

# Comparative analysis of particle irradiation and second-phase additions effects on the critical current densities of $\text{YBa}_2\text{Cu}_3\text{O}_7$ single crystals, thin films, and coated conductors

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**IRradiation Effects on HTS for Fusion**  
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## Motivation

- Radiation damage in the superconducting magnets is a concern for the fusion reactors community. However...
- ...disorder (material defects) is required for vortex pinning
- ReBCO-based CCs have the highest  $J_c$  in any known SC  $\Rightarrow$  effective strong pinning defects
- Added defects in CCs (e.g. second phases) are optimized for high  $J_c$
- Irradiation-induced defects will interact with pre-existing disorder
- Irradiation will start modifying the properties of the CC magnets in fusion reactors from day one of operation

## The LANL team



Boris Maiorov



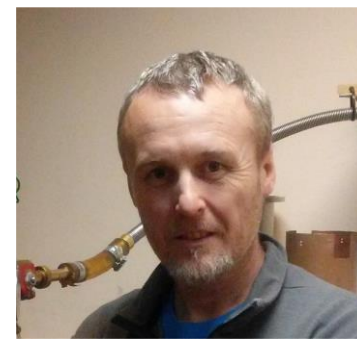
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Ulrich Welp

Alex Koshelev

Wai Kwok

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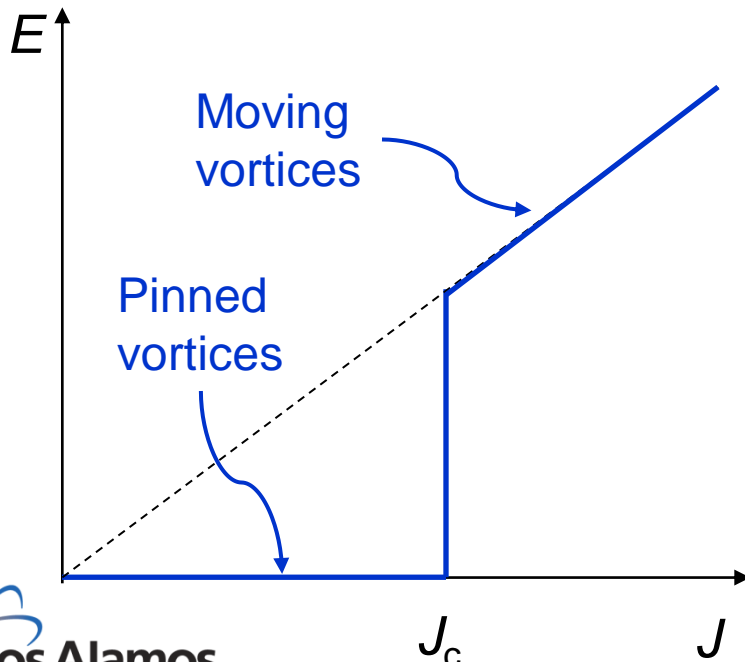
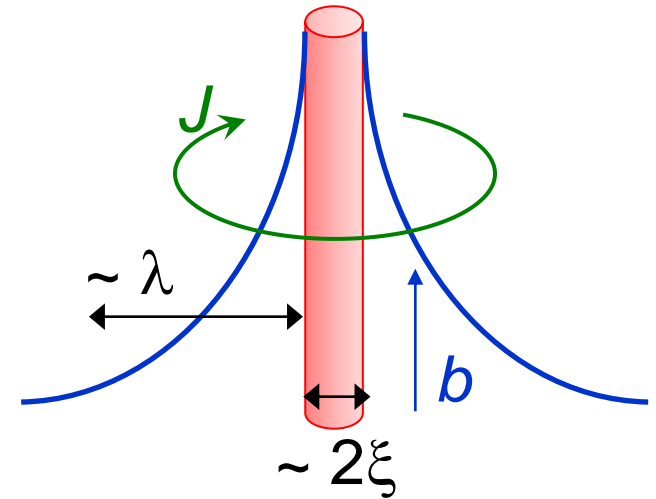
Thanks to all  
the collaborators!!

# Outline

- Introduction to vortex matter – defects, pinning centers and critical currents
- $J_c$  enhancement in YBCO single crystals by particle irradiation
- YBCO films have the highest  $J_c$  of any known SC. Can it be enhanced further?
- Engineering the vortex pinning landscape in YBCO films and coated conductors:
  - ✓ Second phase additions
  - ✓ Particle irradiation: Further  $J_c$  enhancement is still possible!
- Cooperation and competition effects in mixed pinning landscapes
- Conclusions

# Vortices appear in the "mixed state" of type II superconductors

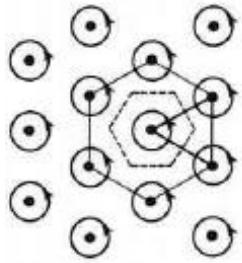
- Quantized "tubes" of magnetic field
  - each carries a flux quantum  $\Phi_0$
- Central filament where superconductivity is suppressed (core) surrounded by circulating currents and associated magnetic field.
- Energy: magnetic + kinetic (currents) + core



- Electric currents exert force on vortices  $\Rightarrow$  vortex motion is dissipative  $\Rightarrow$  resistance
- Motion may be precluded by material disorder (reduced core energy)
- Vortices remain pinned until  $J$  reaches the critical current density  $J_c$



# Vortex matter physics arises from the interplay of 3 energies



$$a = \left(\frac{4}{3}\right)^{1/4} \left(\frac{\Phi_0}{B}\right)^{1/2}$$

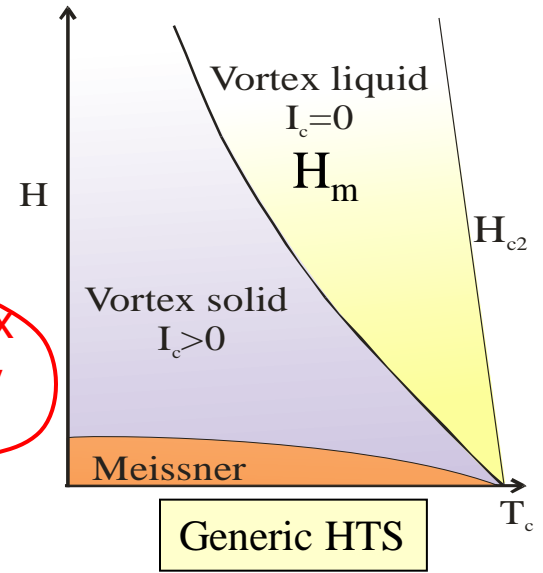
## Vortex-vortex interactions

controlled by “intrinsic” material properties ( $\lambda, \xi, \gamma$ )

## *Gi* Thermal fluctuations

- produce flux creep & vortex liquid phases
- bad for applications

Main source of complex vortex phenomenology in HTS

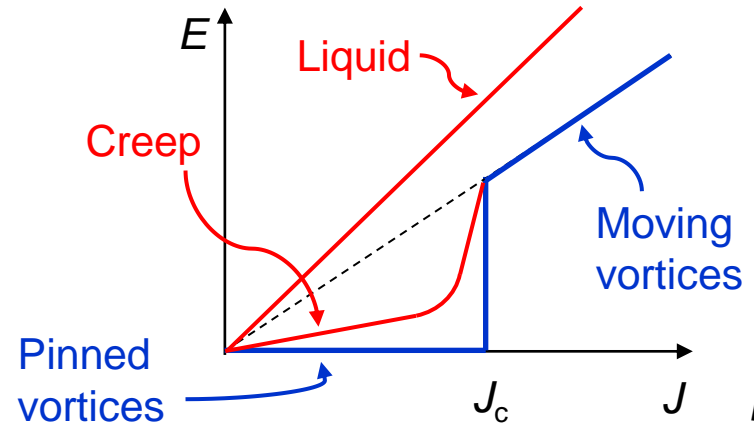


## Vortex-defects interactions

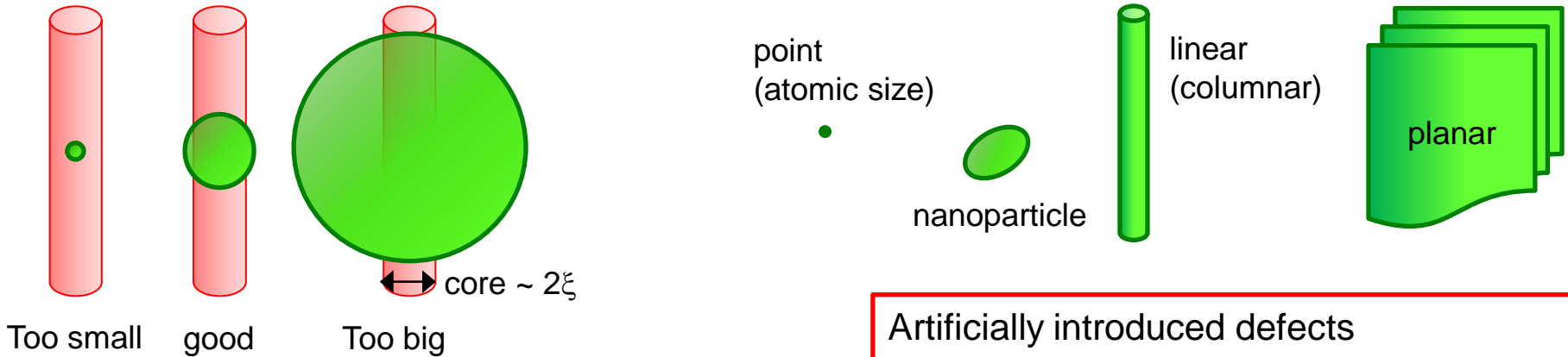
- “Extrinsic” effect responsible for vortex pinning
- $J_c$  can vary by orders of magnitude in the same material
- “Pinning landscape”

