

# Equal-spin supercurrents and magnetization dynamics in high-temperature superconductor/ferromagnet hybrids

Javier E. Villegas

Unité Mixte de Physique CNRS/Thales, France

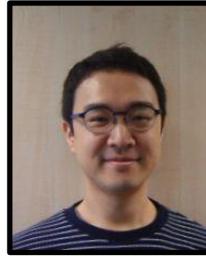


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# Acknowledgements

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**D. Sanchez, S. J. Carreira, S. Mesoraca, M.-W. Yoo, V. Rouco, X. Palermo,  
A. Balan, K. Seurre, A. Sander, A. Anane & Javier E. Villegas**  
Unité Mixte de Physique CNRS, Thales, **FRANCE**

**G. Orfila, A. Cuellar, M. Cabero, L. Marcano, M. Rocci, J. Garcia-Barriocanal  
F. Gallego, J. Tornos, A. Rivera, J. M. Gonzalez-Calbet, C. Leon & J. Santamaria**  
Universidad Complutense Madrid, **SPAIN**

**S. Valencia**  
Helmholtz-Zentrum Berlin, **GERMANY**

**F. Mompean, M. Garcia-Hernandez**  
ICMM-CSIC, **SPAIN**

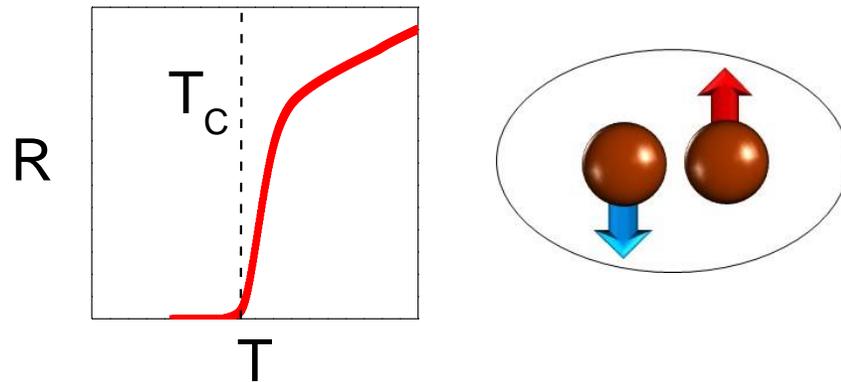
**C. Feuillet-Palma, N. Bergeal & J. Lesueur**  
ESPCI-Paris, **FRANCE**

**A. I. Buzdin**  
Université de Bordeaux, **FRANCE**



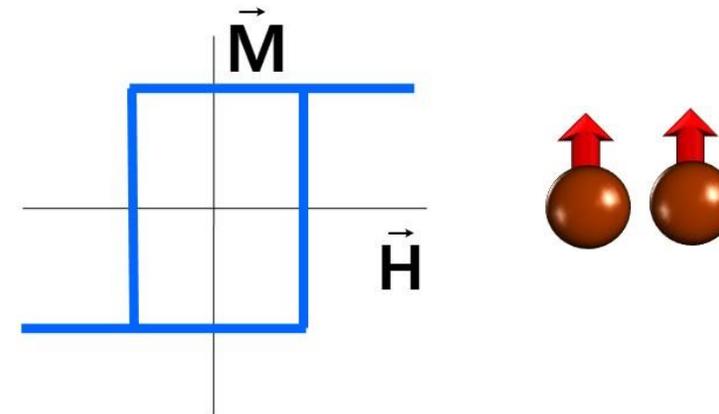
# Superconductor/ferromagnet hybrids

Superconductor



Large resistance variation  
Coherent transport

Ferromagnet



Memory  
Spin-polarized transport

→ OPPORTUNITIES FOR SPINTRONICS

# Main goals in the area

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## Spin-polarized supercurrents for spintronics

Matthias Eschrig *Physics Today*, 64 43 (2011)

A marriage between superconductivity and ferromagnetism is opening the door for new spin-based applications.

## Mesoscopic magnetism and superconductivity

Ali C. Basaran, Javier E. Villegas, J.S. Jiang, Axel Hoffmann, and Ivan K. Schuller



REVIEW ARTICLES

PUBLISHED ONLINE: 2 APRIL 2015 | DOI: 10.1038/NPHYS3242

## Superconducting spintronics

Jacob Linder<sup>1\*</sup> and Jason W. A. Robinson<sup>2\*</sup>



Volume 40, Issue 11 (Mesoscale Materials, Phenomena, and Functionality)

November 2015, pp. 925-932

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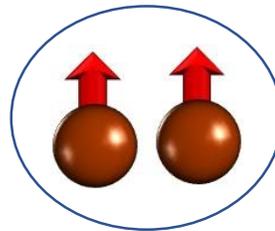


