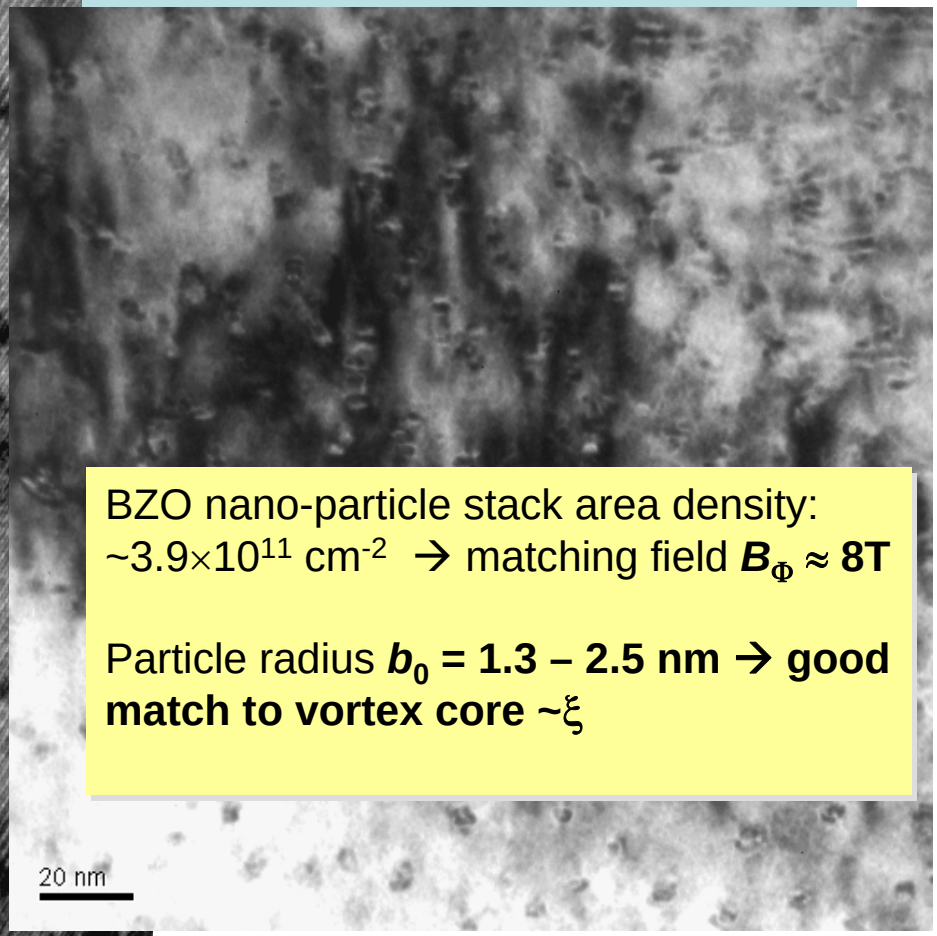
Self-assembly during PLD of $BaZrO_3$ in YBCO

Cross-section TEM of YBCO/RABiTS



Plan view TEM of YBCO/RABiTS



BZO nano-particle stack area density: ~3.9×10^{11} cm^{-2} → matching field $\boldsymbol{B_\Phi \approx 8T}$

Particle radius $\boldsymbol{b_0}$ **= 1.3 – 2.5 nm → good match to vortex core ~ξ**

20 nm

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Model for J_c vs. BZO dopant fraction x

J_c: For non-interacting vortices at low temperatures, Nelson and Vinokur [PRB (1993)] have $J_c = cU_0 / \xi_{ab}\Phi_0$ with pinning energy

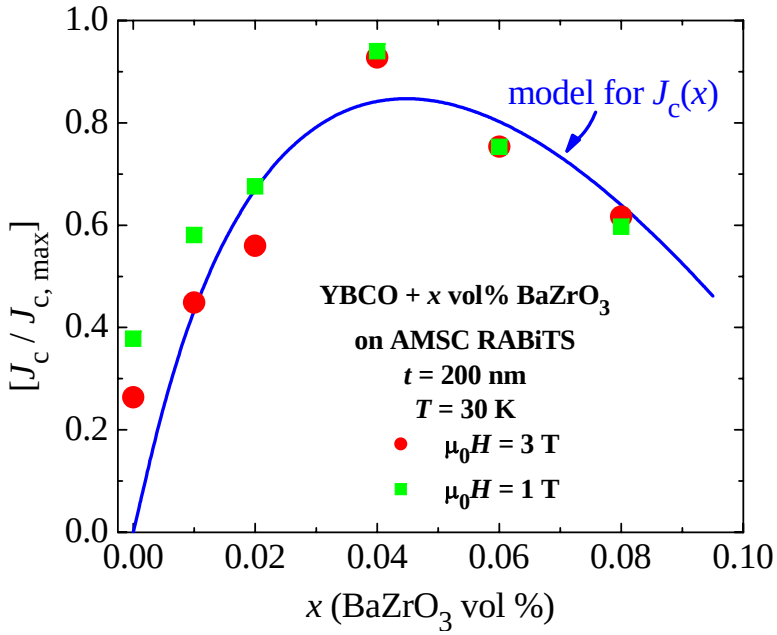
$$U_0 = \frac{1}{2}\left(\frac{\Phi_0}{4\pi\lambda_{ab}}\right)^2 \ln\left[1+\frac{c_0^2}{2\xi_{ab}^2}\right]$$

Here c_0 = effective radius of CD = $kb_0 \propto k\sqrt{x}$ where $k \approx 1$ and x = dopant concentration.

Effect of CDs on penetration depth λ: Wahl, Buzdin, et al. [Physica C (1995)] showed theoretically that adding CDs increases λ (and *reduces the line energy*) as

$$\left(\frac{1}{\lambda(B_\Phi)}\right)^2 = \left(\frac{1}{\lambda(0)}\right)^2\left[1-\frac{2\pi c_0^2 B_\Phi}{\Phi_0}\right]$$

Combining these expressions (and assuming that increasing dopant level x increases CD radius, not B_Φ) gives



$$J_c(x) \approx J_{pb} \ \ln\left[1+k^2 x\frac{B_{c2}}{B_\Phi}\right]\left(1-2k^2 x\right)$$

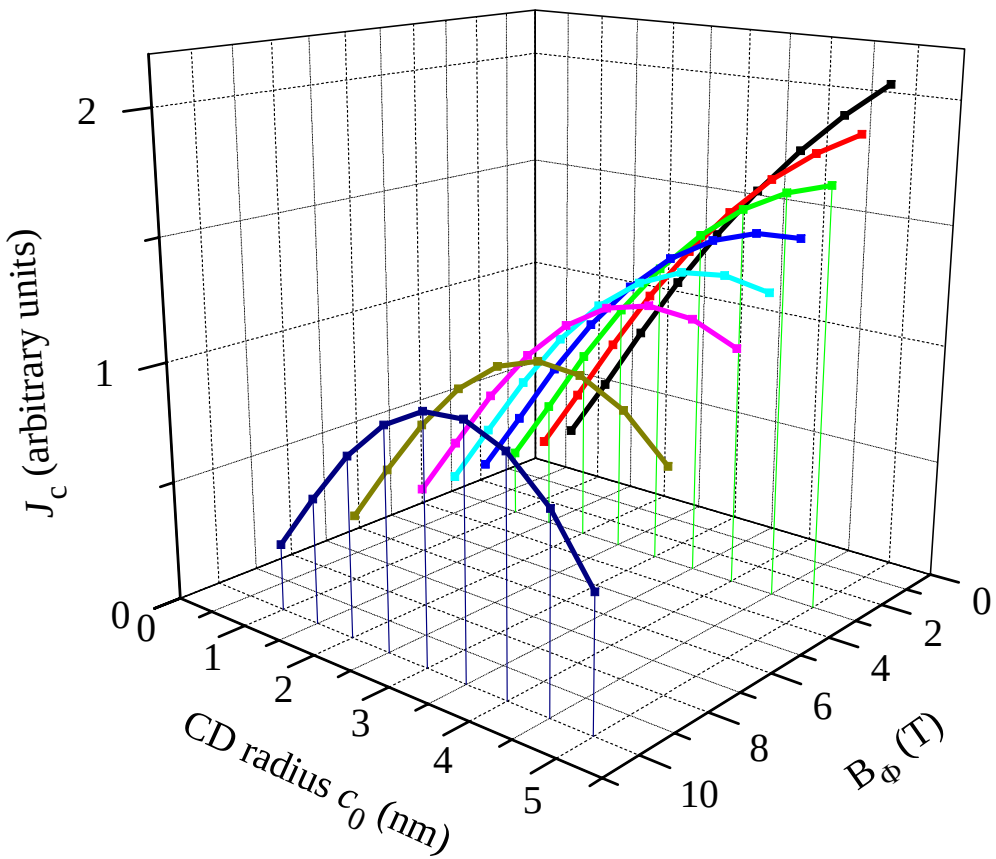
Here $k \approx 2$ => effective radius c_0 > nominal size based on *continuous* BZO stacks..

As with irradiation tracks, changes in equilibrium properties – λ – strongly affect the pinning of vortices [J.R. Thompson et al, *Phys Rev B* (2004)].

Predictive guidance for J_c, assuming:

- Constraints of model are obeyed (e.g., $H << B_\Phi$)
- Processing can control BZO nanoparticle size and column density where, $x = k\, c_o^2\, B_\Phi$

$$J_c(x) \approx J_{pb}\ \ln\left[1+\frac{c_o^2}{2\xi^2}\right]\left(1-2\pi\frac{c_o^2 B_\phi}{\phi_o}\right)$$



Goal: Conduct measurements and analysis of J_c

wide-range thickness dependence of ex situ YBCO

Objectives and Approach:

- **Deposit co-evaporated Y-rich BaF_2-YBCO on RABiTS**
- **Determine defect size and distribution**
- **Series of films with thickness range from 28nm to 1.5 μm**
- **$J_c(H,T)$ measured for $H||c$**
- **Analyze and compare with model calculations**

  [Ijaduola *et al.*, *PRB* 73, 134502 (2006)]

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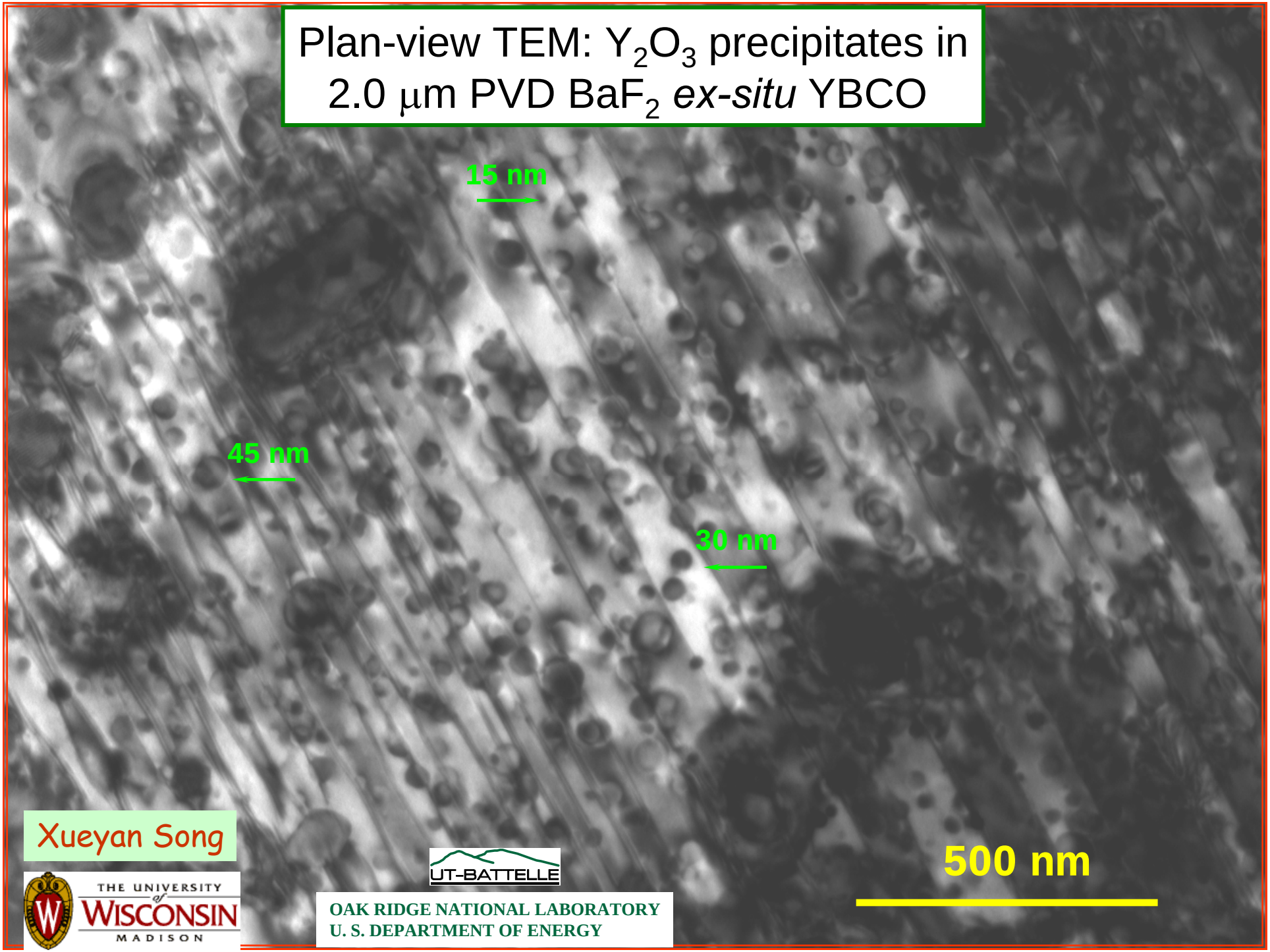
Plan-view TEM: Y_2O_3 precipitates in 2.0 μm PVD BaF_2 *ex-situ* YBCO



Xueyan Song

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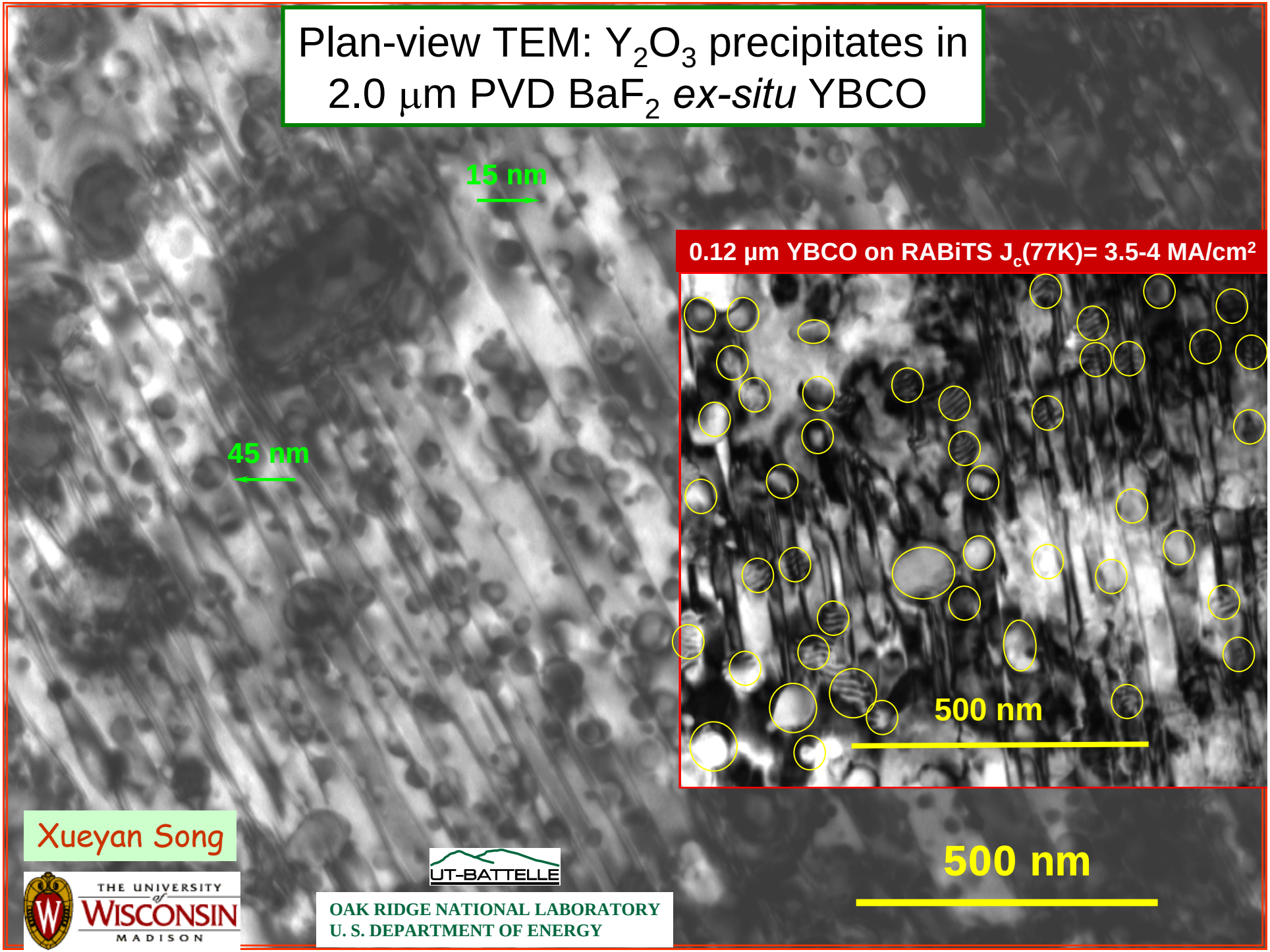