Ordered lattice (Abrikosov) Equilibrium

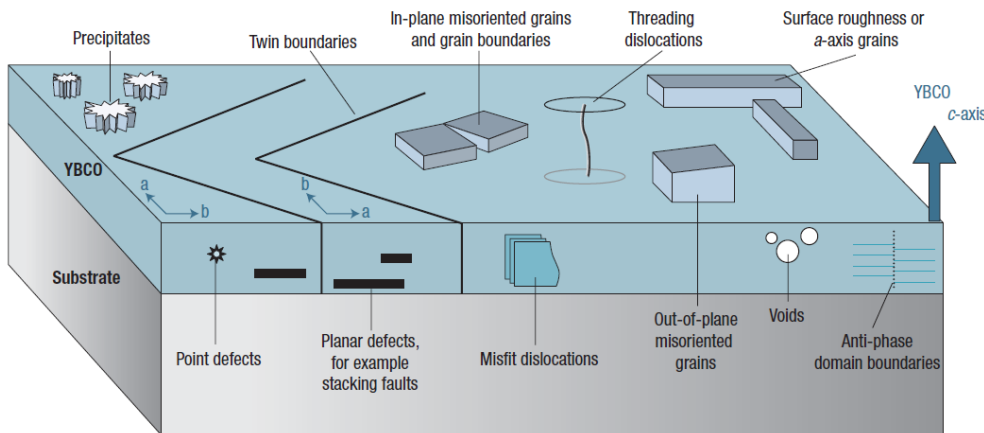
Disordered arrays Metastable states



# Many types of defects can act as pinning centers, some are better than others...

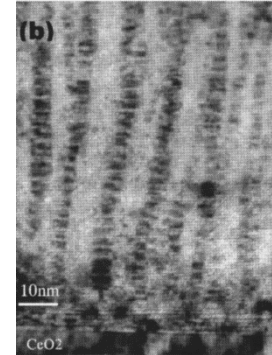


## Defects formed during fabrication (e.g., in YBCO films)

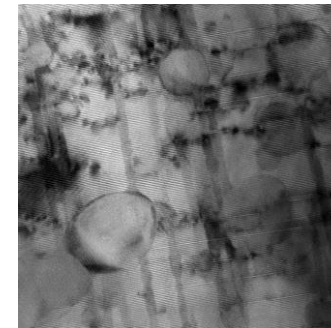
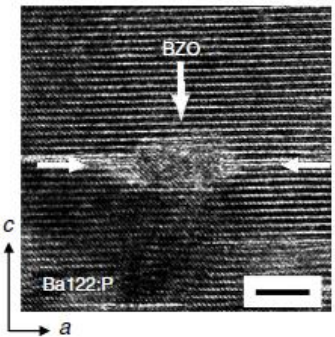


## Artificially introduced defects Popular methods in HTS:

### Particle irradiation



### Chemical incorporation of second phases



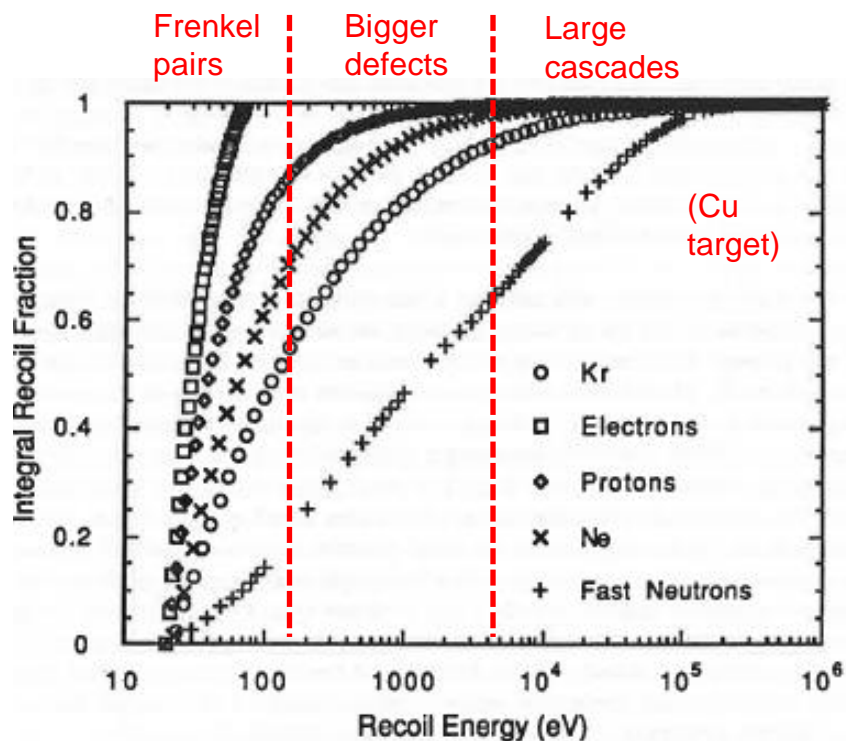
### Combinations

# Particle irradiation of HTS was a very popular activity in the early 1990s

Incident ions transfer energy to the solid by:

Direct collisions with lattice nuclei  
(nuclear or non-ionizing energy loss):  
Dominant for light ions up to few MeV

## Localized uncorrelated disorder

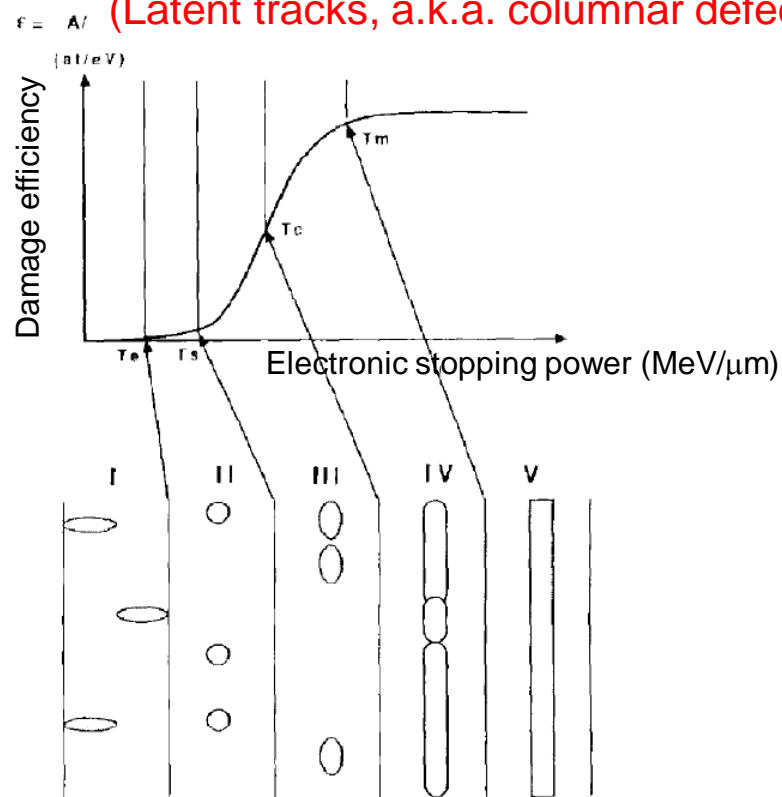


M.A. Kirk & H.W. Weber (1992)

Ionization or electronic excitations  
(electronic or ionizing energy loss):  
Dominant for heavy ions 100s of MeV to GeV

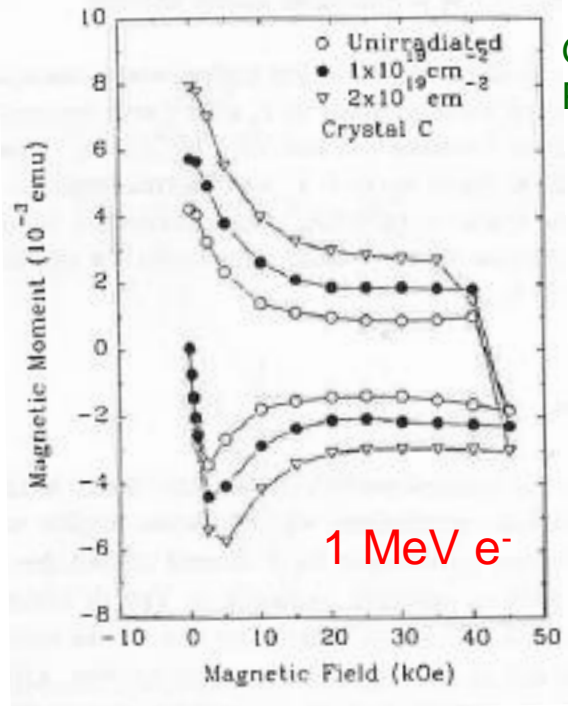
## Correlated disorder

(Latent tracks, a.k.a. columnar defects)

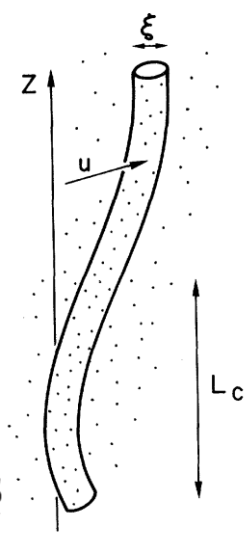


F. Studer & M. Toulemonde (1992)

# Irradiation creates effective pinning centers in clean YBCO single crystals

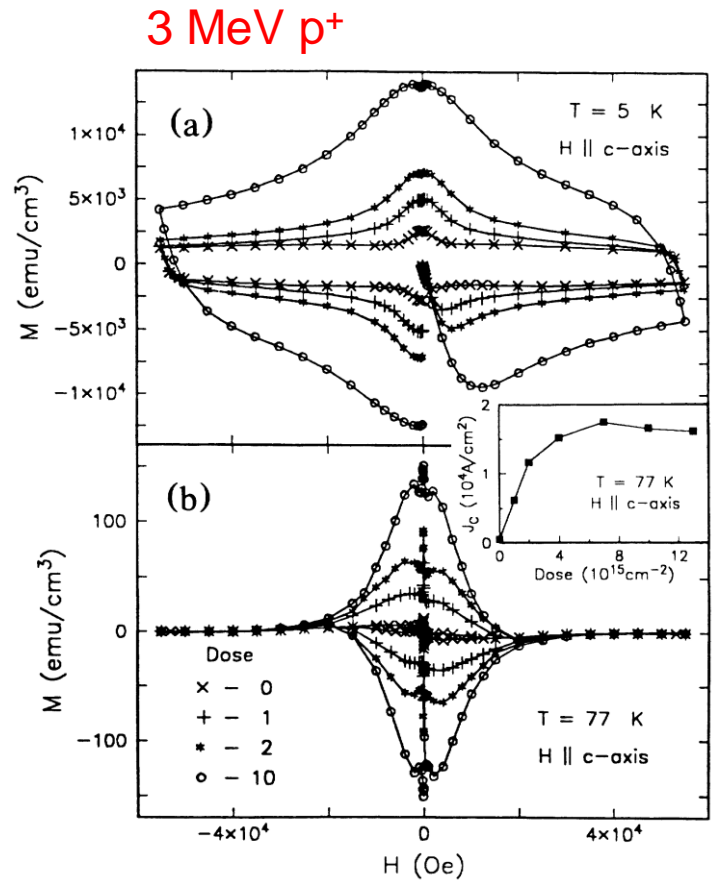


Giapintzakis *et al.*, PRB **45**, 10677 (1992)



Even point defects are effective in YBCO because

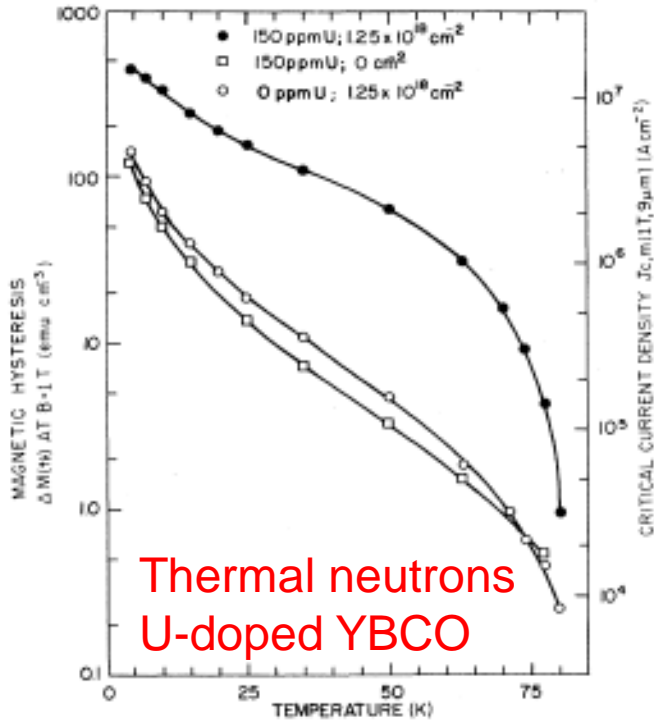
- $\xi$  is small
- affects whole unit cell
- there are many (collective pinning)