Volume 40, Issue 11 (Mesoscale Materials, Phenomena, and Functionality)

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## A) SPIN-POLARIZED SUPERCURRENTS

- Equilibrium
- Low-dissipation spin transport
- Spin torques
- Phase coherence effects (Josephson)
- Large spin signals (spin valves)



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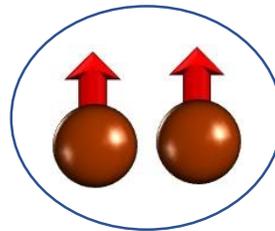


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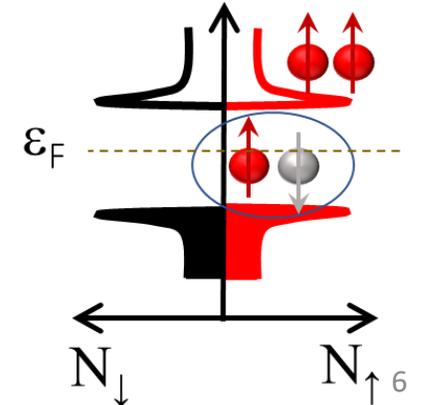
### A) SPIN-POLARIZED SUPERCURRENTS

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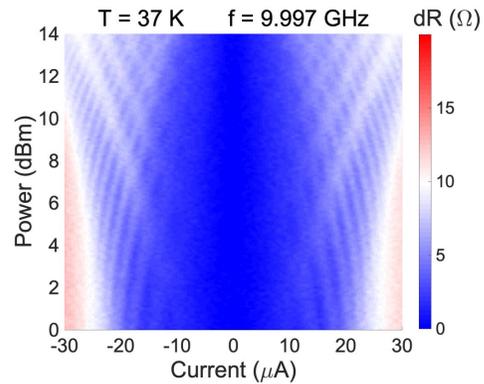
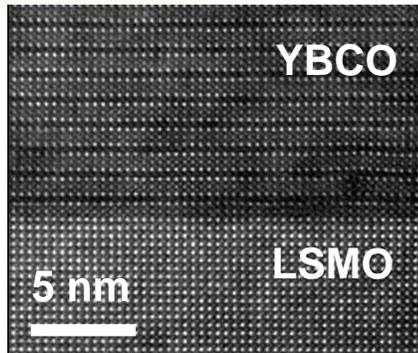
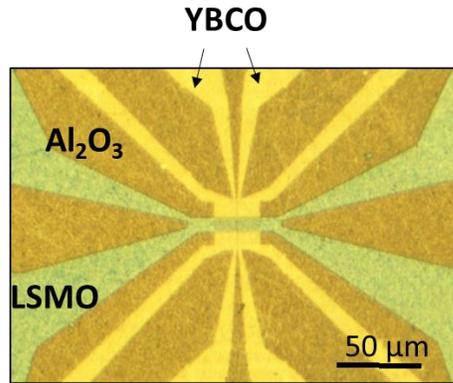
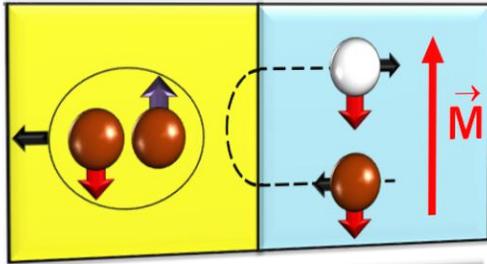
### B) SPIN-POLARIZED QUASIPARTICLES

- Non-equilibrium
- Quantum protection
- Enhanced spin lifetimes
- Enhanced spin Hall effects
- Dynamic coupling