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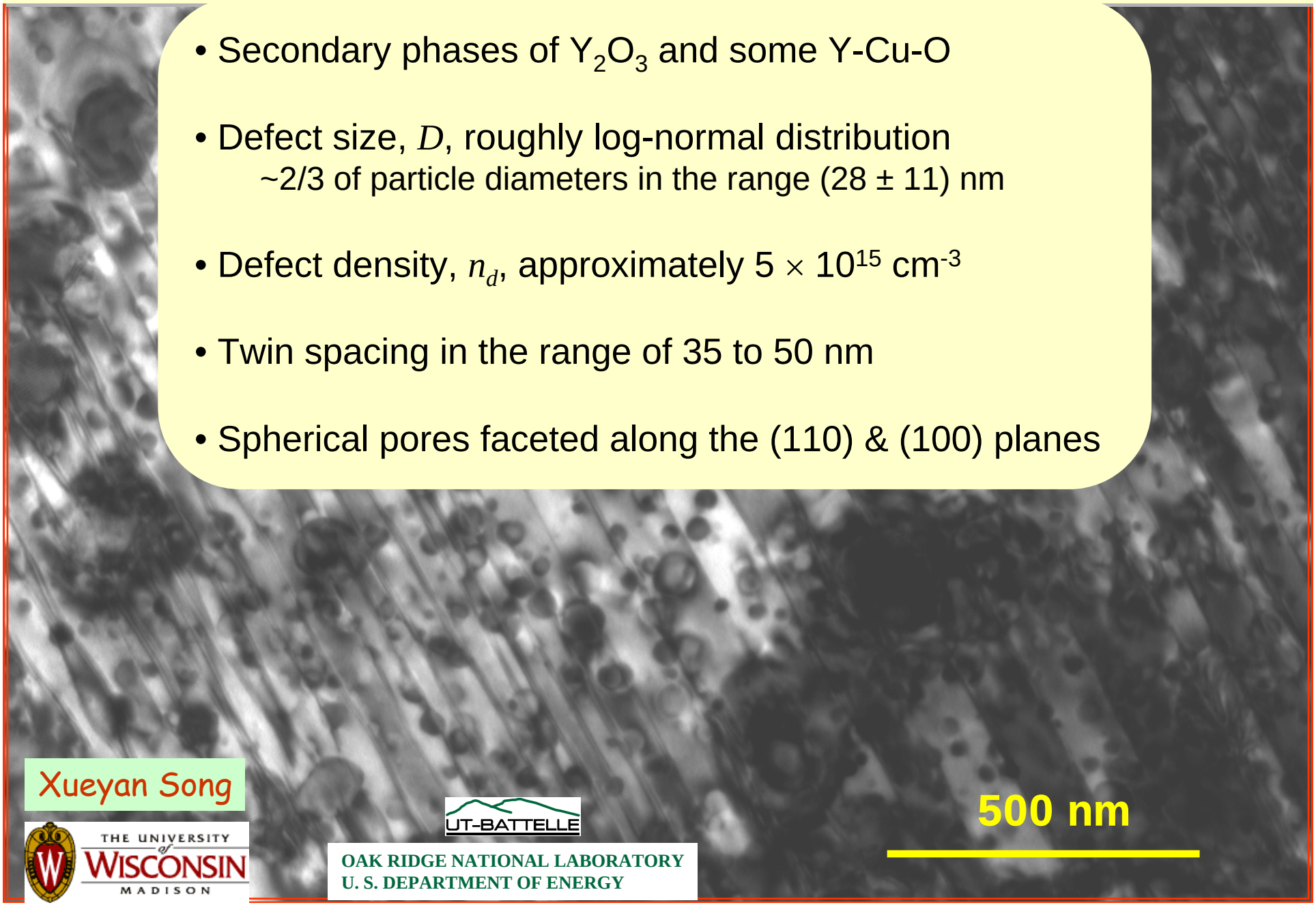
Observed nanostructure

- Secondary phases of Y_2O_3 and some Y-Cu-O
- Defect size, D, roughly log-normal distribution
  ~2/3 of particle diameters in the range (28 ± 11) nm
- Defect density, n_d, approximately 5×10^{15} cm^{-3}
- Twin spacing in the range of 35 to 50 nm
- Spherical pores faceted along the (110) & (100) planes



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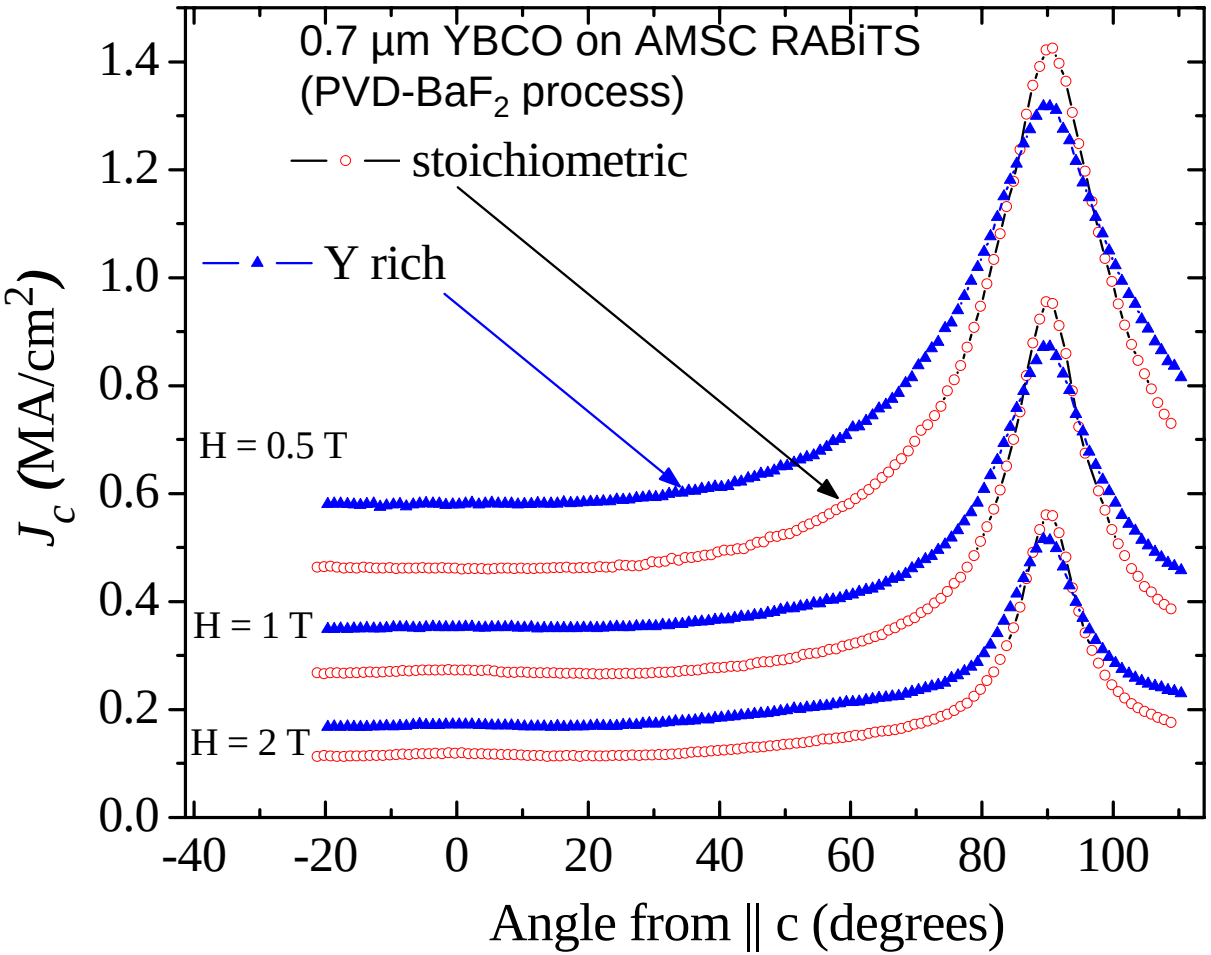
No correlated pinning near *c*-axis in *ex situ* films – "flat" J_c vs. angle

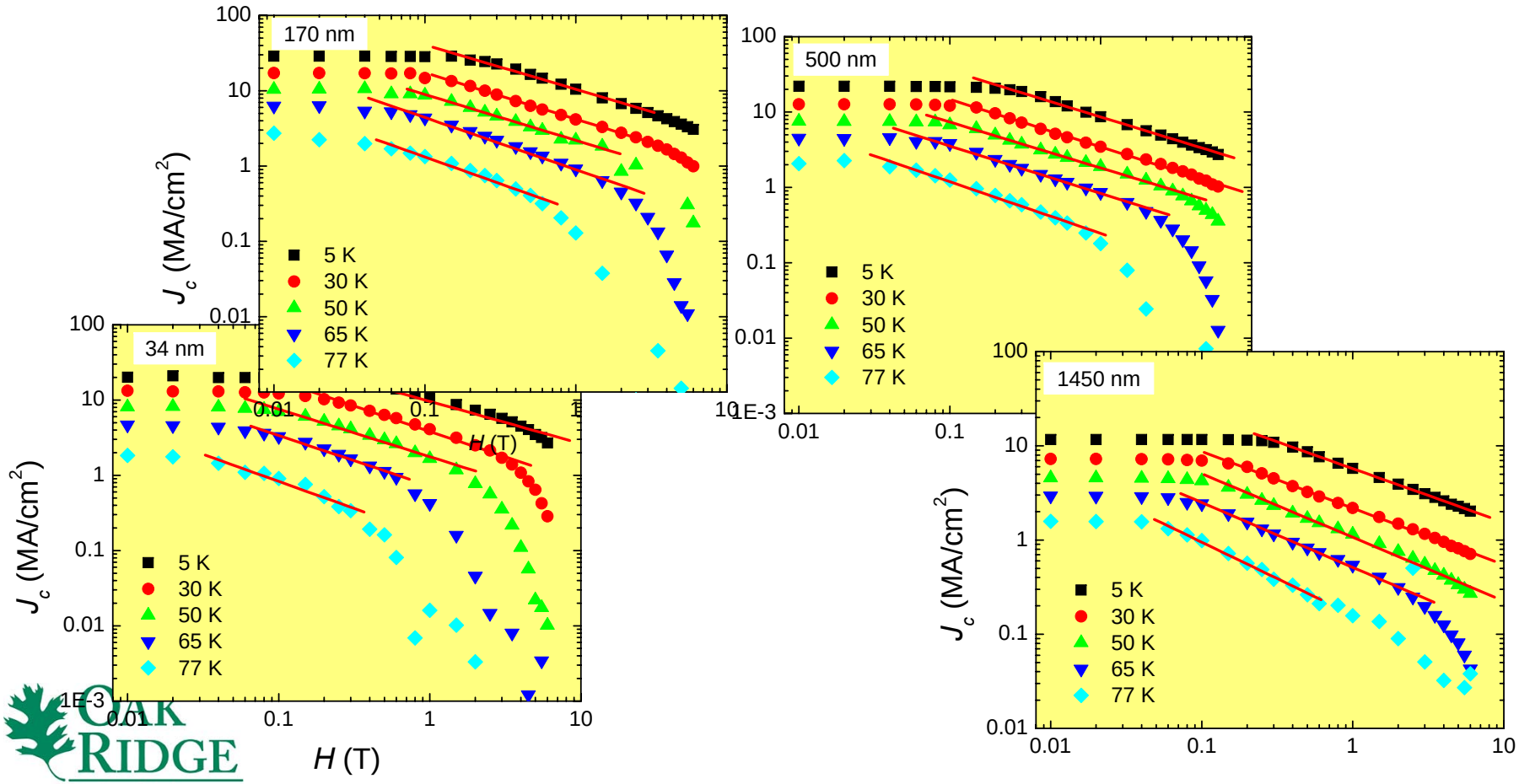


For magnetic field *H* near the *c*-axis, no angularly selective pinning is discernable; this rules out correlated disorder along the *c*-axis as dominant pinning mechanism for this orientation.

(However, for *H* near the *ab* planes, a peak commonly attributed to "intrinsic pinning" or planar defects is evident.)

$J_c(H,T)$ for *ex situ* YBCO: analysis and interpretation

- Eleven samples with thicknesses from 28 nm to 1.5 μm [Feenstra]
- Temperature and field dependencies for $H||c$ [Ijaduola, Thompson, *et al.*]



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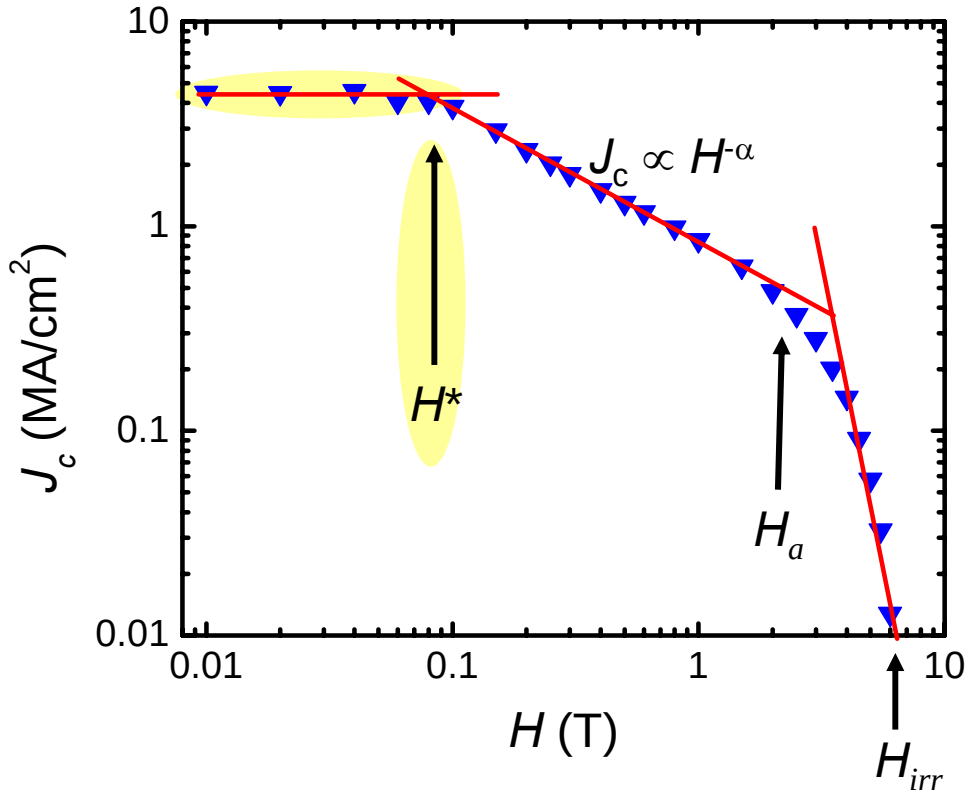
General Characteristics for Analysis

Flux pinning theory of Ovchinnikov and Ivlev [*PRB**43***]

Re-expressed and interpreted by van der Beek *et al.* [PRB**66**]