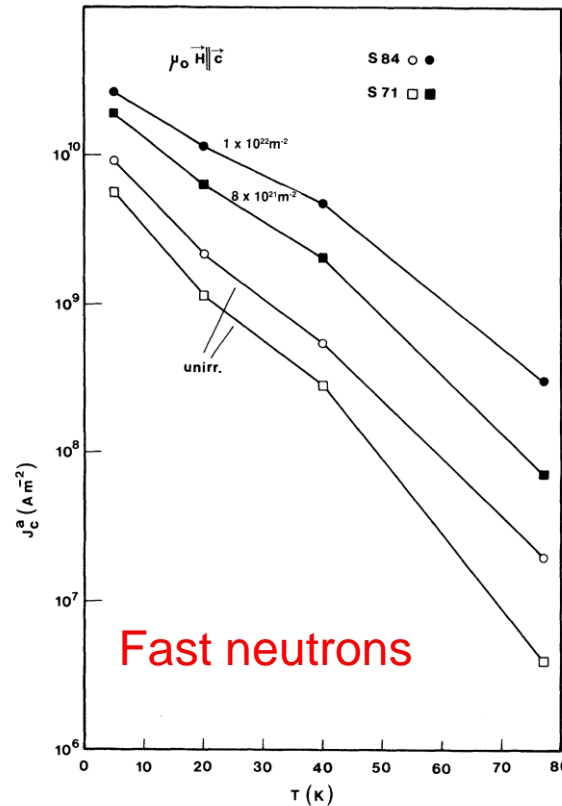
L. Civale *et al.*, PRL **65**, 1164 (1990)



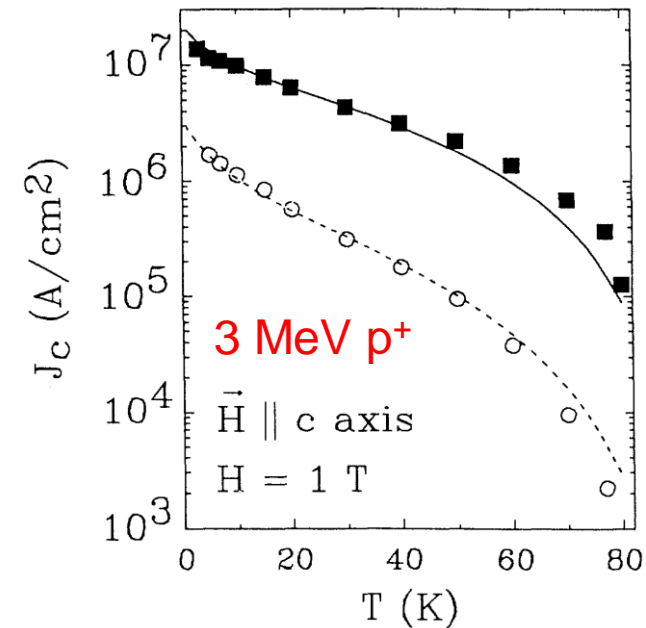
# Orders of magnitude increases in $J_c$ in clean YBCO single crystals



R.L. Fleischer *et al.*,  
PRB **40**, 2163 (1989)



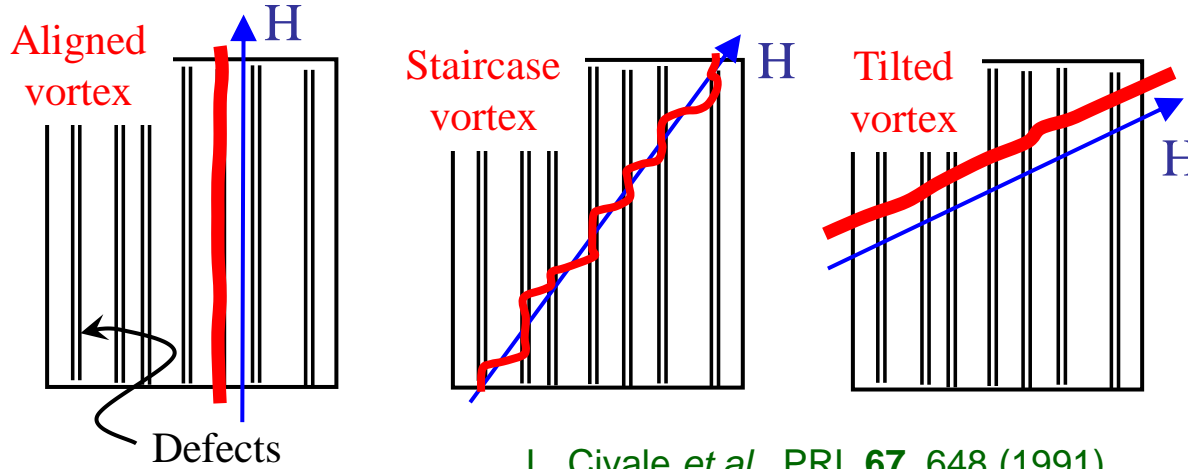
F.M. Sauerzopf *et al.*,  
PRB **43**, 3091 (1991)



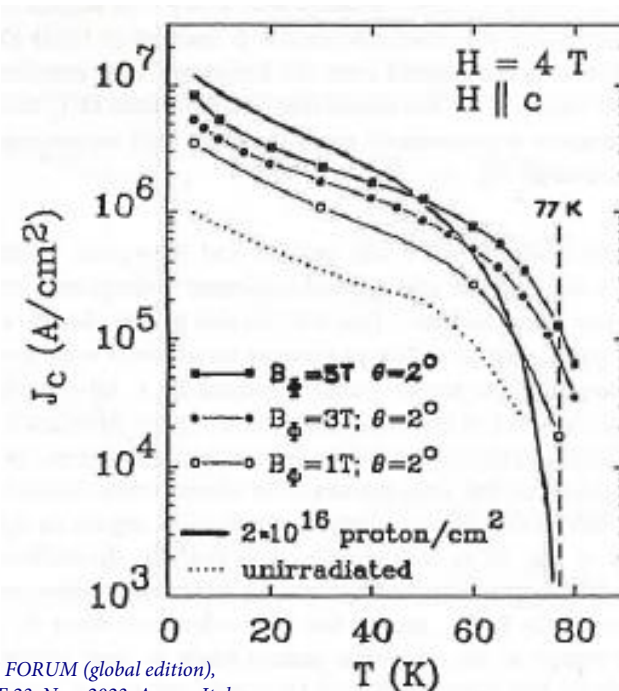
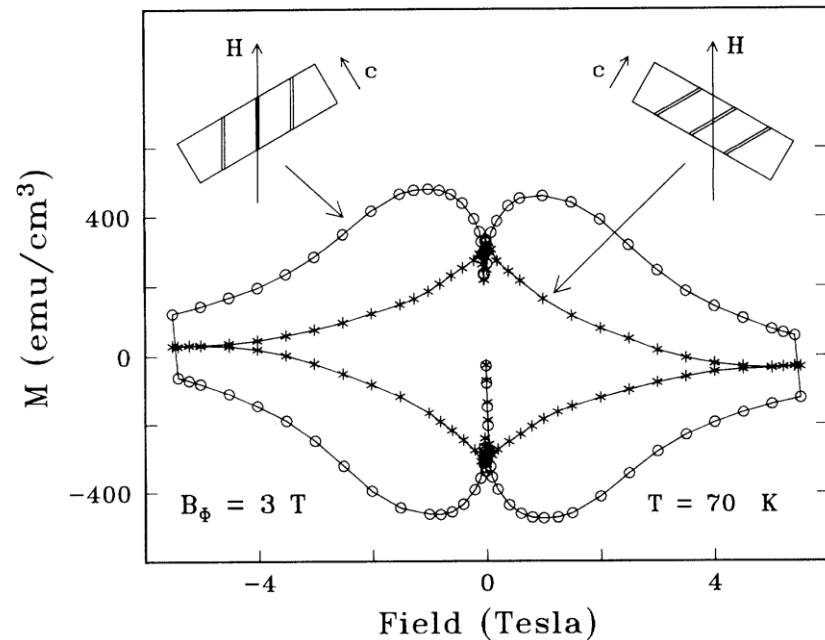
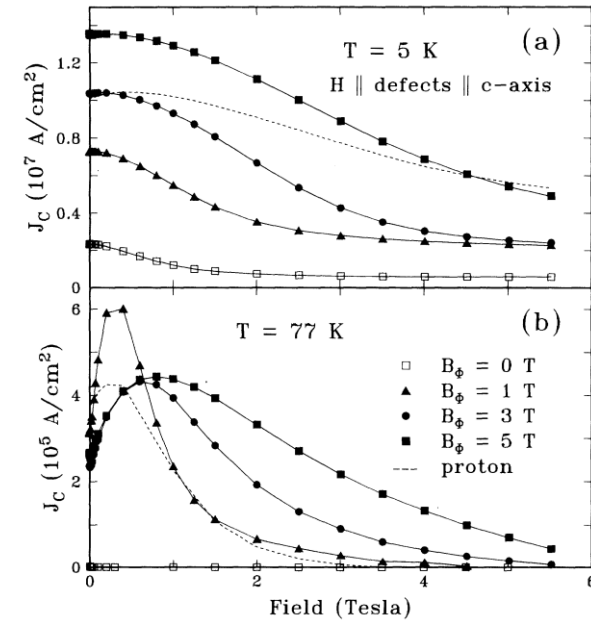
J.R. Thompson *et al.*,  
PRB **47**, 14440 (1993)

# High energy heavy ion irradiation creates aligned columnar defects

## Directional pinning



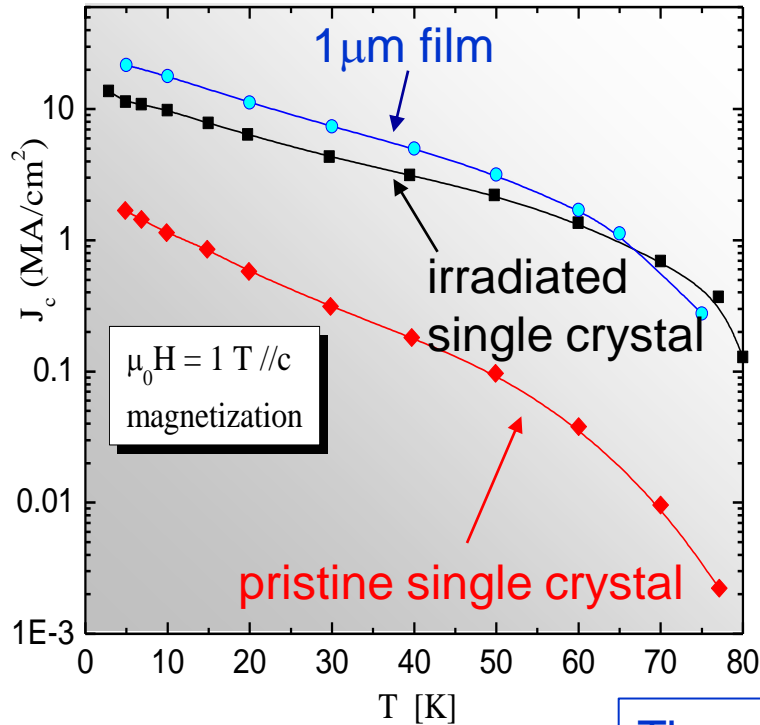
L. Civale *et al.*, PRL **67**, 648 (1991)



CDs are the most effective pinning centers for  $H \parallel \text{defects}$  and below matching field  $B_\Phi$

However....