# The talk in a nutshell

## Proximity effect

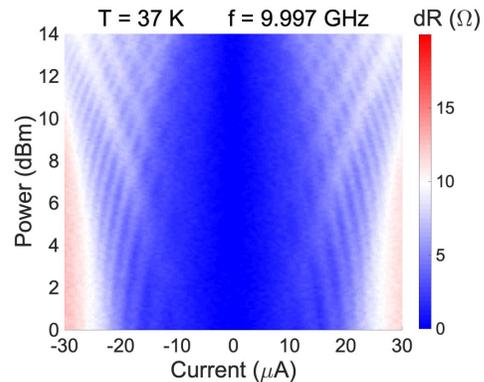
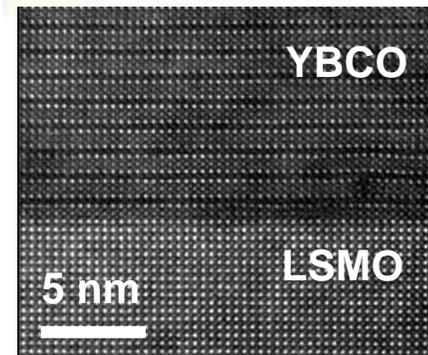
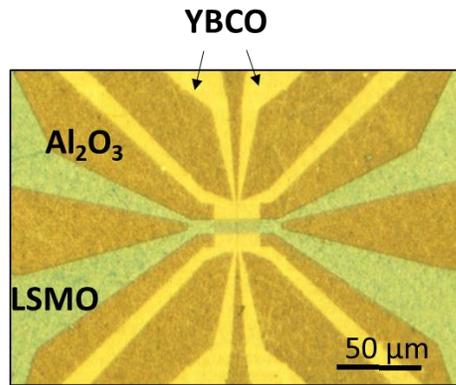
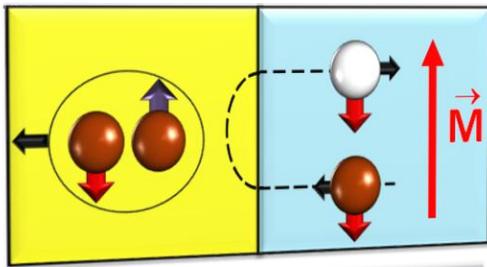


**Fully spin-polarized, high- $T_c$  Josephson supercurrents**  
**Quantum phase coherence effects**

**Sanchez-Manzano *et al.* Nature Materials (2022)**

# The talk in a nutshell

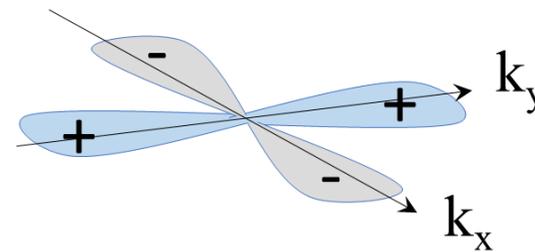
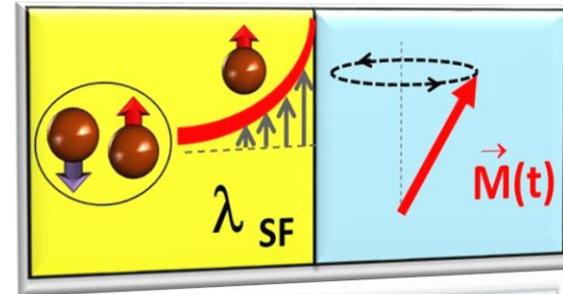
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Fully spin-polarized, high- $T_c$  Josephson supercurrents  
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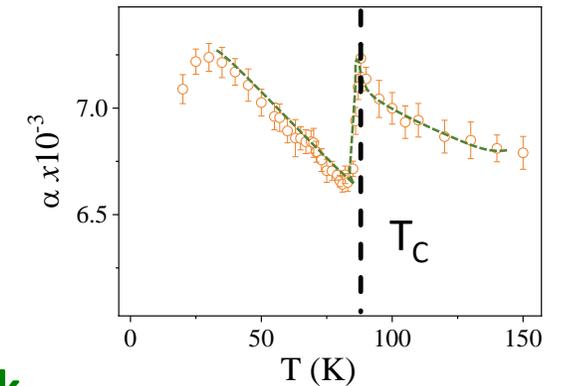
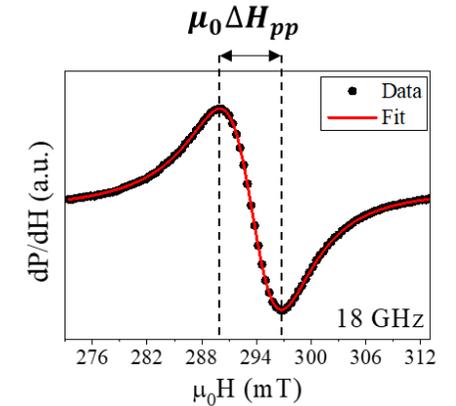
Sanchez-Manzano *et al.* Nature Materials (2022)