- Nearly field independent low-field J_c ($H<H^*$)
- power law dependence $J_c \propto H^{-\alpha}$ ($H^*<H<H_a$)
- Rapid decay to $J_c \rightarrow 0$ at $H_{irr}<H_{c2}$



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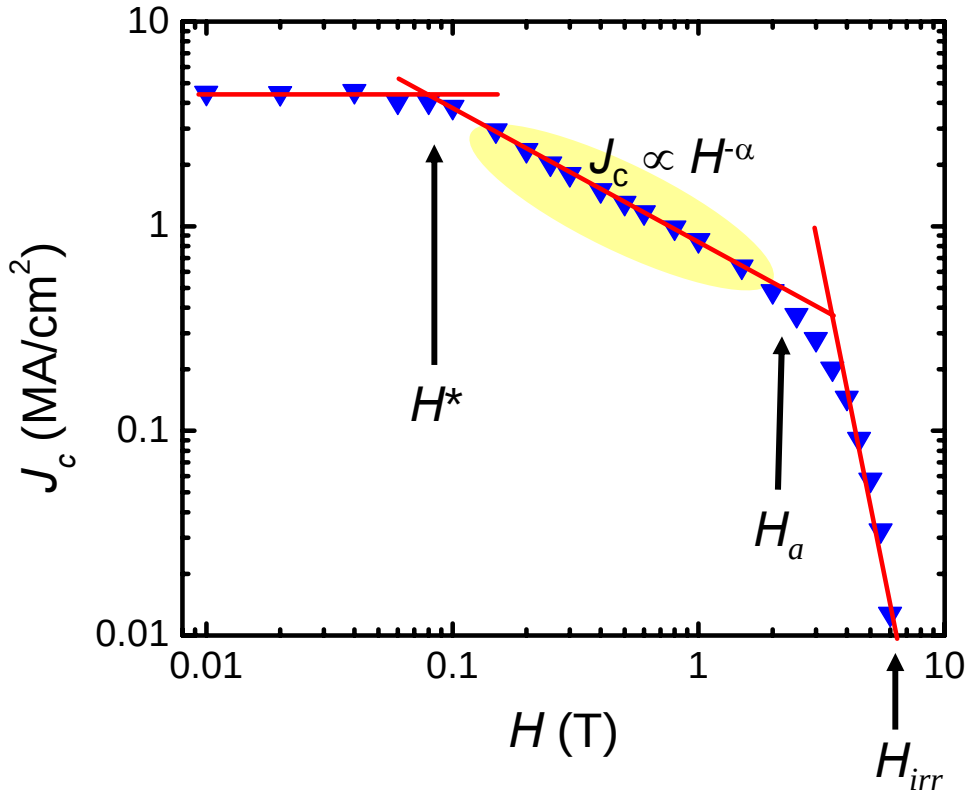

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Interpretation of data

Flux pinning theory of Ovchinnikov and Ivlev [*PRB**43***]

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For large, sparse normal-state defects,

- $J_c \propto [1-(T/T_c)^2]^m$, consistent with m=7/6 for single-vortex pinning by normal precipitates ("δT_c" pinning) [Griessen *et al., PRL**72***]

- $H^* \propto n_d \ln[1 + D^2 / 2\xi^2(T)]$

  Temperature dependence described for T >30 K

- $J_c(H) \propto n_d\ D^{9/4} H^{-5/8}; \quad H^* < H < H_a$

  ✓ Observed power-law α in range 0.56 – 0.69

  For all sample thicknesses and temperatures

  ✗ but, magnitude of J_c too small by factor ~ 20

- Thickness dependence: $J_c / d \propto n_i D U_p$

  Crossover to thickness independent: $d^* \propto (U_p\, n_d)^{-1/2}$

Temperature dependence of low-field J_c

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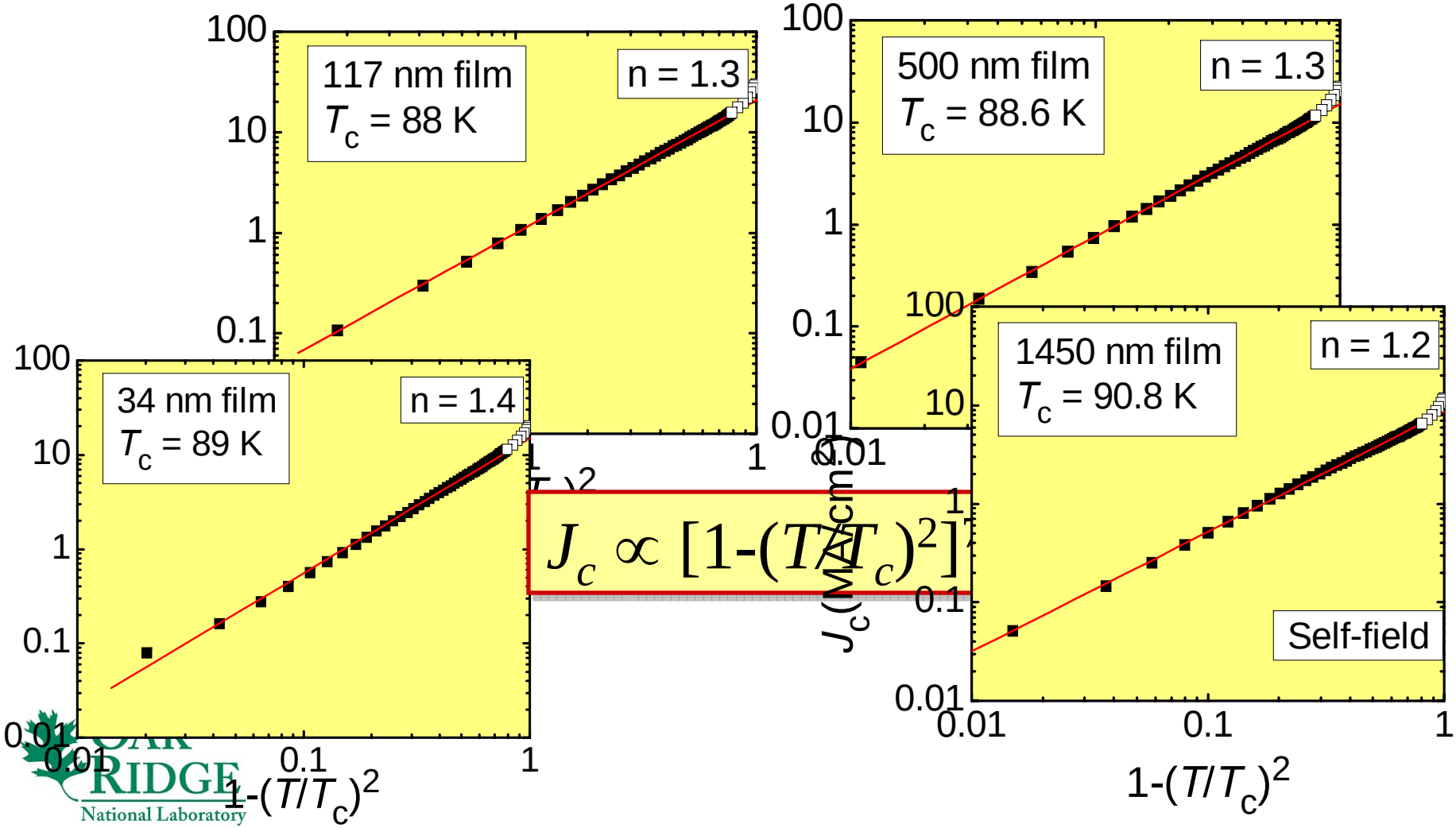

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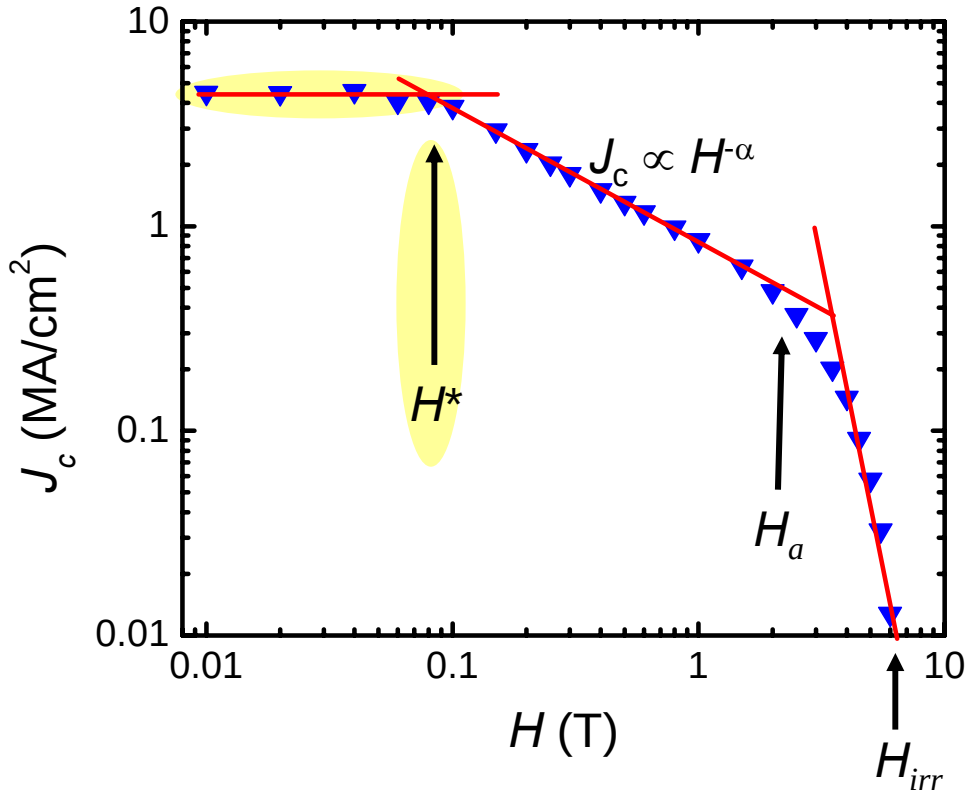

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  rossover to thickness independent: $d^* \propto (U_p\, n_d)^{-1/2}$

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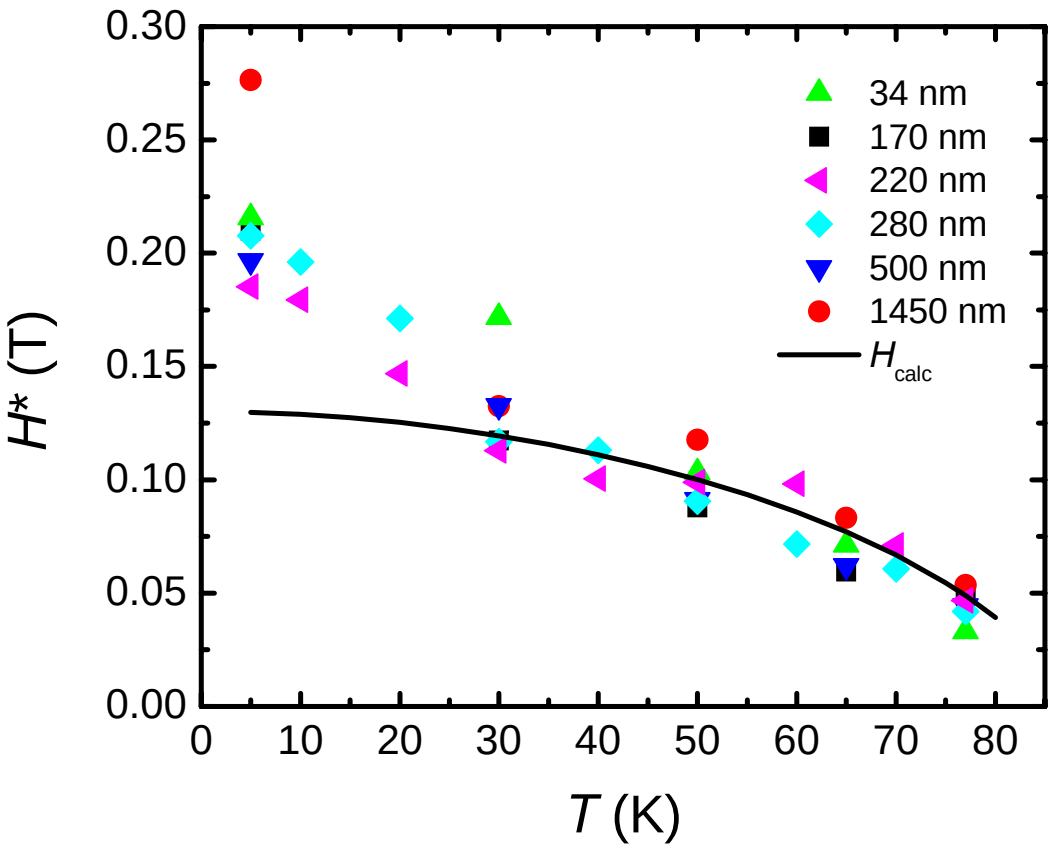
Temperature dependence of crossover field H^*

- H^* not sensitive to film thickness
- Approximate agreement with TEM-determined n_d and ~D
- Consistent with J_c magnitude
- Low-T upturn due to weak collective pinning?



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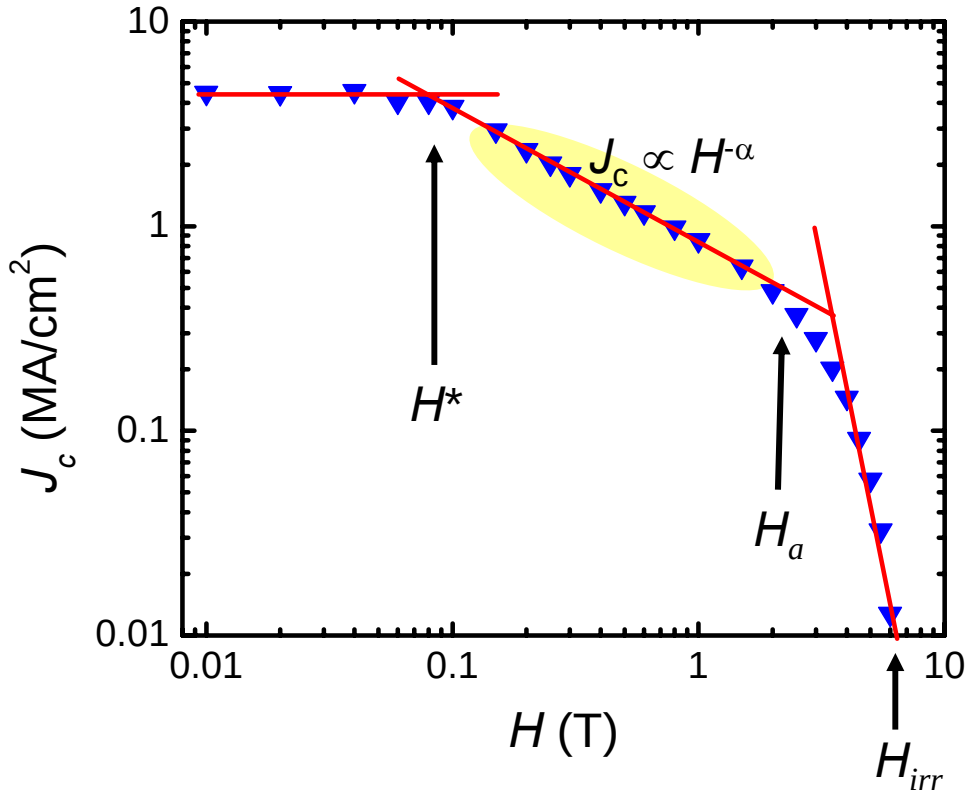

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  ✓ Observed power-law α in range 0.56 – 0.69