However,  $J_c$  in “standard” YBCO films is higher than the best that could be achieved in irradiated single crystals



The maximum possible  $J_c$  is the depairing current density

$$J_0(T) = \frac{cH_c(T)}{3\sqrt{6}\pi\lambda(T)}$$

YBCO films have the highest

$J_c \sim 100 \text{ MA/cm}^2$

$J_c/J_0 \sim 0.3$

of any known superconductor

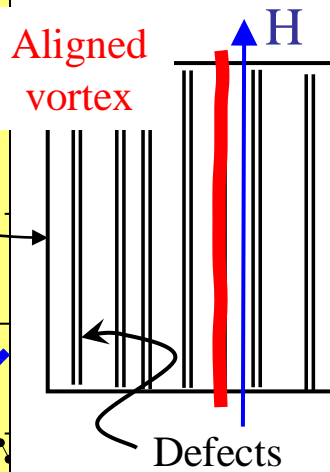
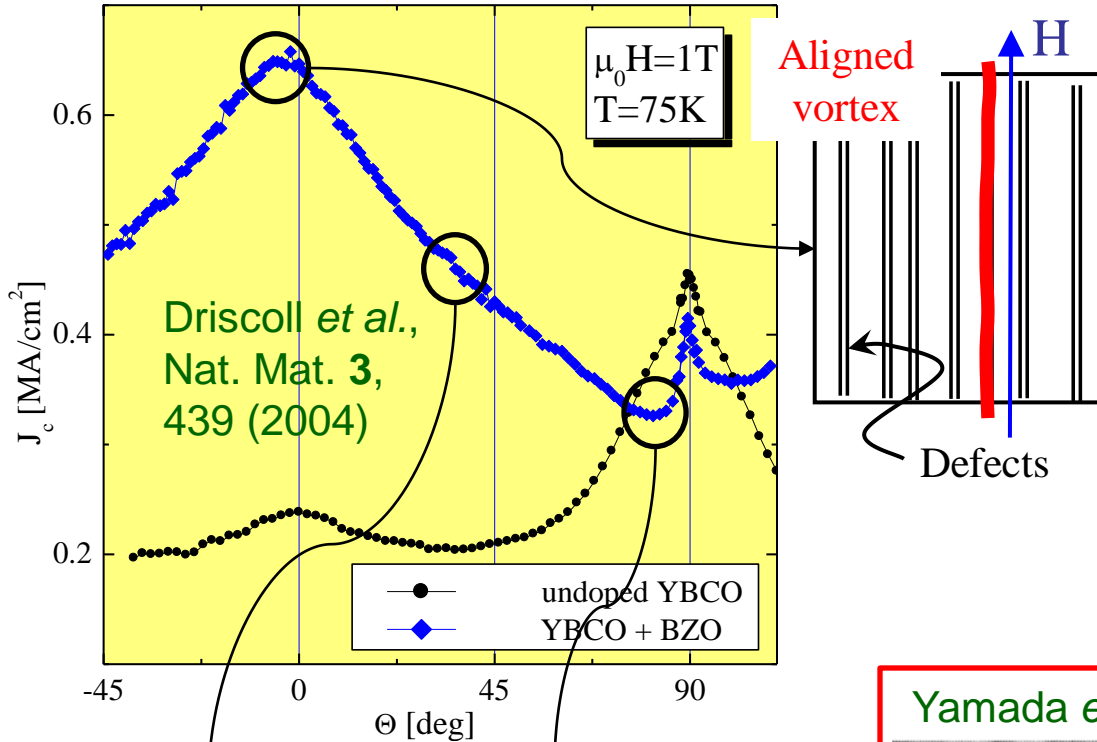
There are two reasons to study YBCO films:

- Technological applications (obvious)
- Basic science: they are an “extreme case”

Why is the  $J_c$  of YBCO films so high?  
 This was a big mystery for the CC community in the early 2000s

A successful approach: pinning landscape engineering

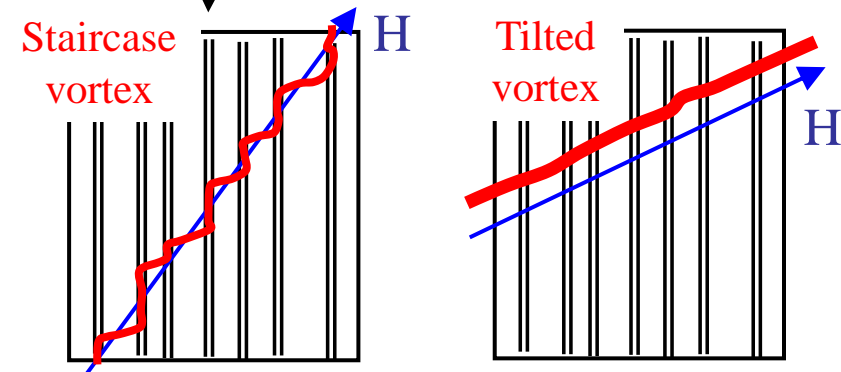
# $J_c$ in YBCO films can be increased by chemical introduction of defects



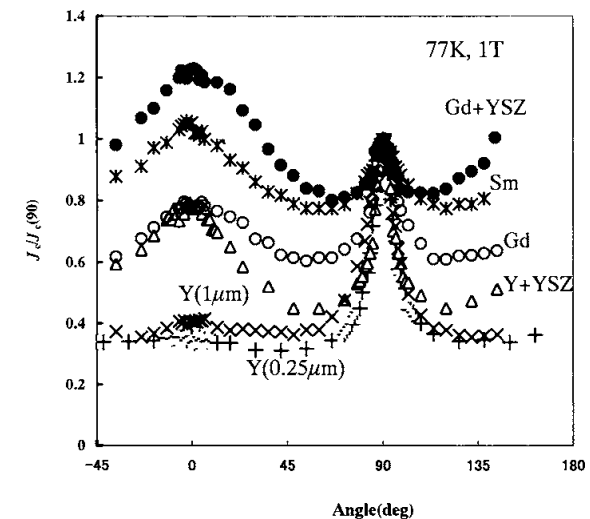
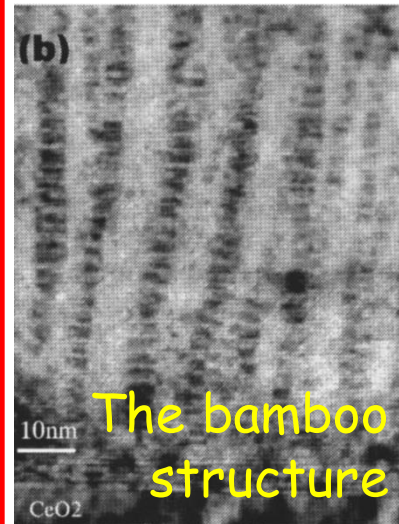
We produced large  $J_c$  increases in PLD YBCO films by adding BaZrO<sub>3</sub> second phases

Angular dependence of  $J_c$ : large peak for  $H//c$ ,  $\Rightarrow$  correlated pinning by self-assembled nanorods (columnar defects).

The BZO doping did not increase the **self-field**  $J_c$ , but produced a much improved in-field performance



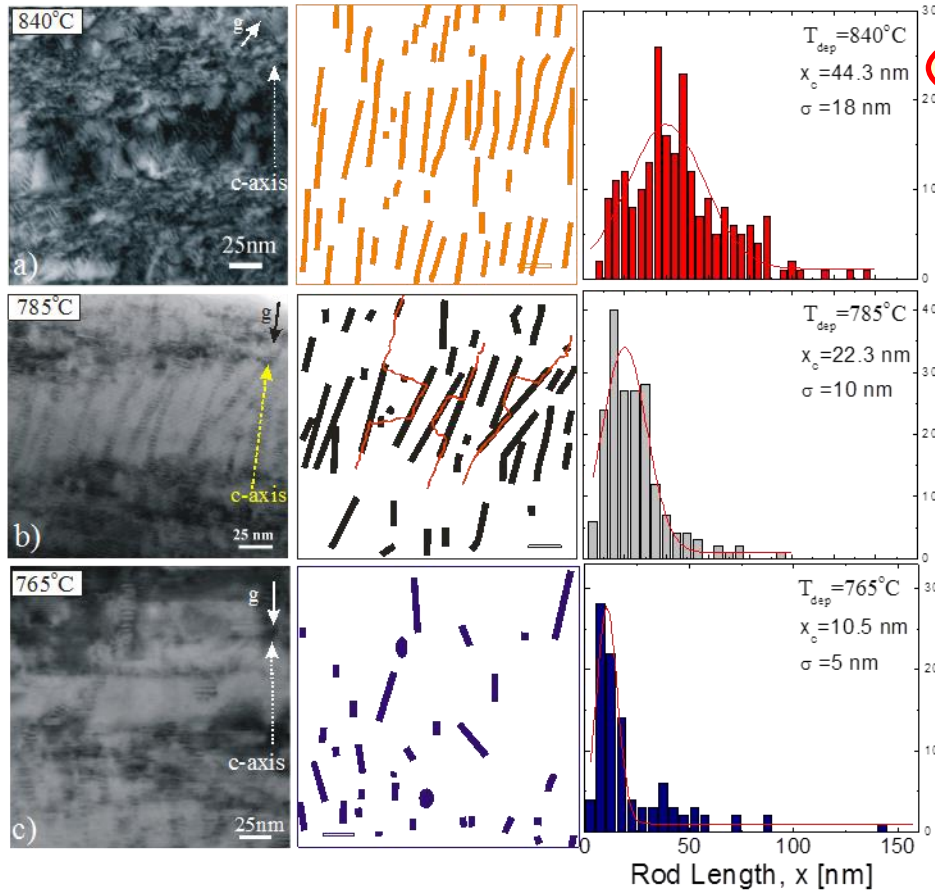
Yamada *et al.* Appl. Phys. Lett. 87, 132502, 2005