  For all sample thicknesses and temperatures

  ✗ but, magnitude of J_c too small by factor ~ 3 - 20

- Thickness dependence: $J_c / d \propto n_i D U_p$

  rossover to thickness independent: $d^* \propto (U_p n_d)^{-1/2}$

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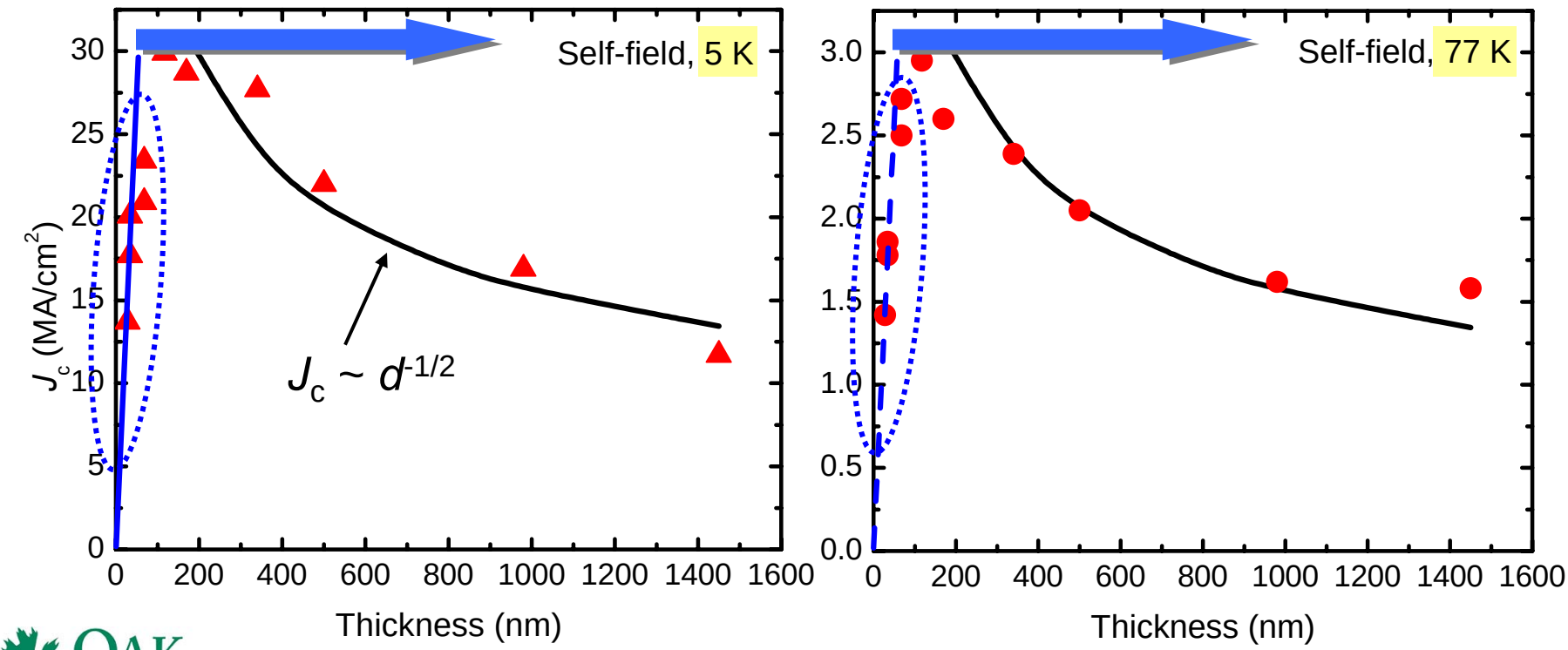
- Thickness dependence: $J_c \propto (n_i DU_p)\, d; \quad d < d^*$

  Crossover to thickness independent: $d^* \propto (U_p\, n_d)^{-1/2}$

Dependence of J_c on thickness

- J_c peaks at thickness d ~ 120 – 140 nm
- $J_c(d)$ is nearly temperature and field independent

  Decrease at small d not thermal effect?
- Observed low-T $J_c/d \approx 5 \times 10^8$ A/cm^2/μm-thickness
- Decay at large d not predicted; calculated d^*~40 nm



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Goal: Continue optimizing substrate surface modification for flux pinning enhancements

Background and Motivation:

- Pre-treatment of substrate surfaces with nanoscale particles can lead to improved flux pinning in YBCO

- For extension to coated conductors, optimize:
  - ➥ Different species
  - ➥ Decoration techniques
  - ➥ Properties improvements

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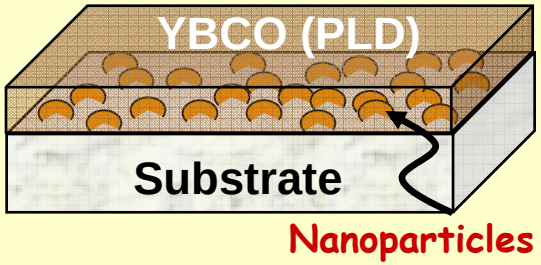
Substrate pre-treatment with nanoparticles

YBCO thickness = 0.2-0.3 μm



Approach:

Modify single crystal $SrTiO_3$ & biaxially textured tape templates by applying second-phase nanoparticles using:

- PVD-based (sputtering) → Iridium
- Solution-based (MOD) → MgO & $BaZrO_3$
- Suspension-based → CeO_2, $BaTiO_3$, $BaZrO_3$ (pre-formed)

Advantages:

- Applicable to any substrate template
- Simple technique for induced growth defects in HTS
- Easy control of number density & size by adjusting the process parameters

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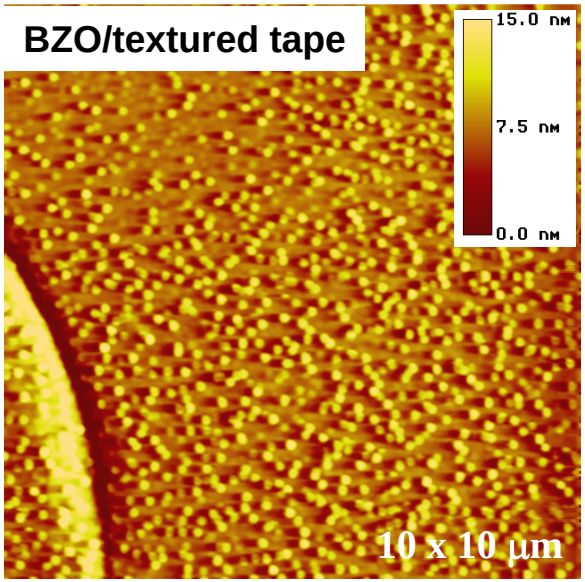
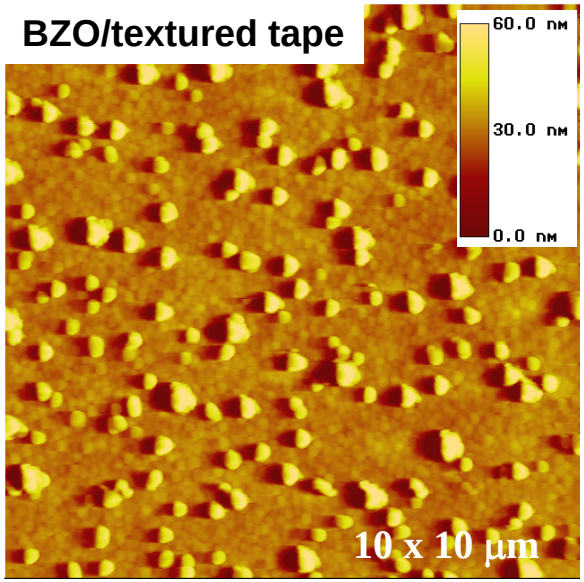
Nanostructured templates: Interfacial engineering by substrate surface decoration

Sputter deposited



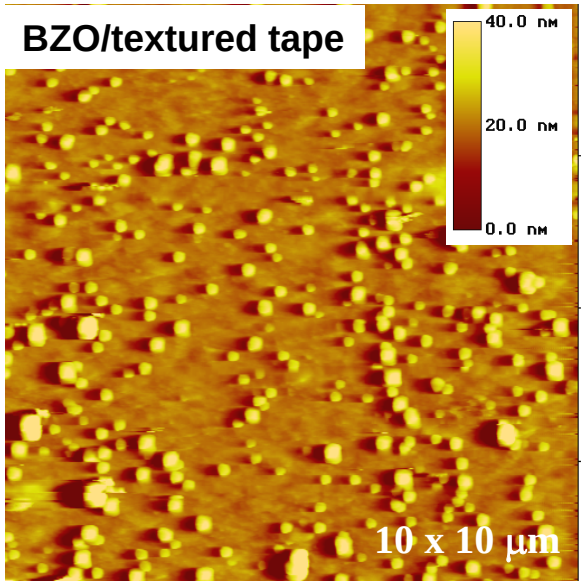
Size= 200 nm
Density, n = 10 μm^{-2}
$B_\phi = n\phi_o$ = 0.2 Tesla

Solution (MOD) processed



Size= 250-500 nm
Density, n = 2 μm^{-2}
$B_\phi = n\phi_o$ = 0.004 Tesla

Suspension deposited Pre-formed



Size= 250 nm
Density, n = 3.2 μm^{-2}
$B_\phi = n\phi_o$ = 0.007 Tesla

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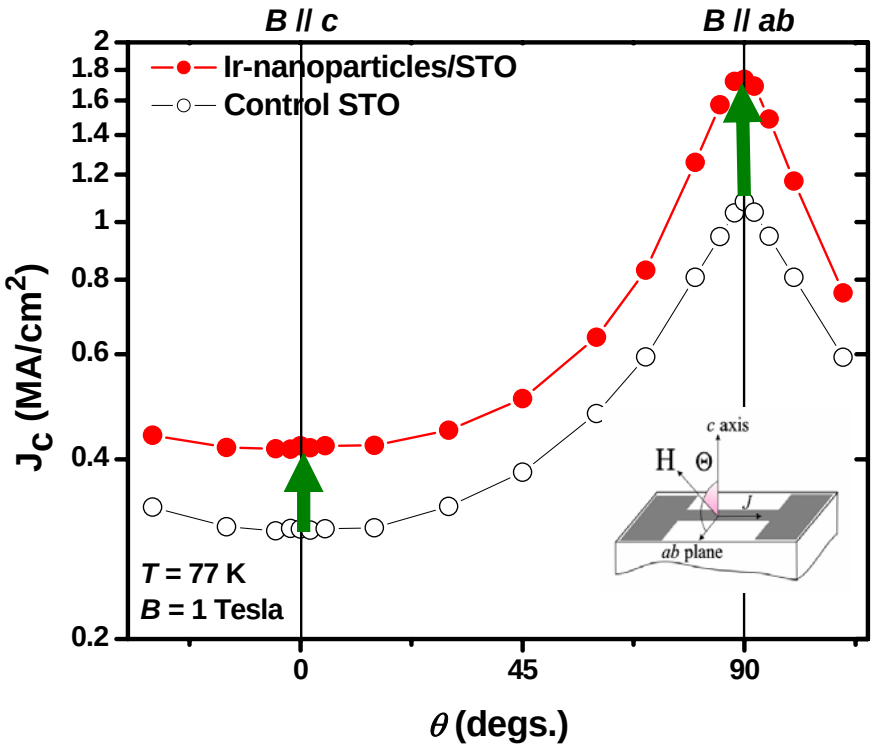
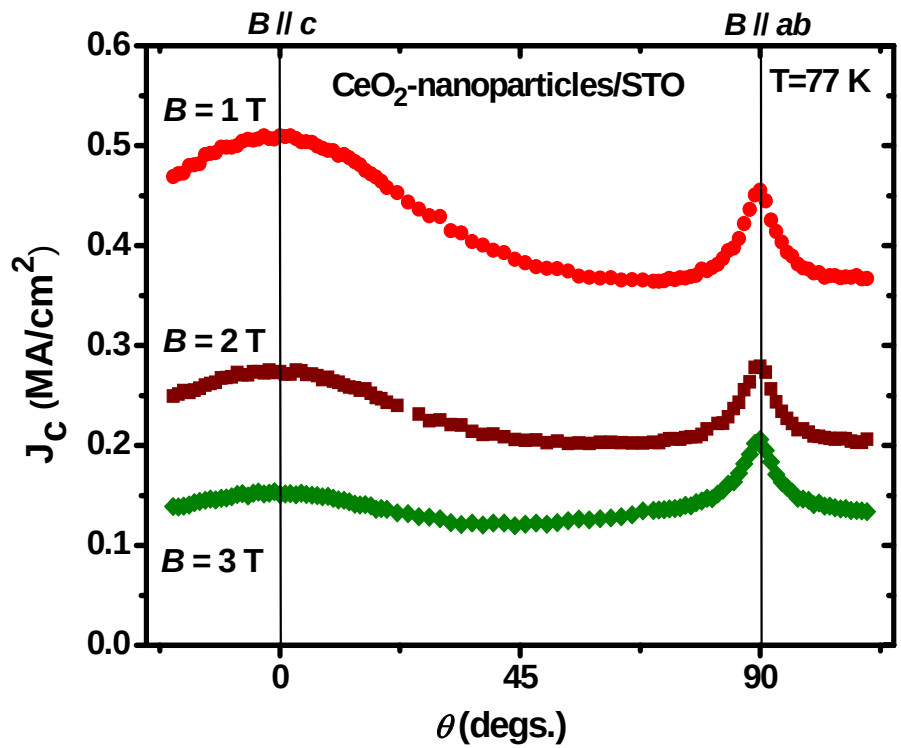
Angular dependence of J_c: (FY 2005 result) With/without nanoparticle modifications

Single crystal substrates

Sputter deposited Ir nanoparticles



Suspension deposited preformed CeO_2 nanoparticles



For Ir: Uncorrelated, random isotropic defects acting over large angular regime
For CeO_2: *c*-axis correlated pinning by extended defects, smaller $\alpha \sim 0.3$

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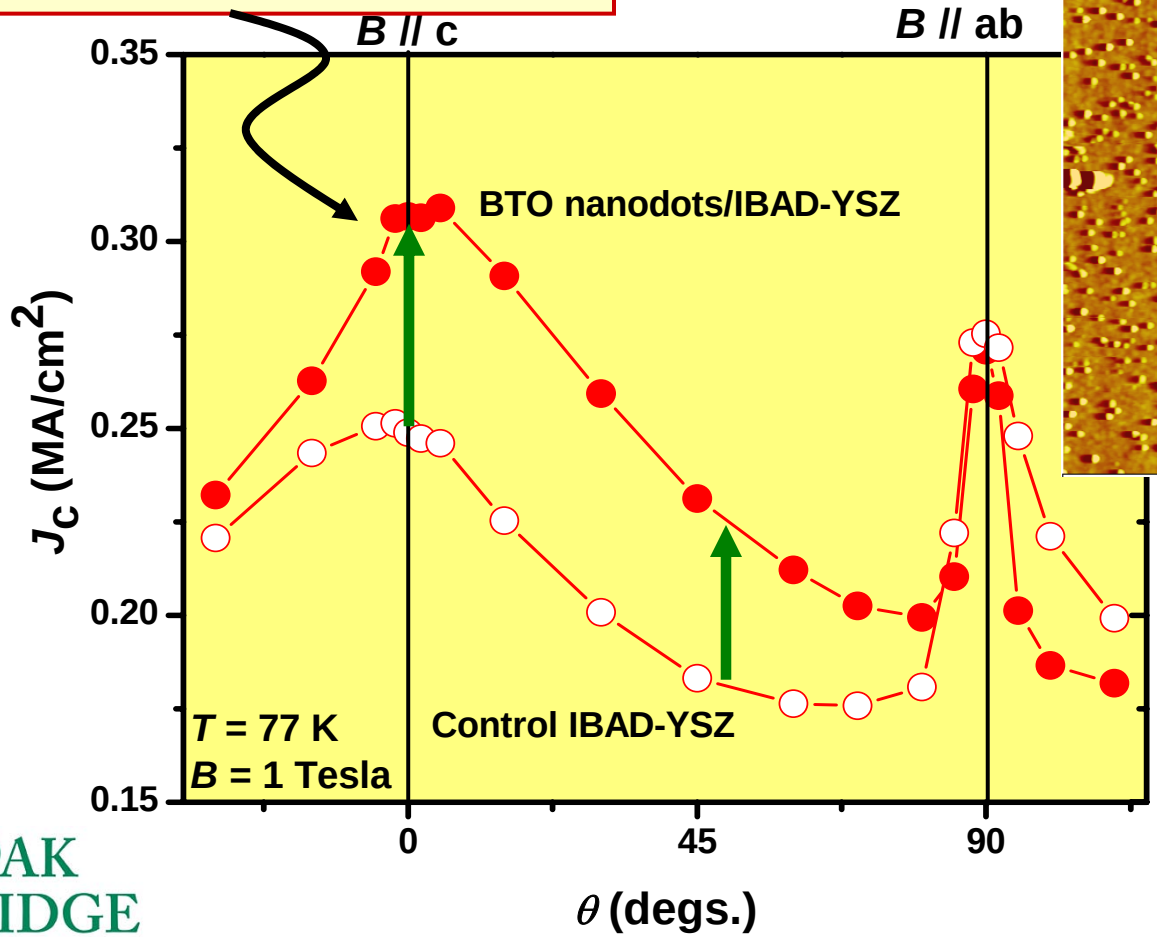
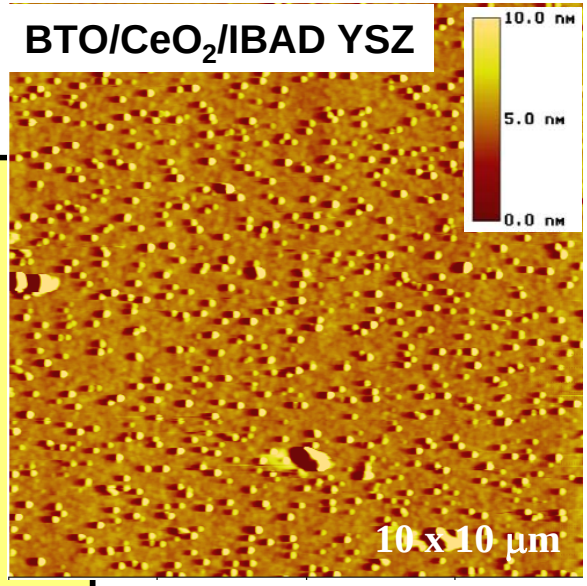
Excellent properties obtained using pre-formed nanoparticles

$BaTiO_3$ nanoparticles on CeO_2/IBAD-YSZ template

Standard PLD YBCO on LMO/IBAD(MgO or YSZ) shows both *c*-axis & *ab*-plane peak
Improved in-field J_c performance is related with broad peak about *c*-axis

$BaTiO_3$ nanoparticles induces additional *c*-axis correlated defects





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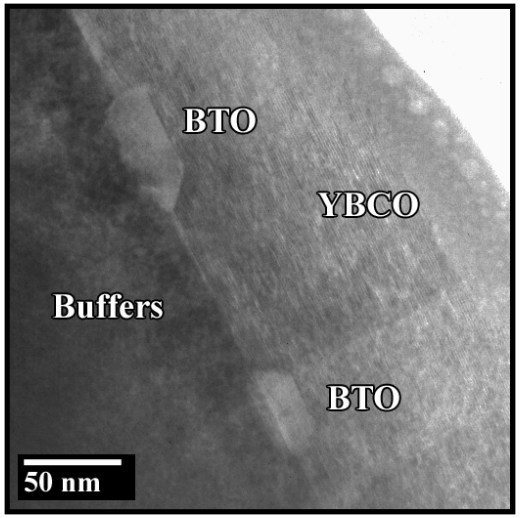
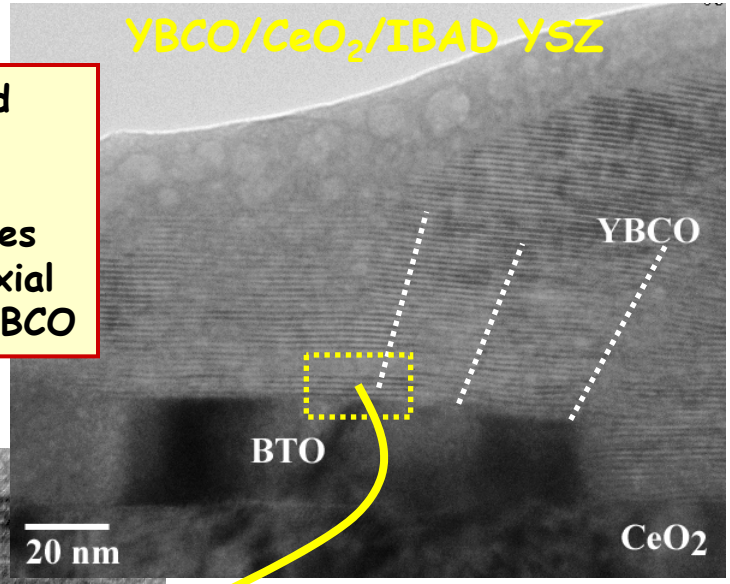
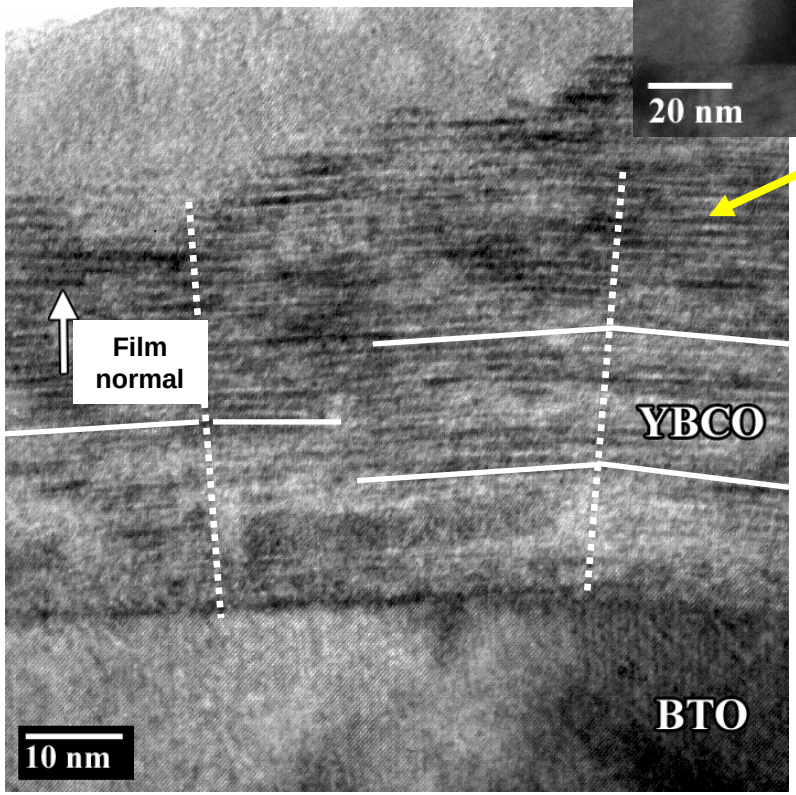
SuperPower Inc.

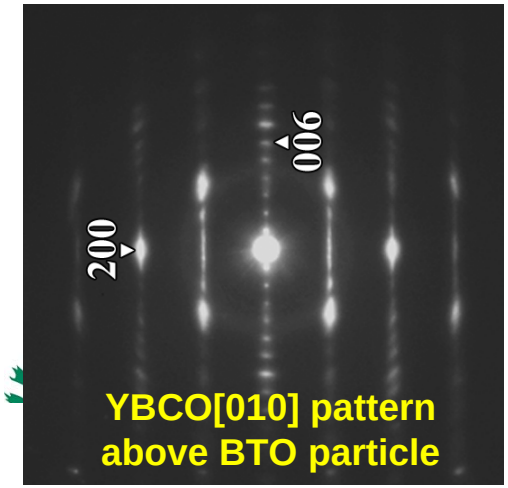
Surface BTO nanoparticles produce distortion of YBCO planes leading to subgrain boundaries



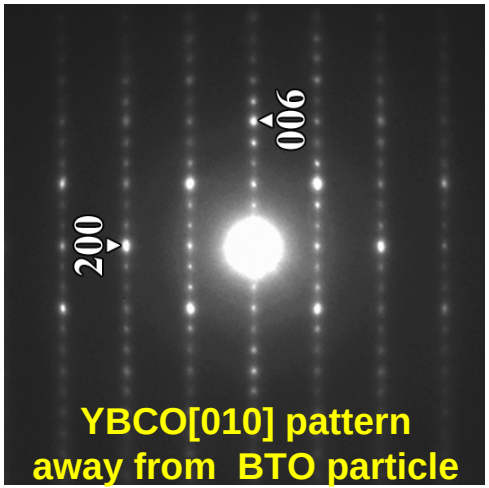
- TEM analysis shows extended defects along c-axis
  - ➥ Distortion of YBCO planes
  - ➥ Low-angle subgrain boundaries
  - ➥ BTO particles are not epitaxial
  - ➥ No reaction between BTO/YBCO





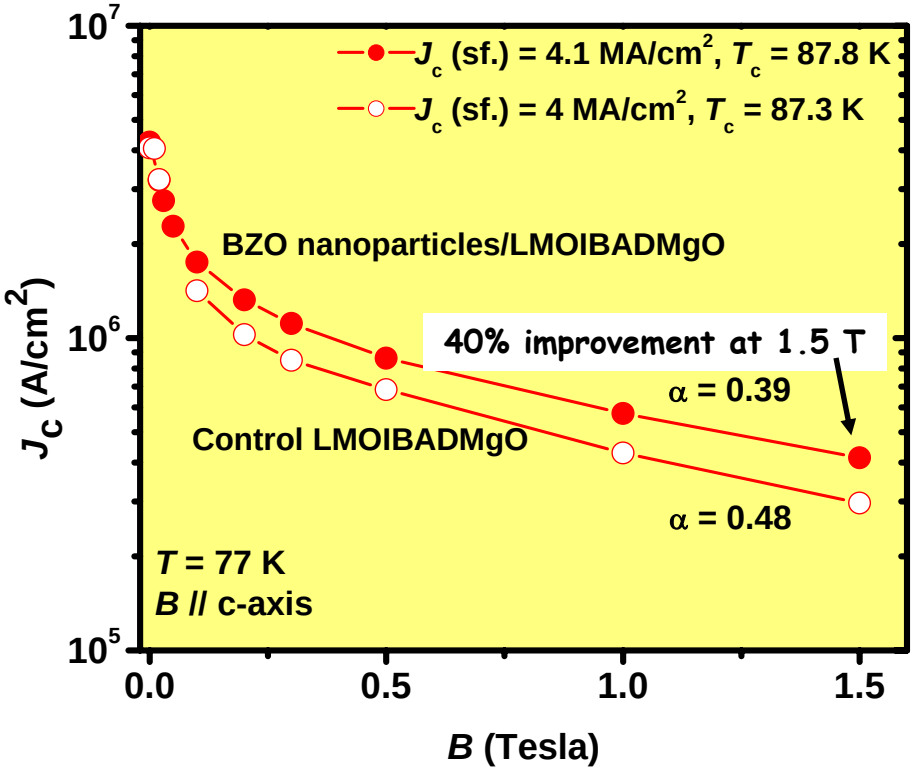


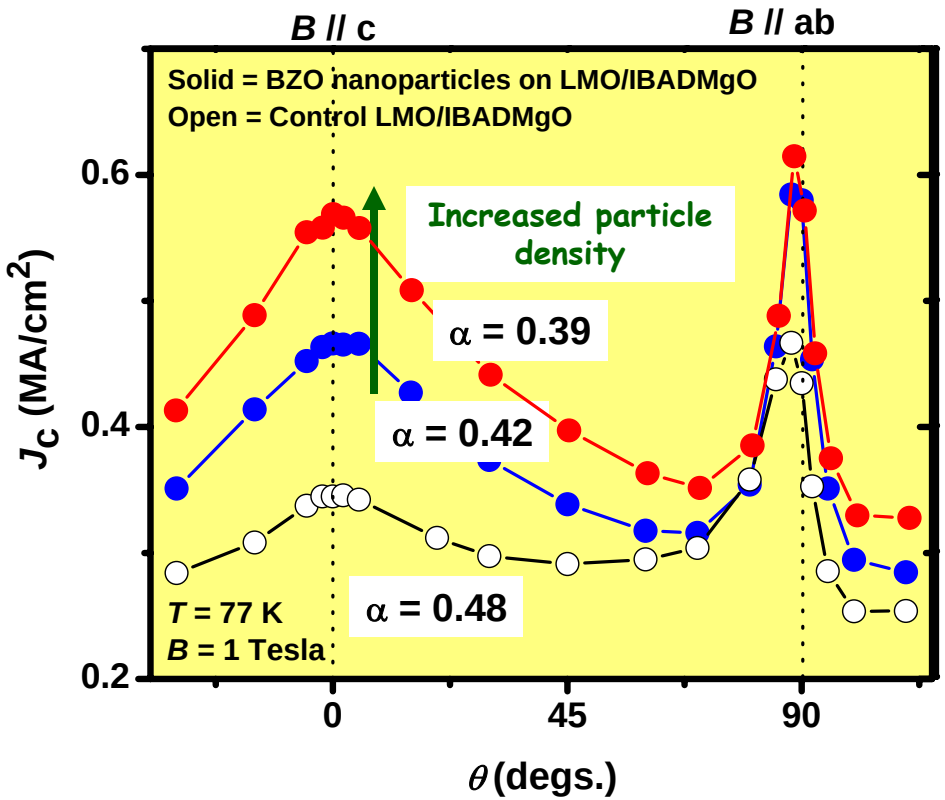
YBCO[010] pattern above BTO particle



YBCO[010] pattern away from BTO particle

Similar in-field performance is obtained by using pre-formed $BaZrO_3$ nanoparticles on SuperPower LMO/IBAD-MgO





- Improved J_c-B performance, smaller α ~ 0.39
- Strong pinning by defects distributed over large angular range

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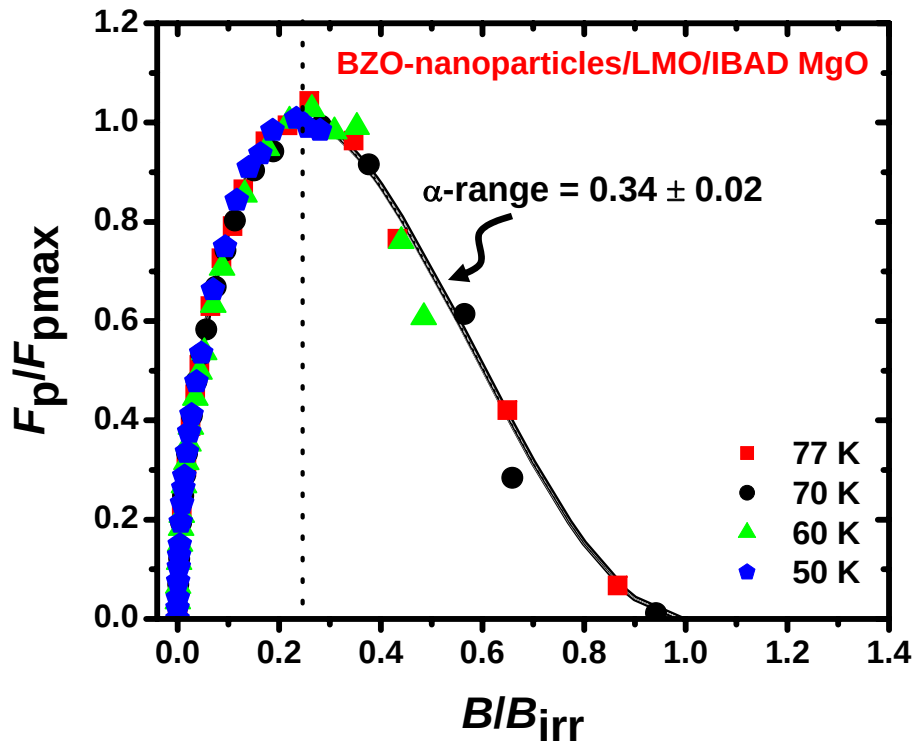
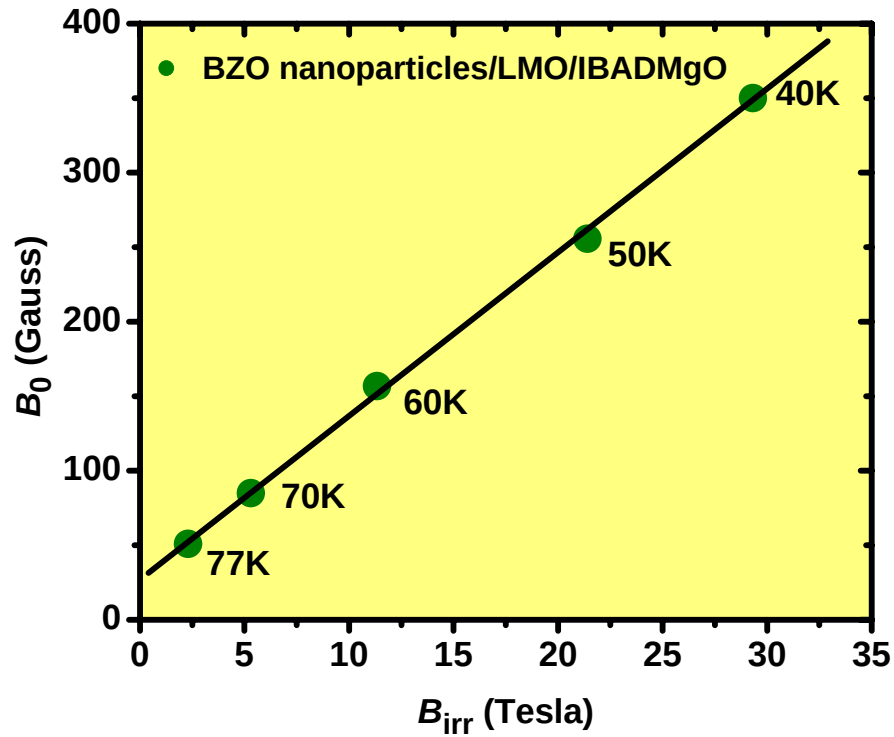
Scaling behavior of pinning force density F_p; F_p shows consistency of pinning mechanism wrt temperature