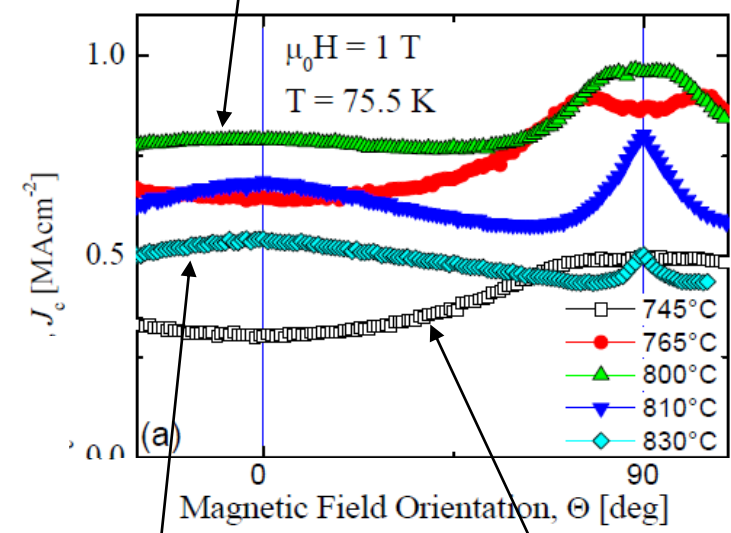
# We learned to nanoengineer the pinning landscape by tuning the growth conditions in PLD YBCO+BZO

Low growth temperature or High rate = random nanoparticles

High growth temperature or Low rate = self-assembled nanorods



*Mixed pinning landscape (splayed nanorods + nanoparticles): best  $J_c$*



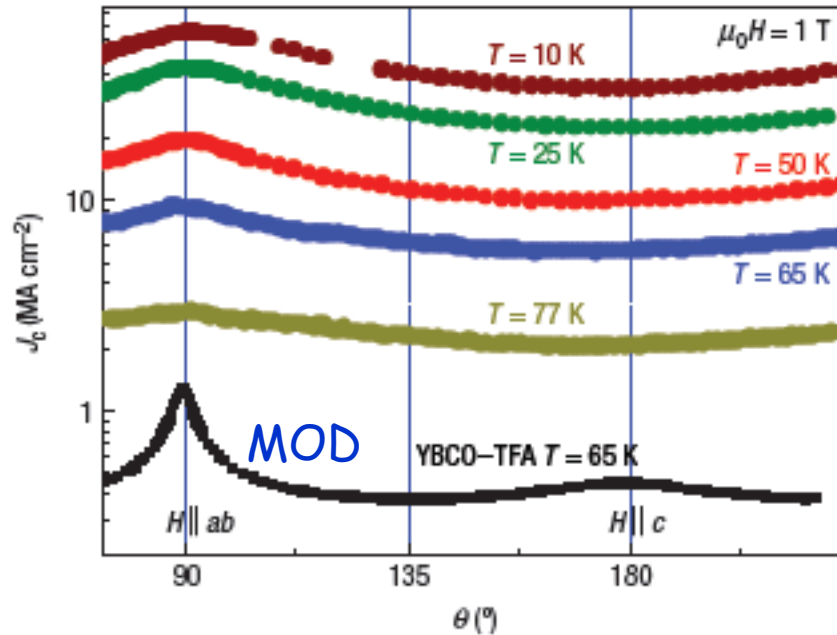
Columnar defects:  
large c-axis peak

Random nanoparticles:  
no c-axis peak

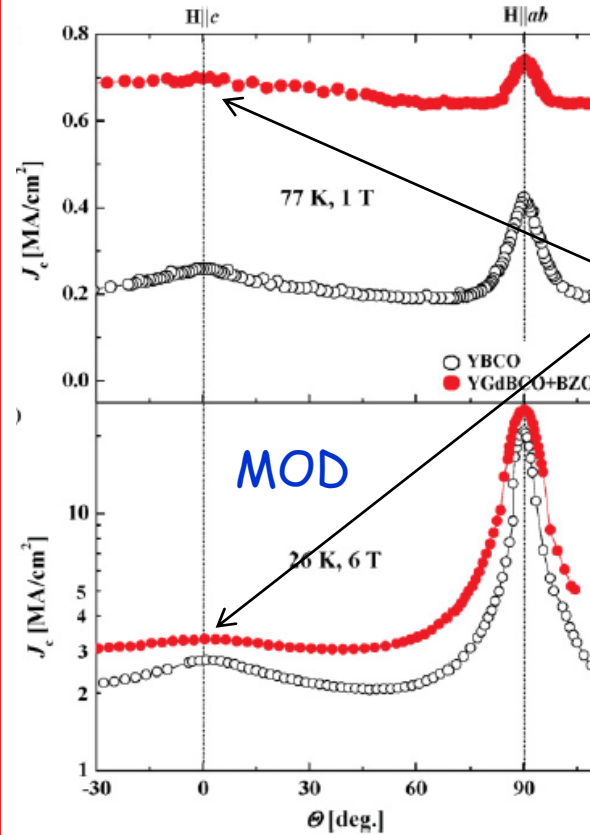
Processing → properties correlations: The columnar growth of Pulsed Laser Deposition (PLD) films promotes the self-assembly of BZO nanorods

# Same BZO additions in films grown by different methods (MOD) or under different conditions may produce strong pinning by random nanoparticles

J. Gutierrez *et al.*, Nat Mat. **6**, 367 (2007)

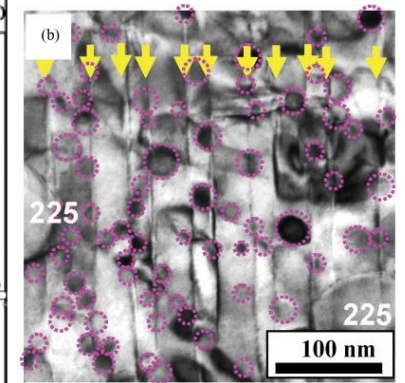


Very strong pinning by nanoparticles and no evidence of c-axis correlated disorder



M. Miura *et al.*, PRB **83**, 184519 (2011)

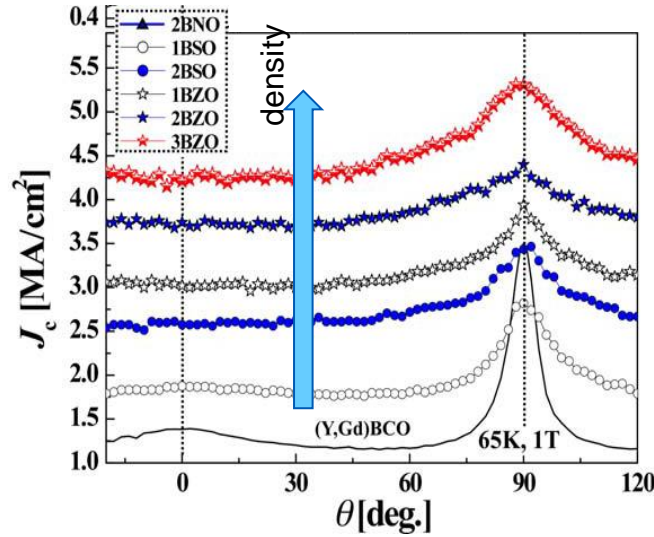
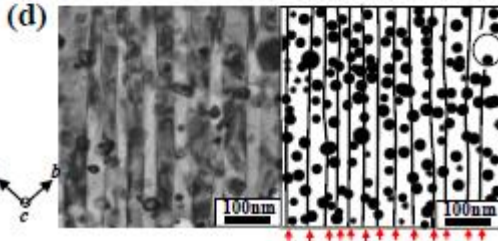
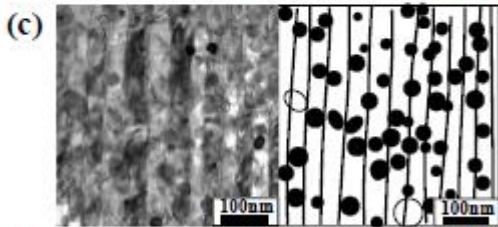
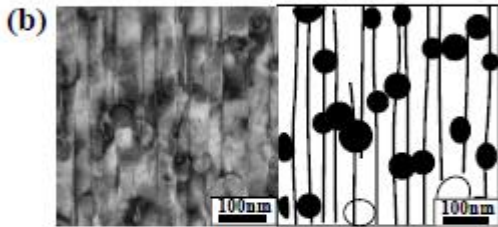
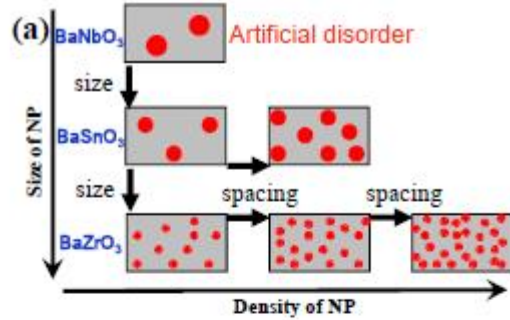
c-axis peak: pinning by twin boundaries



Correlated pinning from twin boundaries is present, but at high  $T$  pinning is dominated by the random nanoparticles

# Influence of the density and size of added random nanoparticles on vortex behavior in YBCO-based CC grown by MOD

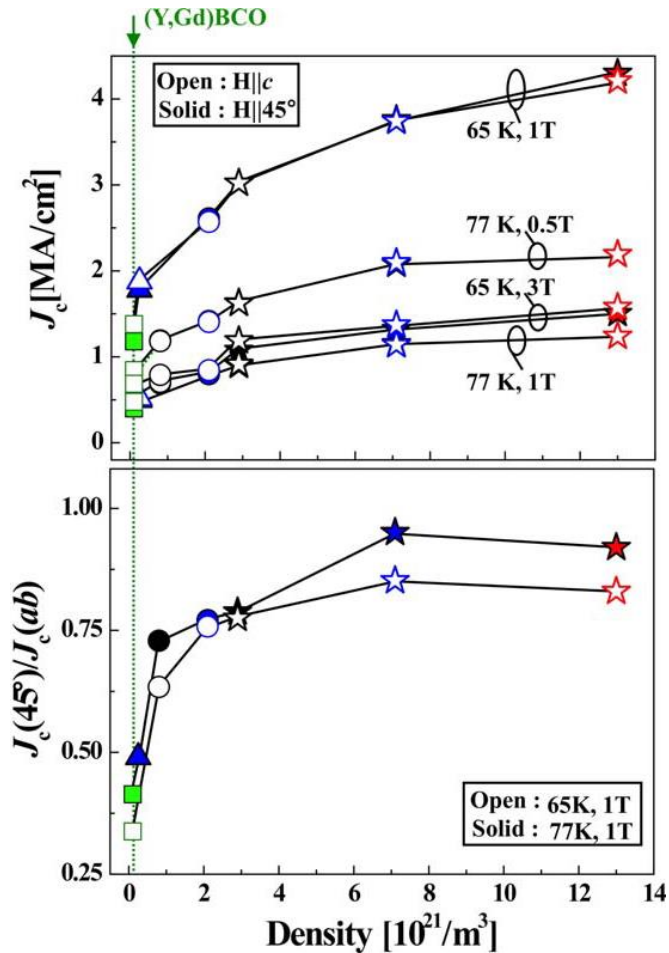
M. Miura *et al.*, *SuST* **26**, 035008 (2013)