Scaling provides a powerful tool for predicting J_c at arbitrary B, T





$$F_p = \frac{J_c(0)B}{(1+B/B_o)^{\alpha}}(1-B/B_{irr})^2$$

3-parameters: α, B_o, B_{irr}

- Model fits data very well
- α is ~ independent of T
- F_p peaks at $B/B_{irr} \cong 0.25$
- $B_o(T)$ scales with $B_{irr}(T)$

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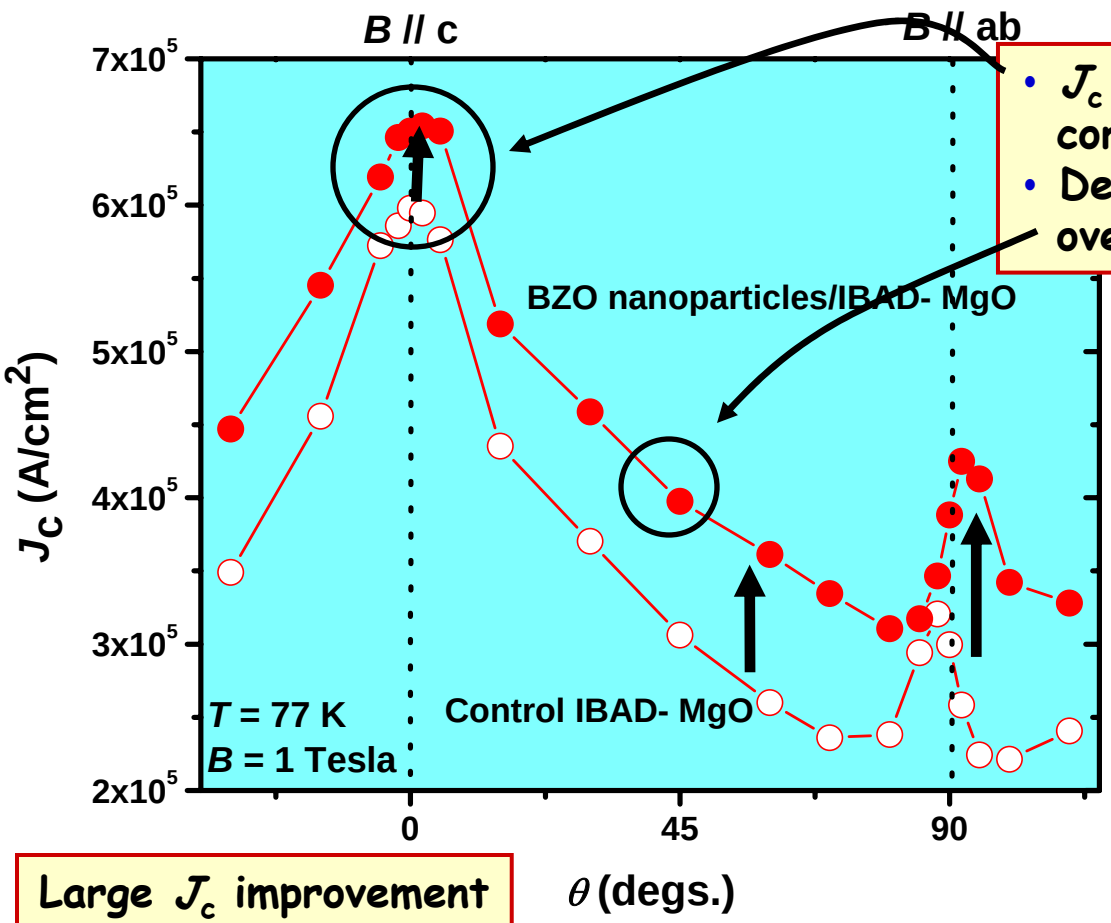
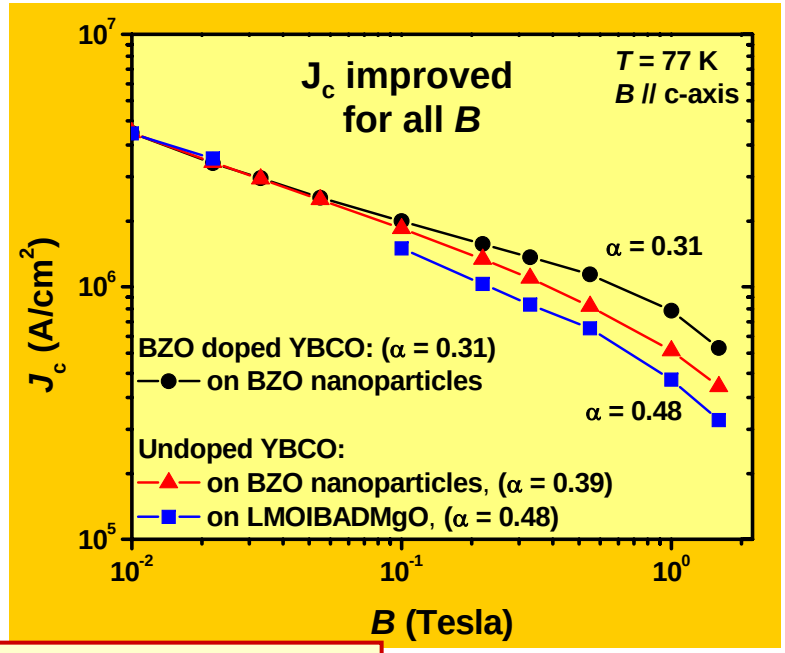
Combine substrate surface modifications + compositional BZO nanoparticle additions

BZO nanoparticles on *BZO doped PLD YBCO* on LMO/IBAD-MgO



- J_c is dominated by *c*-axis correlated defects
- Defects are operative over wide angular range



Large J_c improvement but, similar α = 0.31

Similar self-field J_c but, smaller α

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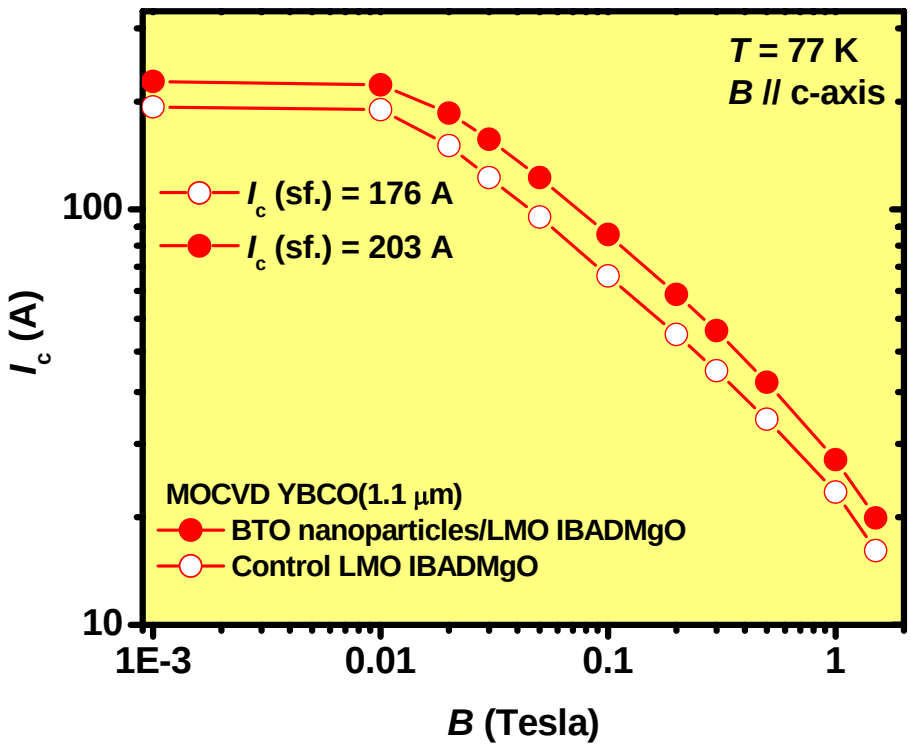
Nanoparticle surface modifications extended to thicker SuperPower MOCVD YBCO (1.1 μm) films: Transport J_c

BTO & BZO nanoparticles on LMO/IBAD-MgO



Surface nanoparticles produce improved J_c over control LMO/IBAD-MgO

SAMPLE ID:	End-to-end I_c (A):
Control LMOIBADMgO	168
TBTOLMOIBADMgO-5	213
TBTOLMOIBADMgO-6	203
TBTOLMOIBADMgO-7	187
TBZOLMOIBADMgO-3	210
TBZOLMOIBADMgO-4	183

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Presentation Outline

- **FY 2006 Results**

  Flux pinning in YBCO & REBCO films with columns of insulating, self-assembled nanodots **(Goyal)**

  Systematics of BZO doped YBCO **(Christen)**
  J_c analysis for Y_2O_3 doped YBCO ex situ vs thickness
  Nanoparticle substrate decoration for J_c enhancements

  Epitaxial growth of buffer layers **(Parans)**

- **FY 2006 Performance and FY 2007 Plans** **(Parans)**
- **Research Integration**

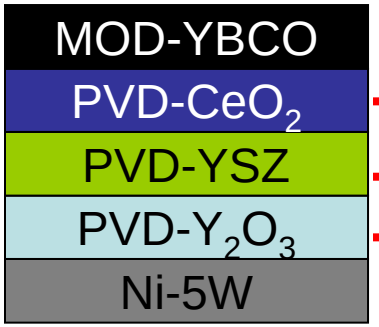
Epitaxial Growth of Buffer Layers

FY06 Objective: Fundamental studies of epitaxial growth of multifunctional buffers on textured substrates.

- **Goal: To demonstrate an I_c comparable to that of standard PVD buffers**

- **MOD CeO_2 cap development**
  Need to reduce the formation of $BaCeO_3$ with MOD-YBCO to match the performance of all-PVD buffers

- **Is $La_2Zr_2O_7$ (LZO) the best MOD barrier layer to scale up?**
  I_c of 266 A/cm achieved on slot-die coated LZO



Compatible with MOD-YBCO

Excellent barrier for Ni

Seed; W barrier; improves out-of-plane texture

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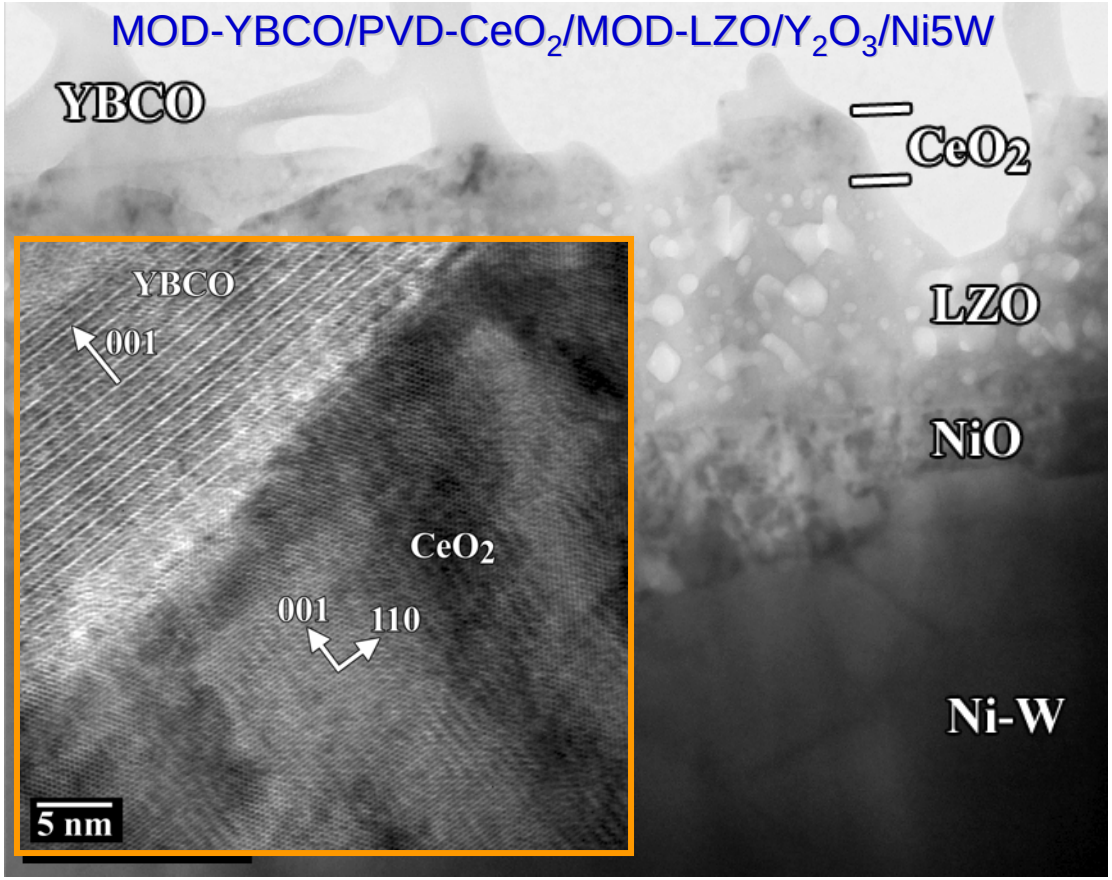
What is limiting the performance of MOD CeO_2 cap layers?