As the NP density increases:

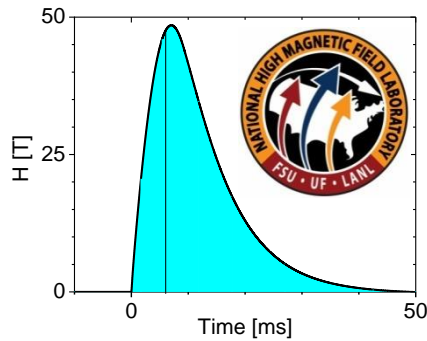
- $J_c$  increases
- The  $J_c$  anisotropy decreases

We need: smaller NPs, higher density



# Influence of the density and size of added random nanoparticles in YBCO-based CC grown by MOD at very high fields

Angular dependent resistivity measurements in pulsed magnetic fields



First measurement of a CC in pulsed fields

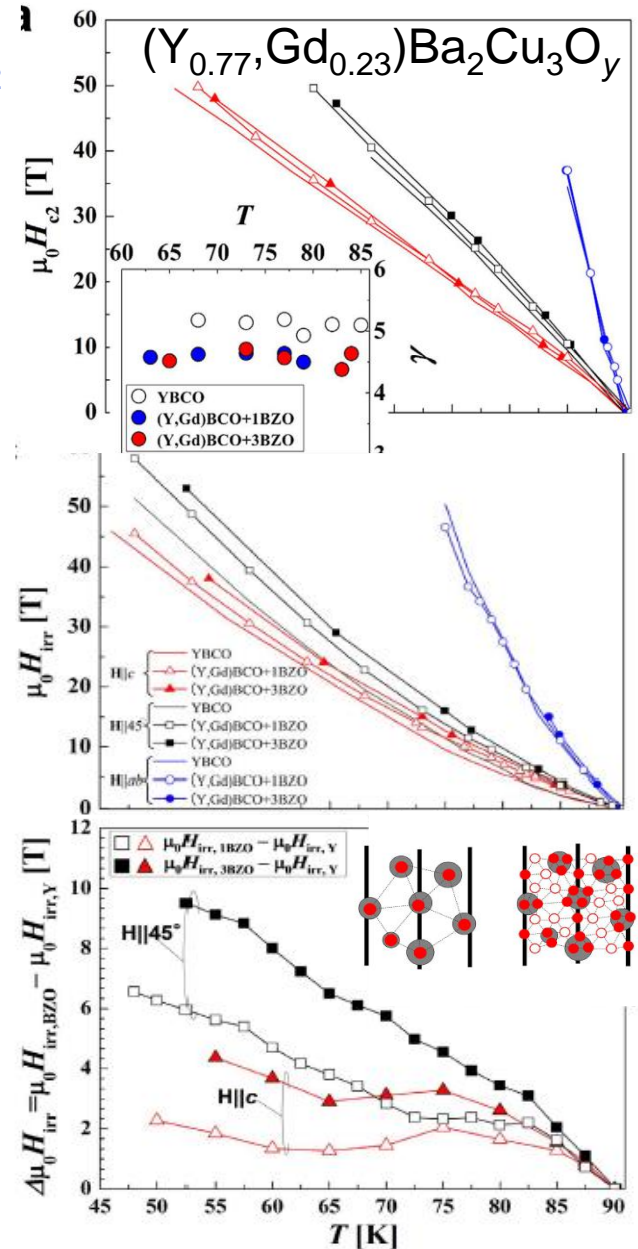
M. Miura *et al.*, *Appl. Phys. Lett.* **96**, 072506 (2010)

No changes in  $H_{c2}$  and  $\gamma$  (because  $\xi_0$  is very small)

$H_{irr}$  (melting line) still increases for fields as high as 60T !  
Largest increase at intermediate angles

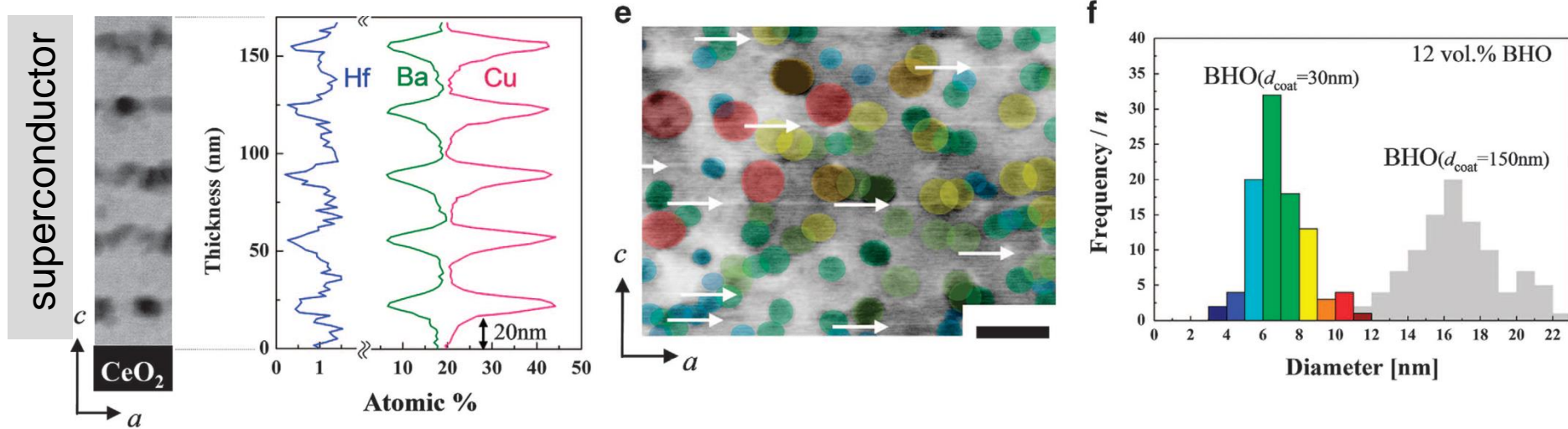
Record high  $H_{irr}$  no saturation with NP density: further increases possible

M. Miura *et al.*,  
*Sci. Rep.* **6**, 20436 (2015)





# We developed a multilayer deposition method enables introduction of even smaller random nanoparticles in YBCO-based CC grown by MOD



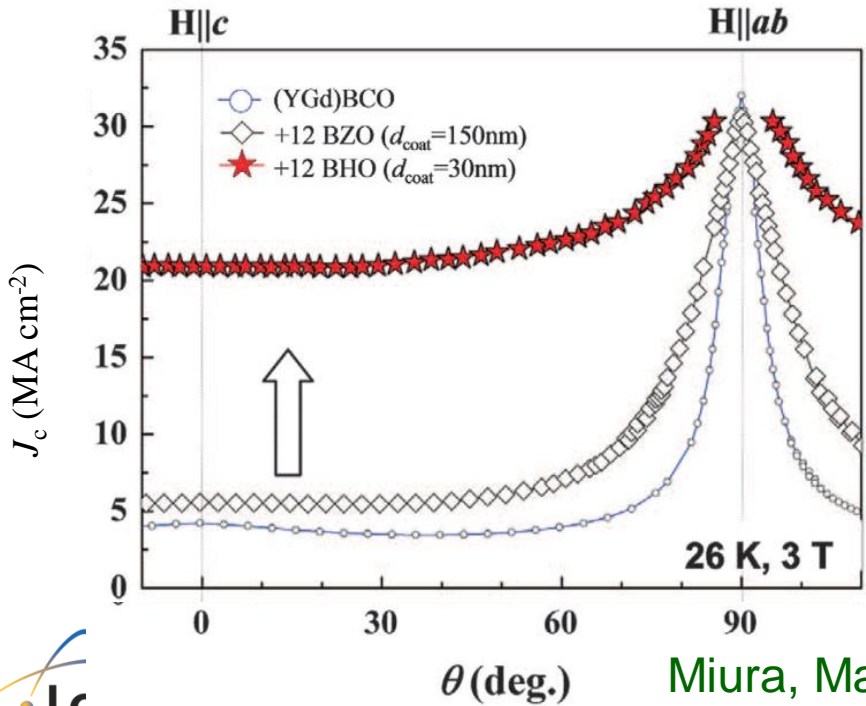
Matching size of nanoparticle to vortex size is essential for effective pinning

In previous studies nanoparticles in MOD were much larger than needed at low  $T$

Multilayer deposition creates Ba-poor regions that stop the growth of BaHfO<sub>3</sub> NPs

Particle size reduced x5

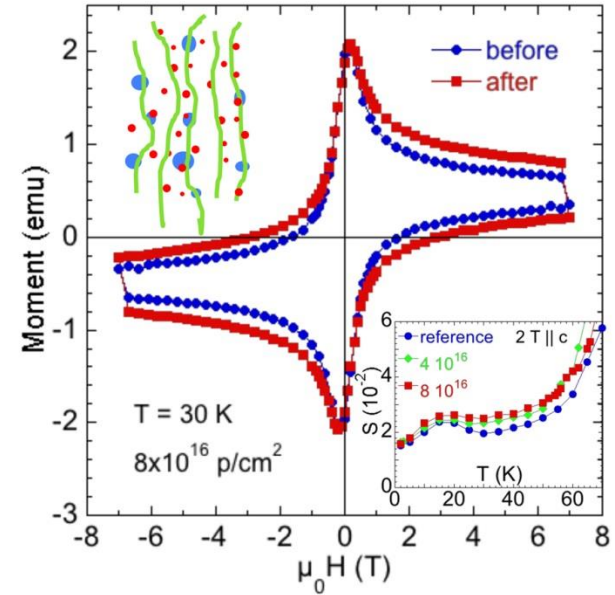
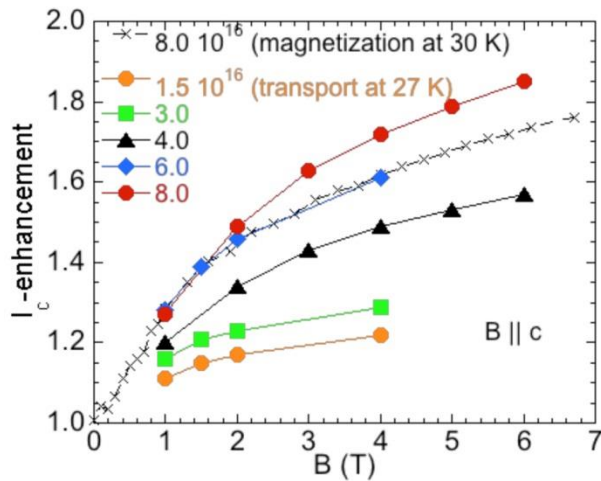
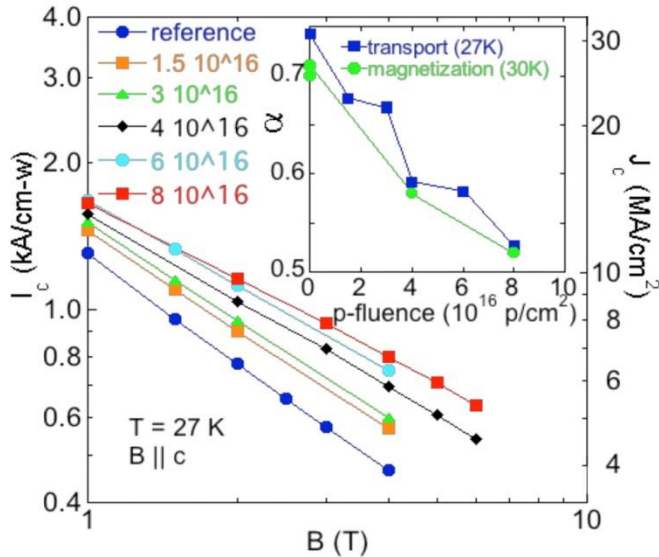
Pinning at low  $T$  is increased dramatically



Miura, Maiorov, *et al*  
 NPG Asia Materials **9**, e447 (2017)

# Pinning in commercial $\text{YBa}_2\text{Cu}_3\text{O}_7$ coated conductors can still be substantially enhanced by irradiation with 4 MeV protons.

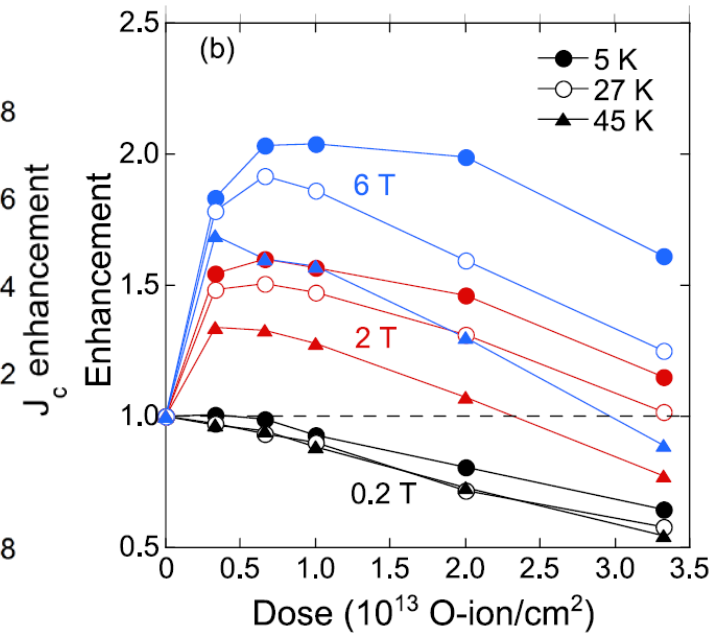
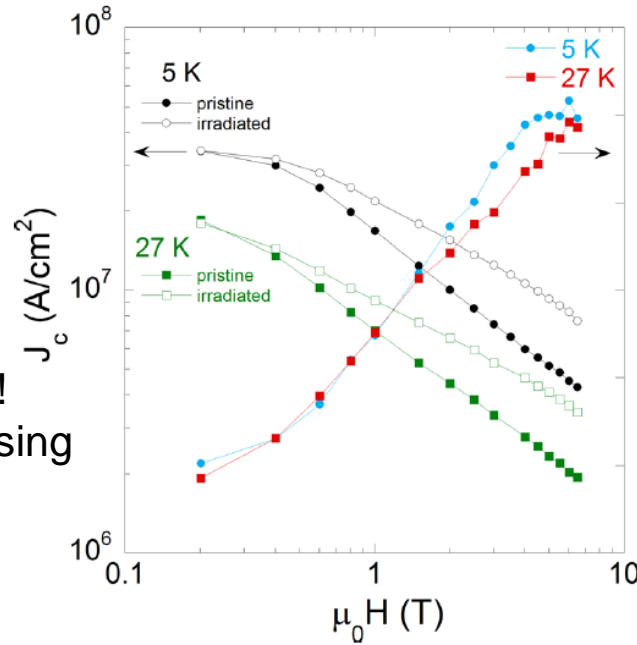
- CC from AMSC
- Irradiation: 4 MeV protons, fluence  $8 \times 10^{16}$  p/cm<sup>2</sup>
- Study lead by ANL - creep studies at LANL



- Near doubling of  $J_c$  in fields  $\sim 6$ T at  $\sim 27$  K
- A mixed pinning landscape of preexisting precipitates and twin boundaries and small, finely dispersed irradiation induced defects.
- No significant changes in creep rates.

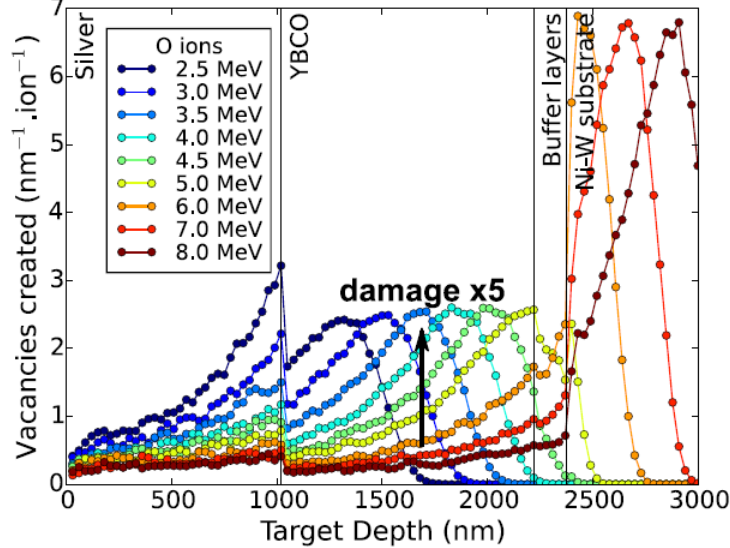
The same  $J_c$  enhancements (in the same coated conductors) can be obtained by oxygen irradiation, but with 1000 times smaller doses!

M. Leroux *et al.*,  
APL **107**, 192601 (2015)

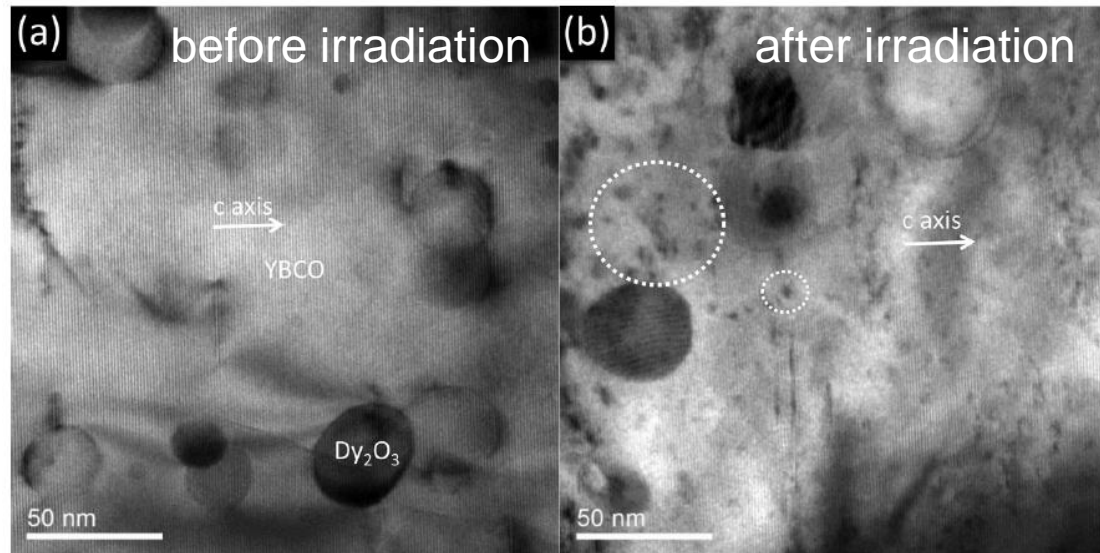


Irradiation done in 1 second!  
Enables commercial processing

SRIM-TRIM simulations



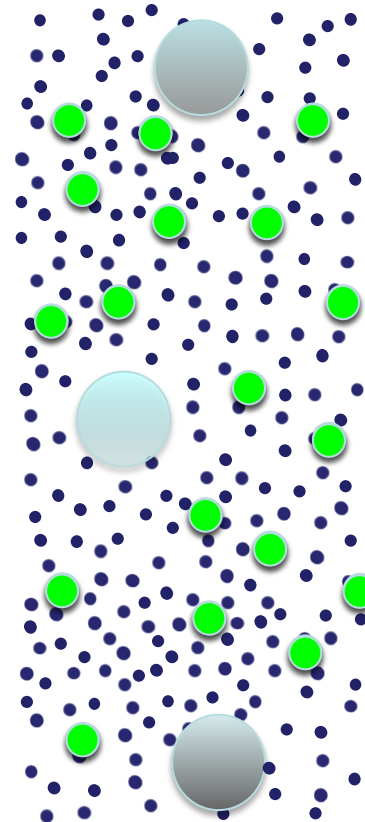
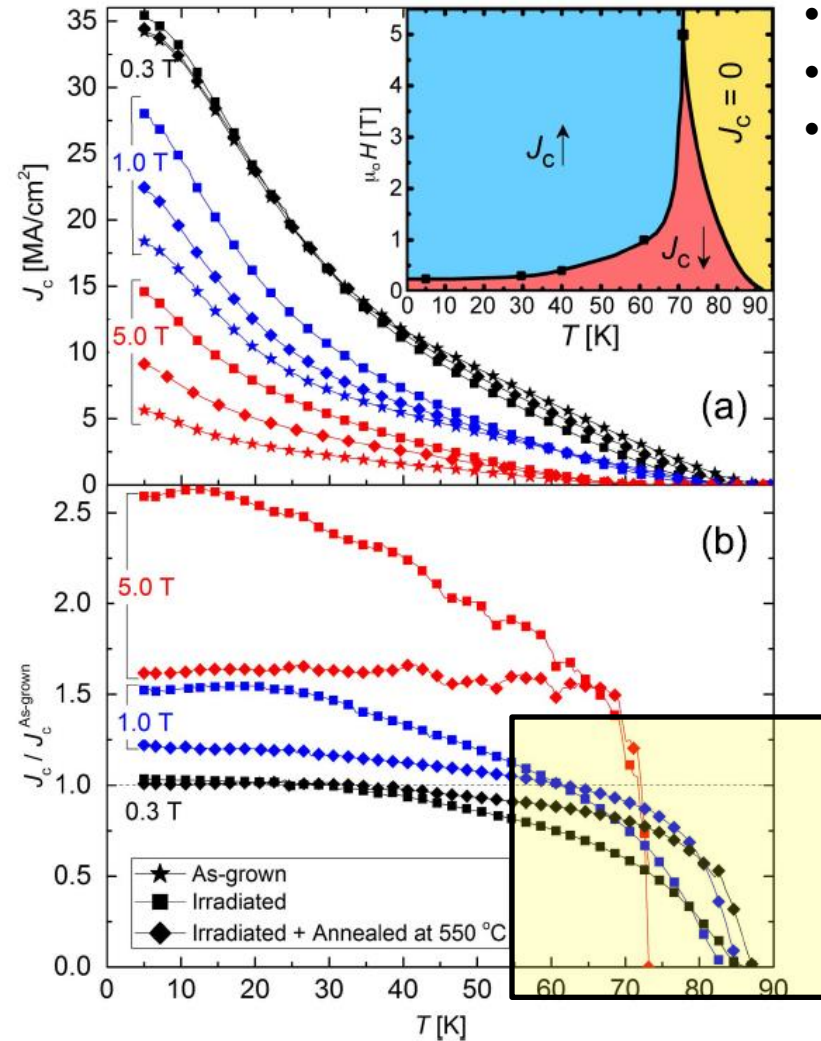
Irradiation introduces clusters and point defects



# Oxygen irradiation improves $J_c$ over most of the H-T phase diagram, but...

... reduces  $J_c$  at low H, high T

- Irradiation introduces clusters and point defects
- $J_c$  reduction  $\Rightarrow$  Evidence for competing effects
- Annealing (removing point defects) increases  $J_c$  !!