MOD-YBCO
PVD-CeO_2
MOD-LZO
PVD-Y_2O_3
Ni-5W

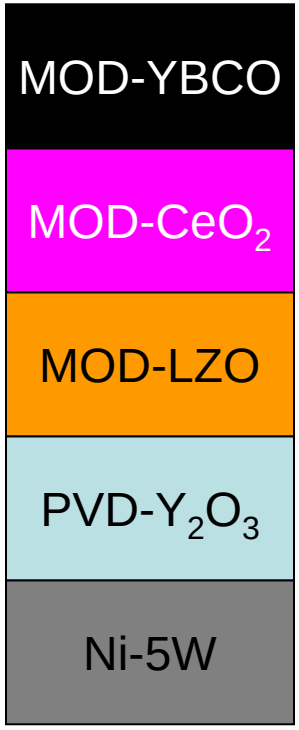
FY05: 255 A
FY06: 266 A

MOD-YBCO
MOD-CeO_2
MOD-LZO
PVD-Y_2O_3
Ni-5W

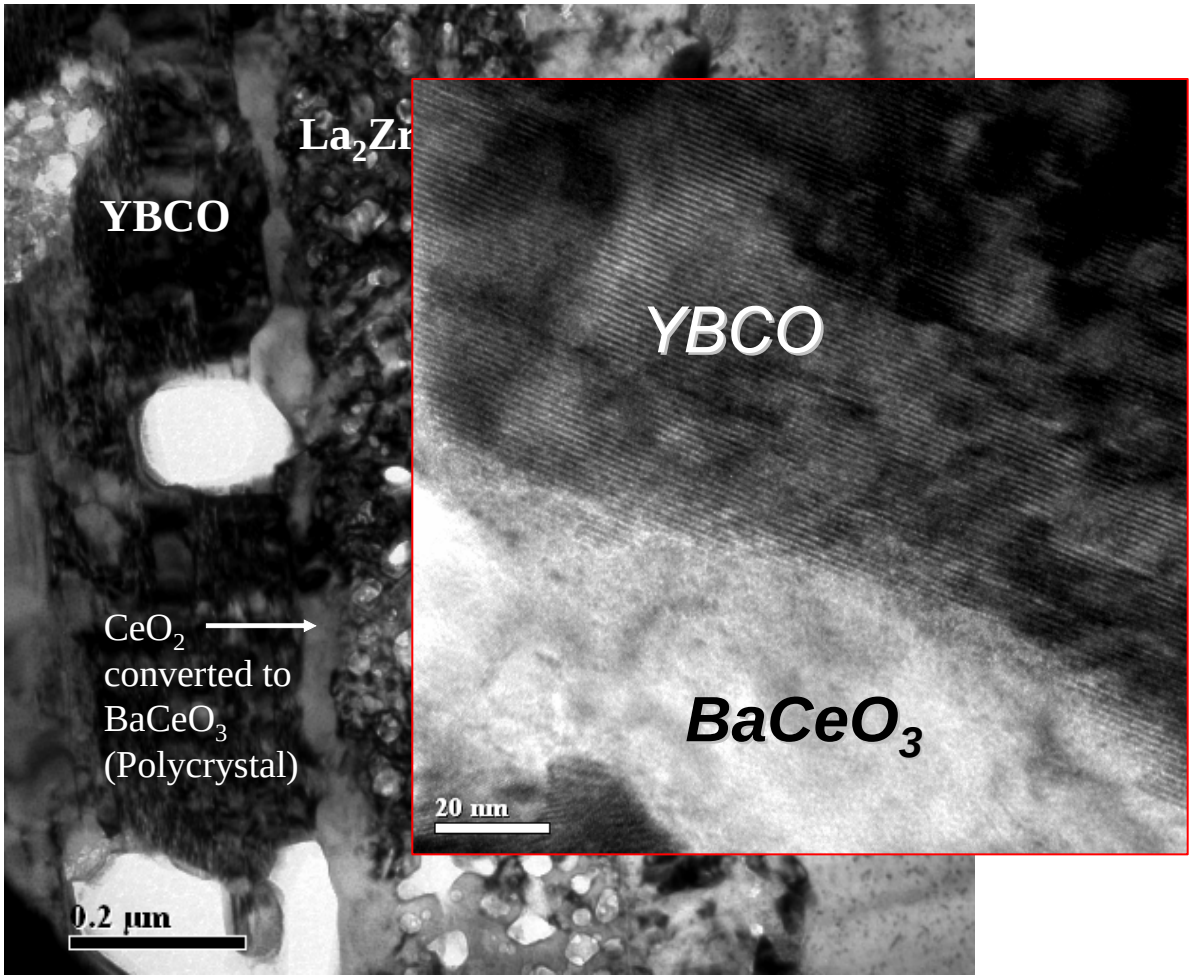
FY05: 200 A



MOD CeO_2 cap layers on MOD LZO react completely to form $BaCeO_3$

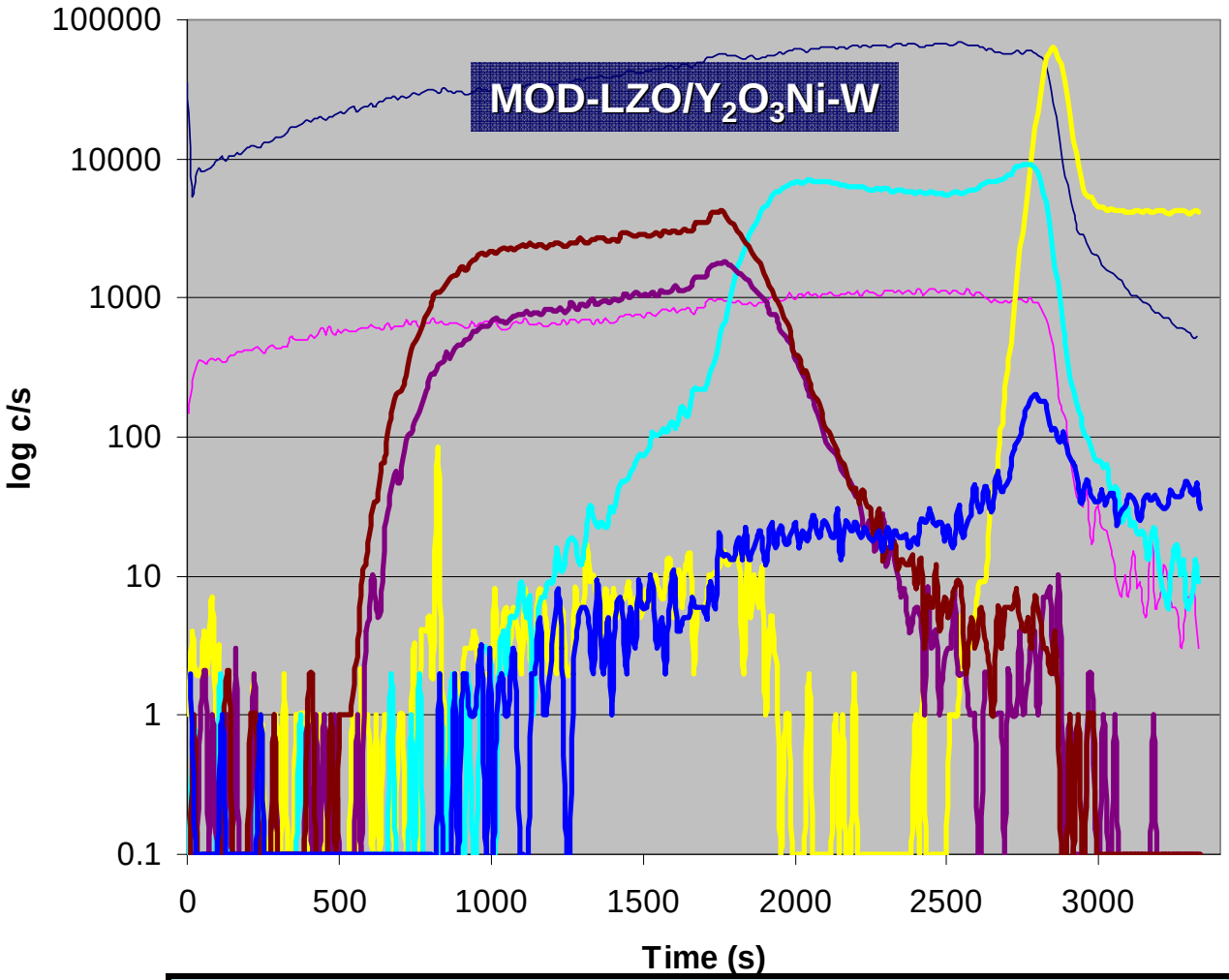


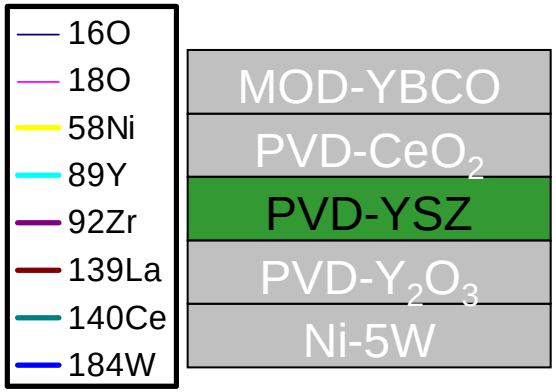
FY05: 200 A



Optimization of MOD CeO_2 cap layer is in progress

Is MOD LZO the best barrier layer for scale-up ?





SIMS depth profiling shows that 75 nm LZO is necessary to be an excellent Ni and W diffusion barrier

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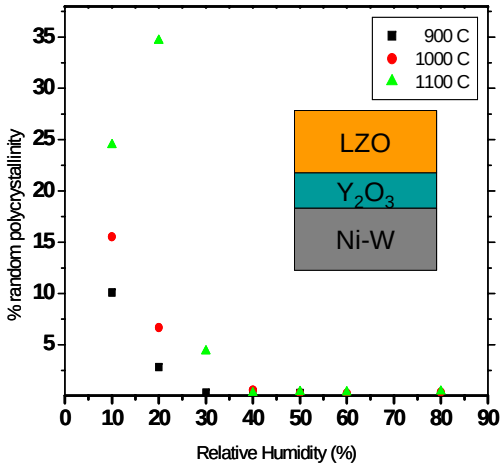
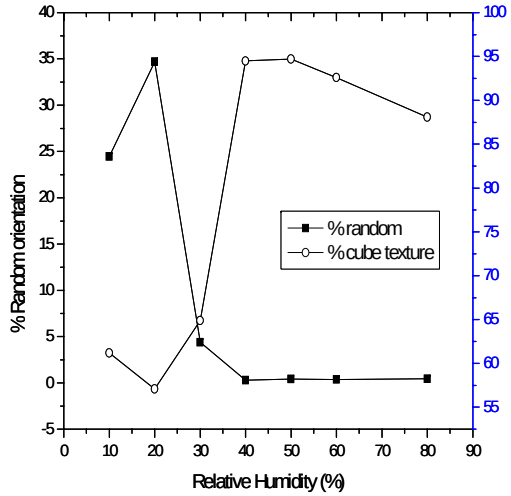
Controlling RH during coating has resulted in processing MOD LZO at wider temperature ranges

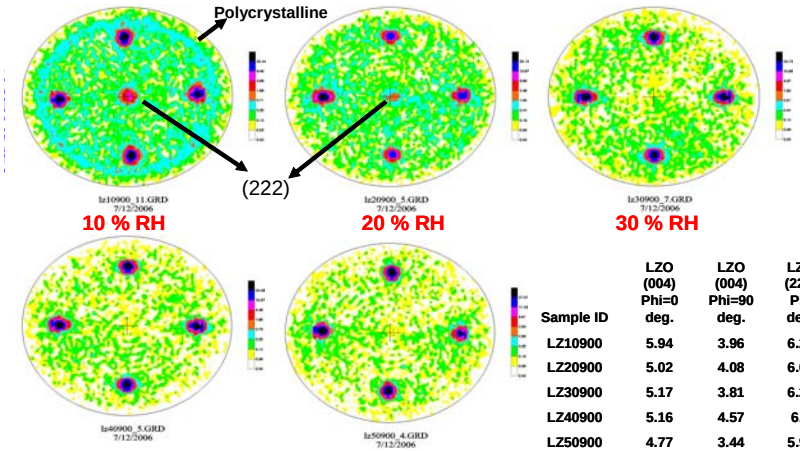




MOD LZO films with good texture was obtained under RH range: 40-60% (during coating) at temps. 900-1100 °C

Both crystallinity and % cube texture improves with higher % RH during coating

(222) LZO Pole figures for films processed at 900 °C

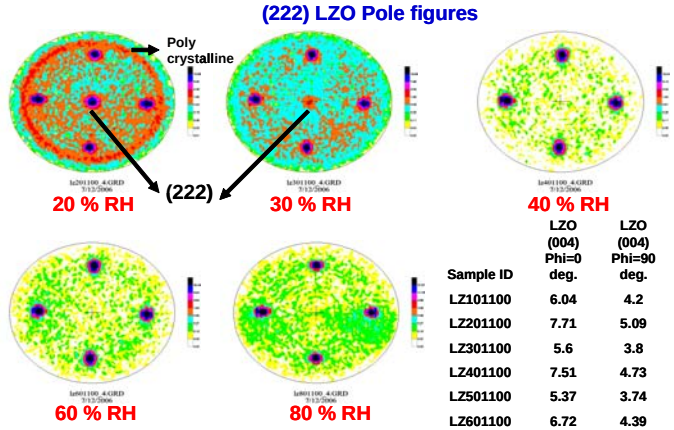


Sample ID	LZO (004) Phi=0 deg.	LZO (004) Phi=90 deg.	LZO (222) Phi deg.
LZ10900	5.94	3.96	6.28
LZ20900	5.02	4.08	6.09
LZ30900	5.17	3.81	6.27
LZ40900	5.16	4.57	6.4
LZ50900	4.77	3.44	5.93

No big change in texture with % RH

Both crystallinity and % cube texture improved with higher % RH during coating for LZO films processed at 1100 °C

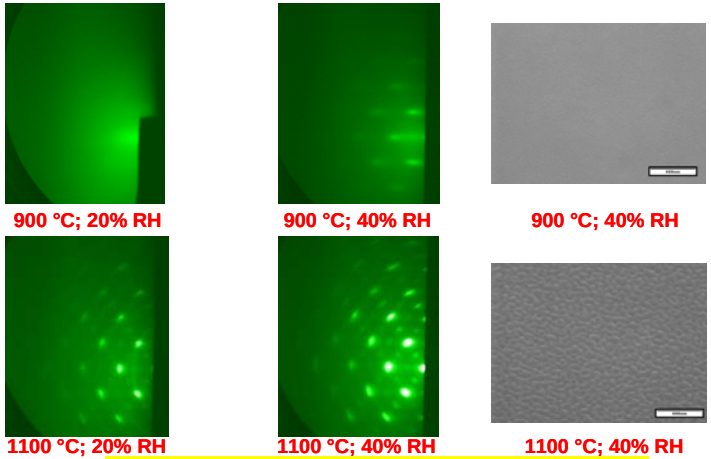
(222) LZO Pole figures



Sample ID	LZO (004) Phi=0 deg.	LZO (004) Phi=90 deg.	LZO (222) Phi deg.	Cube texture
LZ101100	6.04	4.2	5.9	61.20%
LZ201100	7.71	5.09	6.42	57.10%
LZ301100	5.6	3.8	5.87	64.90%
LZ401100	7.51	4.73	6.64	94.50%
LZ501100	5.37	3.74	6.63	94.70%
LZ601100	6.72	4.39	6.7	92.60%
LZ801100	4.23	3.11	5.98	88.10%

No big change in texture except % cube texture with % RH

RHEED and SEM analysis of MOD LZO surface processed at 900 and 1100 °C under 20% RH and 40% RH during coating



Smooth films at higher % RH during coating and 900 °C
Process conditions transferred to slot-die coating conditions

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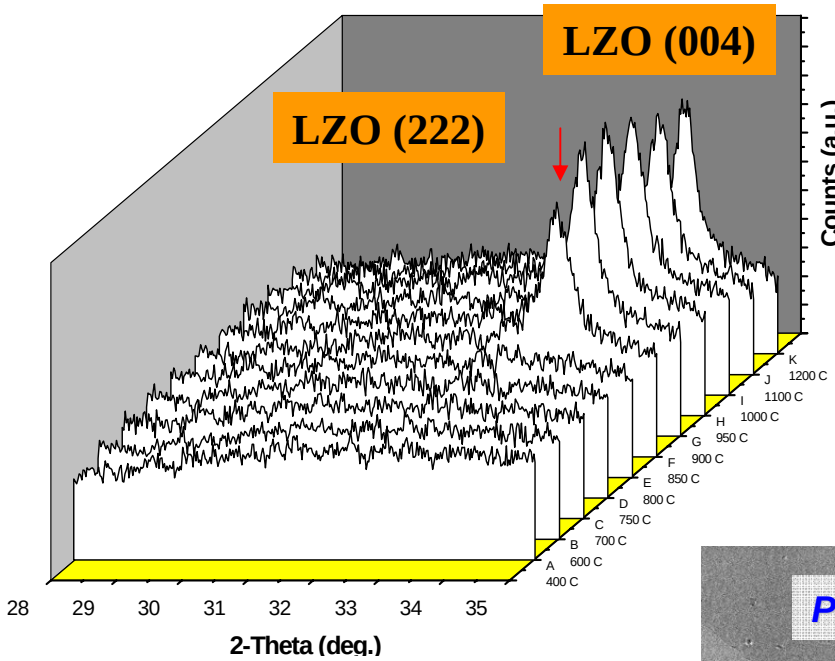
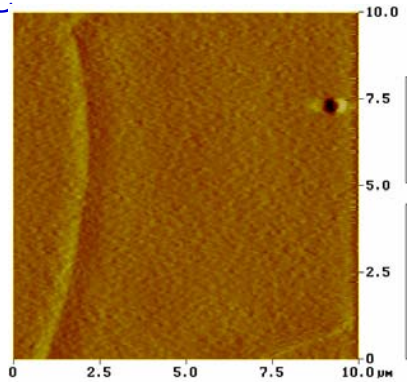
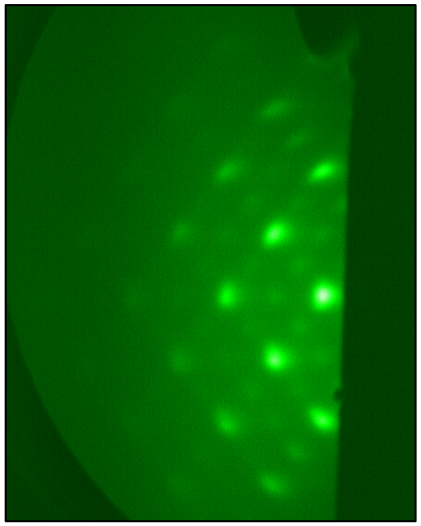
Highly textured and smooth LZO films obtained at 900 °C



- HTXRD @ various T
- LZO *(00l)* nucleate at 850°C
- LZO crystallization complete at 900°C in 15 min.