Mixed pinning landscapes exhibit complex cooperation and competition effects among different types of disorder

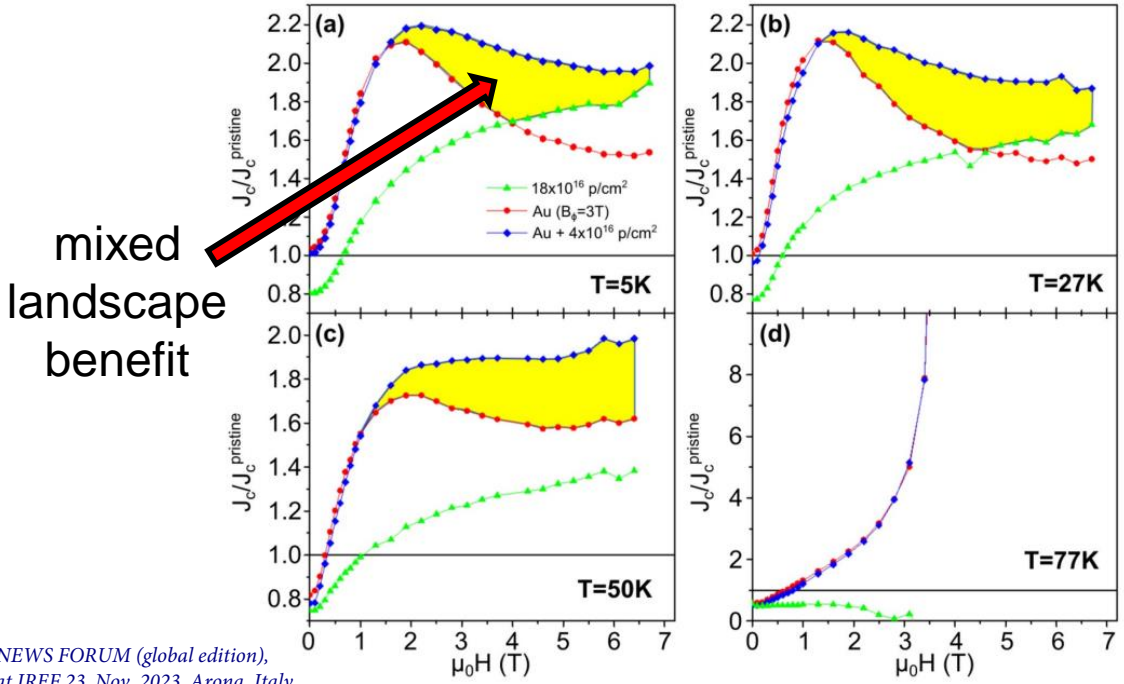
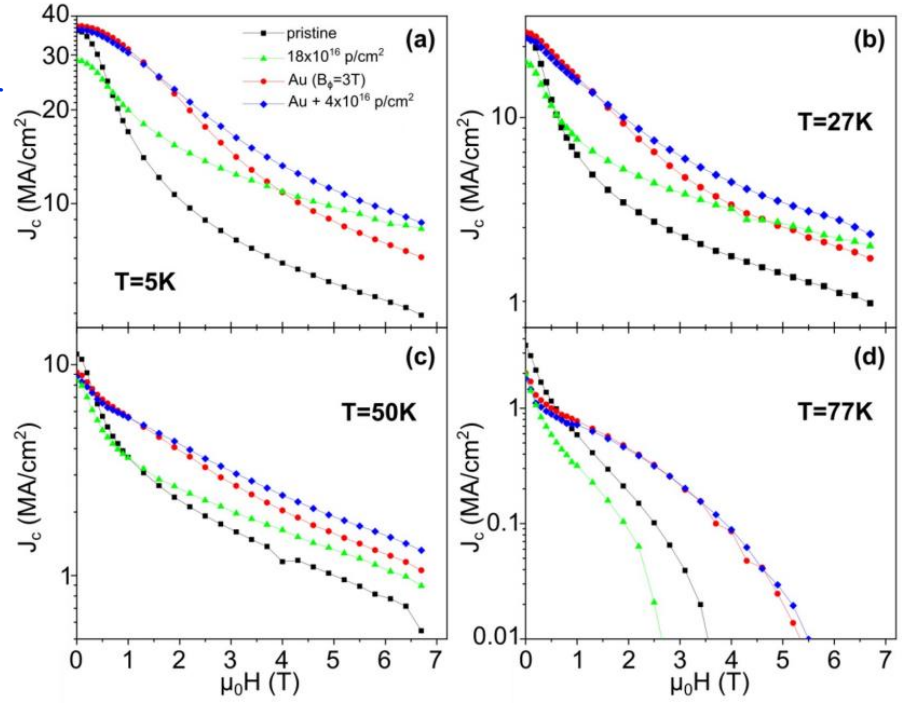
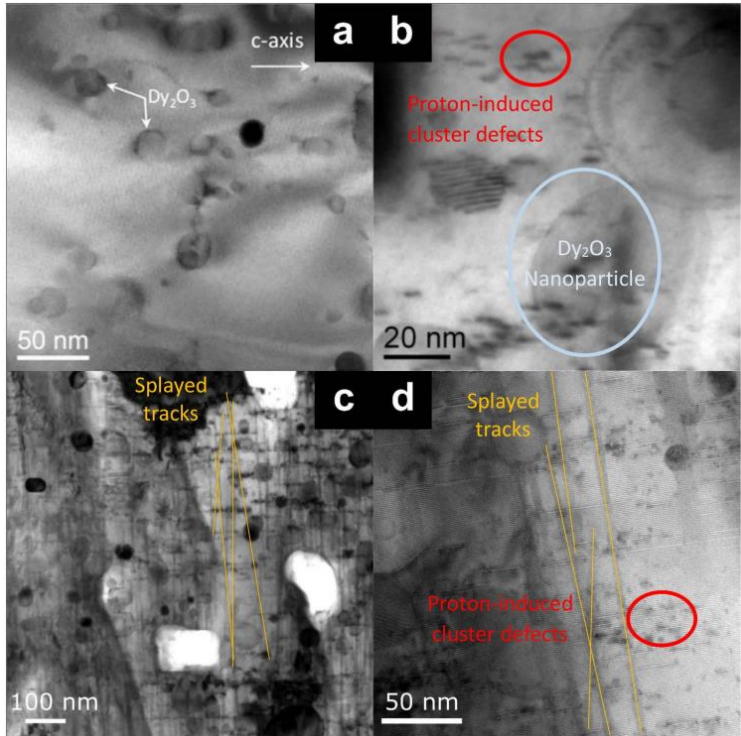
S. Eley, *et al.*, SuST **30**, 015010 (2017)  
 Collaboration with ANL (EFRC Center for Emergent Superconductivity)

# Confirmation of the benefits of mixed pinning landscapes by combined 4MeV p<sup>+</sup> and 250 MeV Au irradiations

K J Kihlstrom<sup>1,7</sup>, L Civale<sup>2</sup>, S Eley<sup>2,8</sup>, D J Miller<sup>1</sup>, U Welp<sup>1</sup>, W K Kwok<sup>1</sup>, P Niraula<sup>3</sup>, A Kayani<sup>3</sup>, G Ghigo<sup>4,5</sup>, F Laviano<sup>4,5</sup>, S Fleshler<sup>6</sup>, M Rupich<sup>6</sup> and M Leroux<sup>1,2,9</sup>

Supercond. Sci. Technol. **34** (2021) 015011 (13pp)

Pristine                      Protons

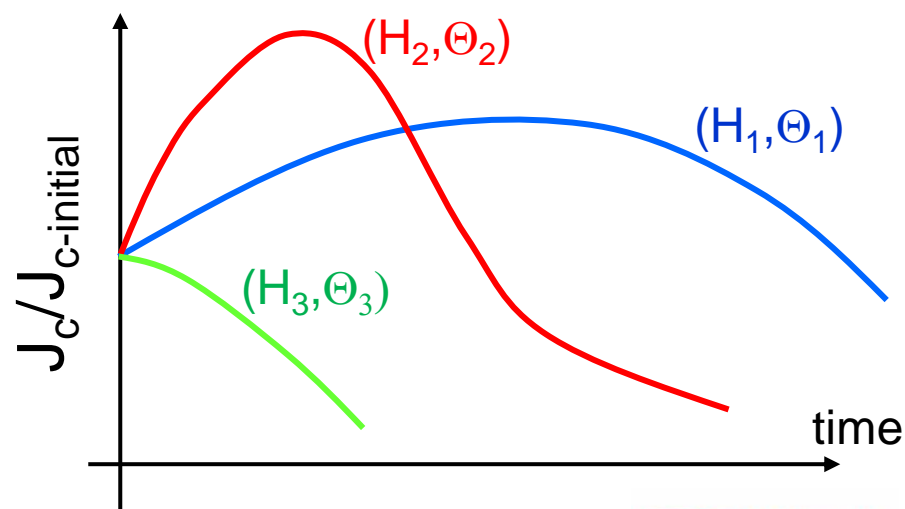


Combined irradiations

## Concluding considerations

- Disorder (material defects) is required for vortex pinning
- *ReBCO*-based CCs have the highest  $J_c$  in any known SC  $\Rightarrow$  many effective strong pinning defects
- Added defects in CCs (e.g. second phases) are wisely optimized for high  $J_c$  – typically “mixed pinning landscapes”
- Irradiation-induced defects will interact with pre-existing disorder. There will be cooperation and competition effects, different for each CC (“initial conditions”)
- Irradiation will start modifying the properties of the CC magnets in fusion reactors from day one of operation - The initial effect on  $J_c$  may be *mostly* positive...

• ...but  $J_c$  will evolve differently for each  $(H, \Theta)$  condition (not just an overall factor)  $\Rightarrow$  the location of the limiting position in the magnet will change.



*Thank you for your attention!*