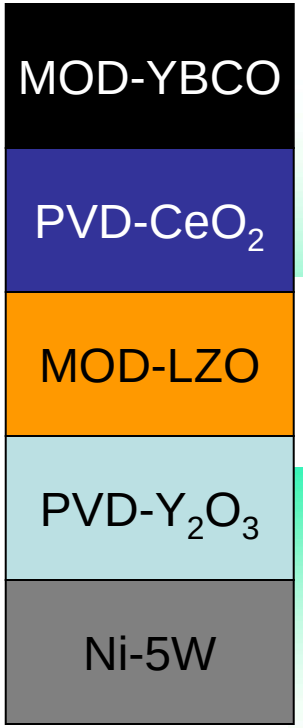
MOD-LZO @900° C/Y_2O_3/NiW

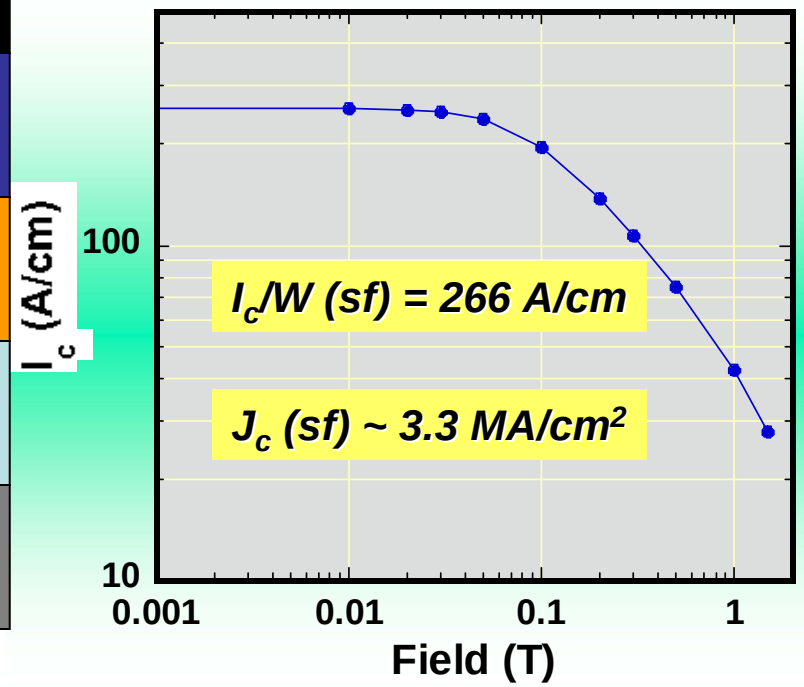


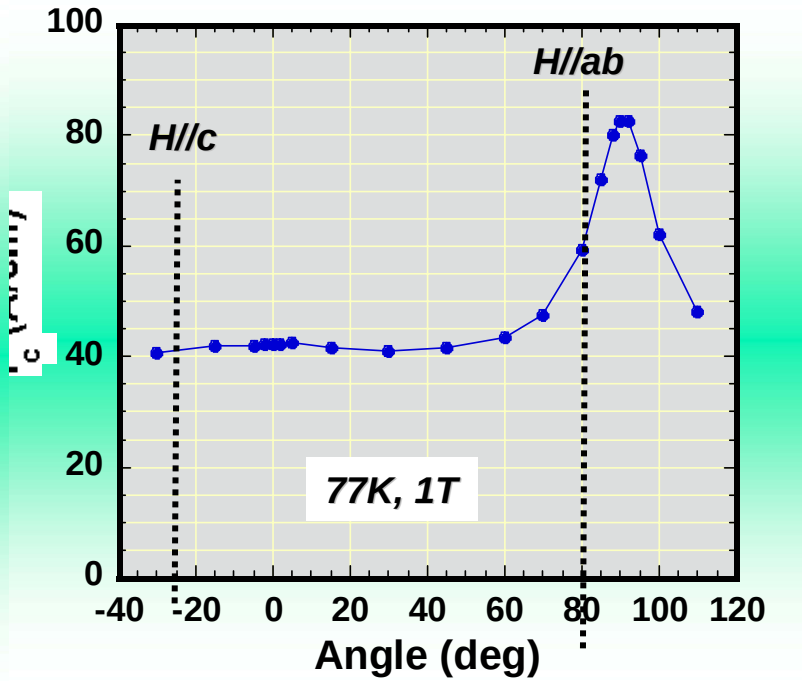
Temp./time (°C/min)	Surface roughness
900 (15)	Ra 0.5 nm
950 (30)	Ra 1.5 nm
1000 (20)	Ra 1.9 nm
1050 (15)	Ra 1.9 nm
1100 (10)	Ra 2.0 nm

We will now follow-up with MOD CeO_2/MOD YBCO

266 A/cm achieved on a continuously processed slot-die coated LZO films at 1100°C







FY05: 255 A
FY06: 266 A

This demonstrates that LZO is an excellent barrier material
Optimization of MOD CeO_2 cap is in progress

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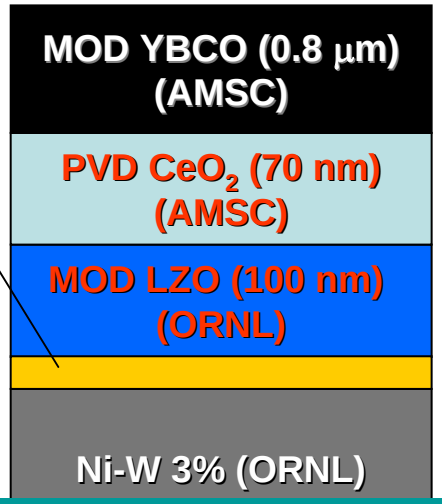
LZO processing temperature reduction raises J_c

FY2006: LZO via high-speed Slot-die coating – continuous process. Annealing conditions: 1100 ºC, 15 min.

FY2006: Short spin-coated sample. Annealing conditions: 900 ºC, 15 min.

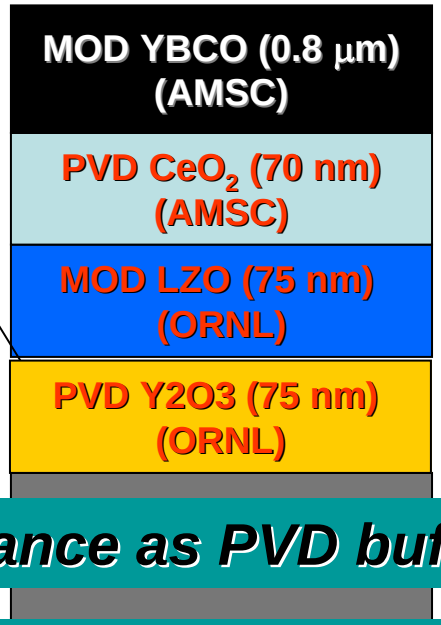
I_c = 266 A/cm-width
J_c = 3.3 MA/cm^2

PVD 10nm Y_2O_3



I_c = 314 A/cm-width
J_c = 3.9 MA/cm^2

75nm Y_2O_3



Solution buffer yields similar performance as PVD buffers

DOE Milestone of 300 A/cm on solution buffer met!

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FY 2006 Plans and Performance

FY2006 Plans

- **Fabricate films of REBCO with and without nanoparticle additions and compare pinning characteristics with YBCO with nanoparticle additions**
- **Further develop nanoparticle additions in the appropriate volume fractions to optimize pinning properties of YBCO at different temperatures (i.e., 77K, 65K, and 40K).**

FY2006 Performance

- ✓ **High-J_c Nd-123, Sm-123, Gd-123 films were fabricated on technical substrates**
- ✓ **REBCO films have better pinning than YBCO films**
- ✓ **Incorporation of columnar defects comprised of self-assembled BZO nanodots within REBCO films, results in significant enhancement in pinning**
- ✓ **Thick REBCO films with columnar defects have a values as low as 0.2. This is because of a high density of c-axis correlated defects**

FY 2006 Plans and Performance

FY2006 Plans (cont'd)

- Correlate HTS properties with the level of $BaZrO_3$ incorporated in films as 3D, self-assembled, columnar nanodot arrays.
- Conduct detailed measurement and analysis of the wide-range thickness dependence of critical currents in *ex situ* YBCO on RABiTS.

FY2006 Performance

✓ Self-assemble linear stacks of BZO nanoparticles approximate columnar defects along *c*-axis. T_c is progressively suppressed by dopant fraction x. Analysis suggests targets for optimization wrt columnar stack size and density

✓ Nanostructure of PVD *ex situ* YBCO/RABiTS controlled by composition and processing: large, sparse Y_2O_3 precipitates. Flux pinning by Y_2O_3 precipitates analyzed with model of Ivlev & Ovchinnikov: Temperature, Field and Thickness dependence

FY 2006 Plans and Performance

FY2006 Plans (cont'd)

- **Continue optimizing nanoparticle substrate surface modifications for flux-pinning in thick *in situ* and *ex situ* YBCO on RABiTS, and extend to YBCO on IBAD Templates.**

FY2006 Performance

- ✓ **Substrate surface pre-treatment has been extended to decoration of metal template surfaces**
- ✓ **Enhancement in J_c observed for all field orientations**
- ✓ **Demonstrated further J_c improvements via combination of nanoparticle BZO surface decoration and YBCO doping**
- ✓ **SuperPower has evaluated ORNL nanoparticle treatment of IBAD-MgO templates: *J_c improvements in 1.1um MOCVD YBCO***

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FY 2006 Plans and Performance

FY2006 Plans (cont'd)

- **Fundamental studies of epitaxial growth of multi-functional buffer layers on textured substrates**

FY2006 Performance

✓ **I_c of 266 A/cm or a J_c of 3.3 MA/cm^2 was demonstrated on slot-die coated LZO using a fully continuous process**

✓ **I_c of 314 A/cm or a J_c of 3.9 MA/cm^2 was demonstrated on a short spin-coated sample with the LZO annealed at 900ºC. PVD performance matched.**

✓ **SIMS studies showed that LZO is still the best MOD barrier layer for scale-up**

FY 2007 Plans

- Conduct fundamental studies of epitaxial growth of simplified and multi-functional buffers on technical substrates.
- Perform experiments to verify and refine predictive model to explain pinning by columnar defects.
- To introduce columnar defects without significant reduction in H //ab on technical substrates.
- Fabricate 3-5 µm with 3D self-assembled nanodots to exceed performance requirements for applications. Demonstrate an I_c of 300 A/cm @ all field angles and 65 K; 3T.
- Create columnar defects on substrates followed by infiltration by superconductor via in-situ and ex-situ techniques.
- Extend nanoparticle decoration to thicker REBCO films on technical substrates.
    - Investigate size, density and material performance on LMO/IBAD-MgO templates.
- Continue correlation of HTS properties in thicker REBCO films with 3D, self-assembled linear nanodot arrays.
- Extend the theoretical model developed for BaF_2 coated conductors to commercial 2G wires.
- Understand the mechanisms of 3D self-assembly of nanodots to enable better control of defect structures to further optimize pinning characteristics. (if committee recommends and funding is available)

Research Integration

Interaction with companies; universities and national labs.

- **Work on IBAD substrates – Part of ORNL-SuperPower CRADA**
- **Work on RABiTS substrates – Part of ORNL CRADA interactions with AMSC and Oxford**
- **MetOx – Provide characterization support for the development of low-cost MOCVD buffers/HTS**
- **University of Tennessee: T. Aytug, J.R. Thompson, S. Sathyamurthy**
- **University of Florida: D.P. Norton**
- **University of Kansas: J.Z. Wu**
- **University of Wisconsin-Madison: E. Hellstrom, D. Larbalestier, M. Feldmann**
- **Columbia University: Sui-Wai Chan**
- **University of Houston: R. Meng, I. Rusakova, C.W. Chu**
- **University of Oslo, Norway: T.H. Johansen**
- **University of Alabama at Mobile: A.A. Gapud**
- **Los Alamos National Laboratory: P.N. Arendt, L. Stan, T.G. Holesinger**
- **National Renewable Energy Laboratory: R.N. Bhattacharya**

Communications:

- **Many publications**
- **Numerous presentations**
- **Nano50 Award**
- **Patents issued and filed disclosures**
- **Information also dispersed via regular CRADA teleconferences/meetings/ annual reports/ ORNL websites: http://www.ornl.gov/sci/htsc/publications.htm**

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Presentations during current review period = 14 invited, 25 total

T. Aytug, et al., *J. Appl. Phys.* **98**, (11) 114309 (December 2005).
T. Aytug, et al., *J. Mater. Res.* **20**, (11) 2988 (November 2005).
T. Aytug, et al., *IEEE Trans. Appl. Supercond.* **15**, (2) 2977 (June 2005).
C. Cantoni, et al., *IEEE Trans. Appl. Supercond.* **15**, (2) 2981 (June 2005).
C. Cantoni, et al., p. 53 in *Interfaces in Electronic Materials*, ed. by L. Cook, D. Misra, S. Mukhopadhyay, W. Wong-Ng, O. Leonte, and K. Sundaram, The Electrochemical Society, Inc., Pennington, New Jersey (2006). **(INVITED)**
N. Cheggour,et al., *Supercond. Sci. Technol.* **18**, (12) S319 (December 2005).
R. Feenstra, et al., *IEEE Trans. Appl. Supercond.* **15**, (2) 2803 (June 2005).
D. M. Feldmann, et al., *J. Mater. Res.* **20**, (8) 2012 (August 2005).
D. M. Feldmann, et al., *J. Mater. Res.* **21**, (4) 923 (April 2006).
A. A. Gapud, et al., *Supercond. Sci. Technol.* **18**, 1502 (November 2005).
A. A. Gapud, et al., *IEEE Trans. Appl. Supercond.* **15**, (2) 2578 (June 2005).
A. Goyal, et al., *Physica C* **426-431**, 1083 (October 2005).
A. Goyal, et al., *Supercond. Sci. Technol.* **18**, 1533 (November 2005).
T. J. Haugan, et al., *Physica C* **425**, (1-2) 21 (September 2005).
T. G. Holesinger, et al., p. 111 in *Interfaces in Electronic Materials*, ed. by L. Cook, D. Misra, S. Mukhopadhyay, W. Wong-Ng, O. Leonte, and K. Sundaram, The Electrochemical Society, Inc., Pennington, New Jersey (2006).
A. O. Ijaduola, et al., *Phys. Rev. B* **73**, 134502 (April 2006).
S. Kang, et al., *Science* **311**, (5769) 1911 (March 2006).
K. Kim, et al., *Supercond. Sci. Technol.* **19**, (4) R23 (April 2006).
K. Kim, et al., *IEEE Trans. Appl. Supercond.* **15**, (2) 2997 (June 2005).
M. Laver, et al., *Phys. Rev. Lett.* **96**, (16) 167002 (April 2006).
M. M. Ozer, J. R. Thompson, and H. H. Weitering, *Nature Physics* **2**, (3) 173 (March 2006).
A. Palau, et al., *Appl. Phys. Lett.* **88**, 122502 (March 2006).
A. Palau, et al., *Appl. Phys. Lett.* **88**, (13) 132508 (March 2006).
A. Palau, et al., *IEEE Trans. Appl. Supercond.* **15**, (2) 2790 (June 2005).
A. Polyanskii, et al., *Phys. Rev. B* **72**, (17) 174509 (November 2005).
M.P. Paranthaman, et al., IEEE Trans. on Appl. Supercond. 15 (2) 2632-2634 (June 2005).
M.P. Paranthaman, et al., Ceramic Transactions, Vol 169, pp. 49-58 (2005).
J. R. Thompson, et al., *Supercond. Sci. Technol.* **18**, 970 (June 2005).
M. Yethiraj, et al., *Phys. Rev. B* **72**, (6) 060504 (August 2005).
W. Wong-Ng, et al., *Appl. Phys. Lett.* **88**, 102507 (March 2006).
Y. Zhang, et al., *IEEE Trans. Appl. Supercond.* **15**, (2) 2659 (June 2005).