## Dynamic coupling



Tunable spin sink  
d-wave effects

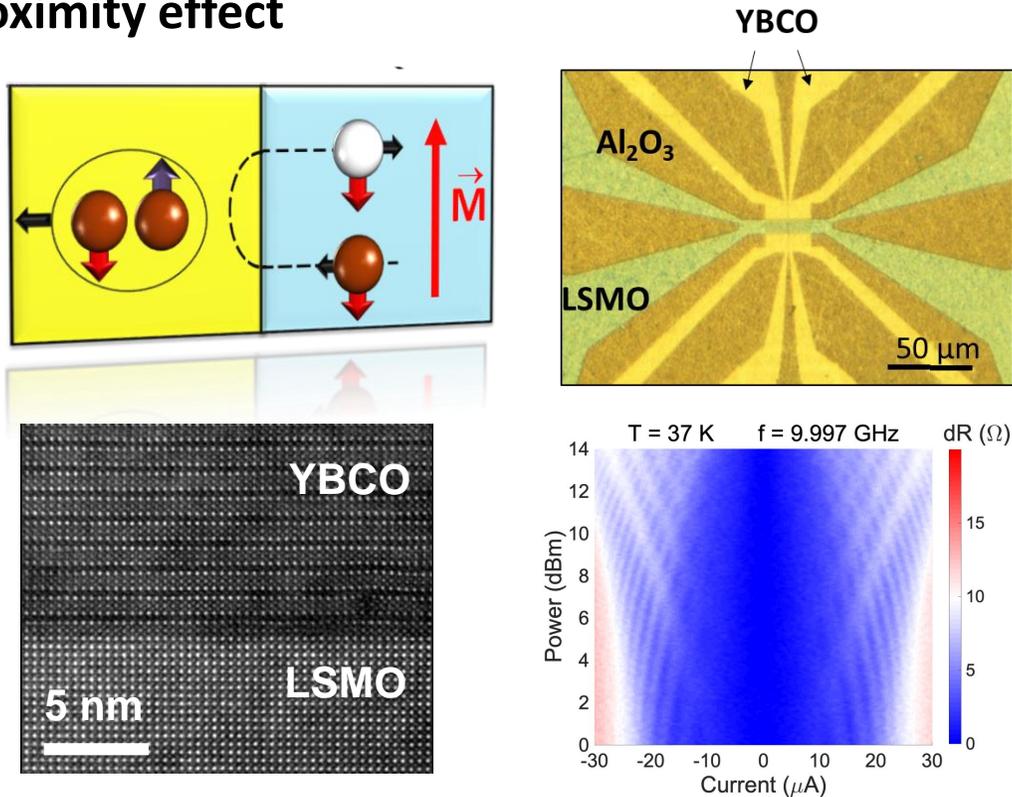
Carreira *et al.* PRB (2021)



→ OPPORTUNITIES FOR HIGH- $T_c$  SUPERCONDUCTING SPINTRONICS 8

# The talk in a nutshell

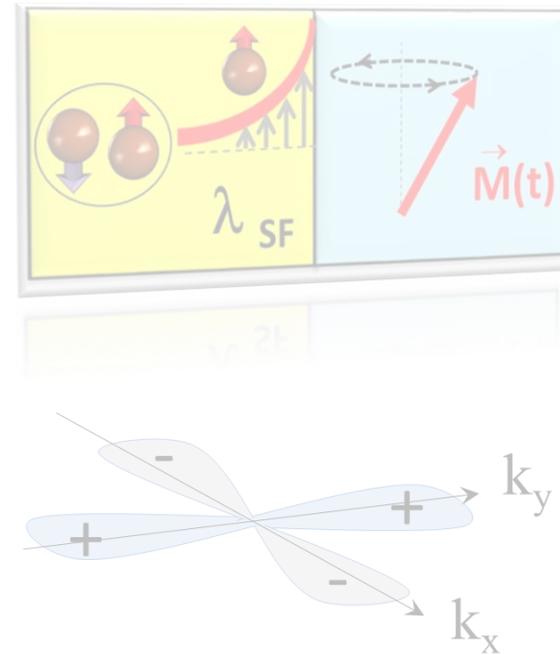
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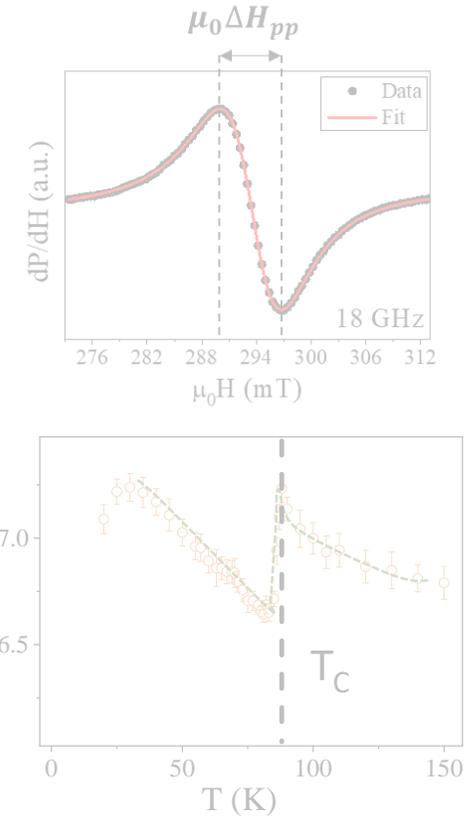
Sanchez-Manzano *et al.* Nature Materials (2022)

## Dynamic coupling



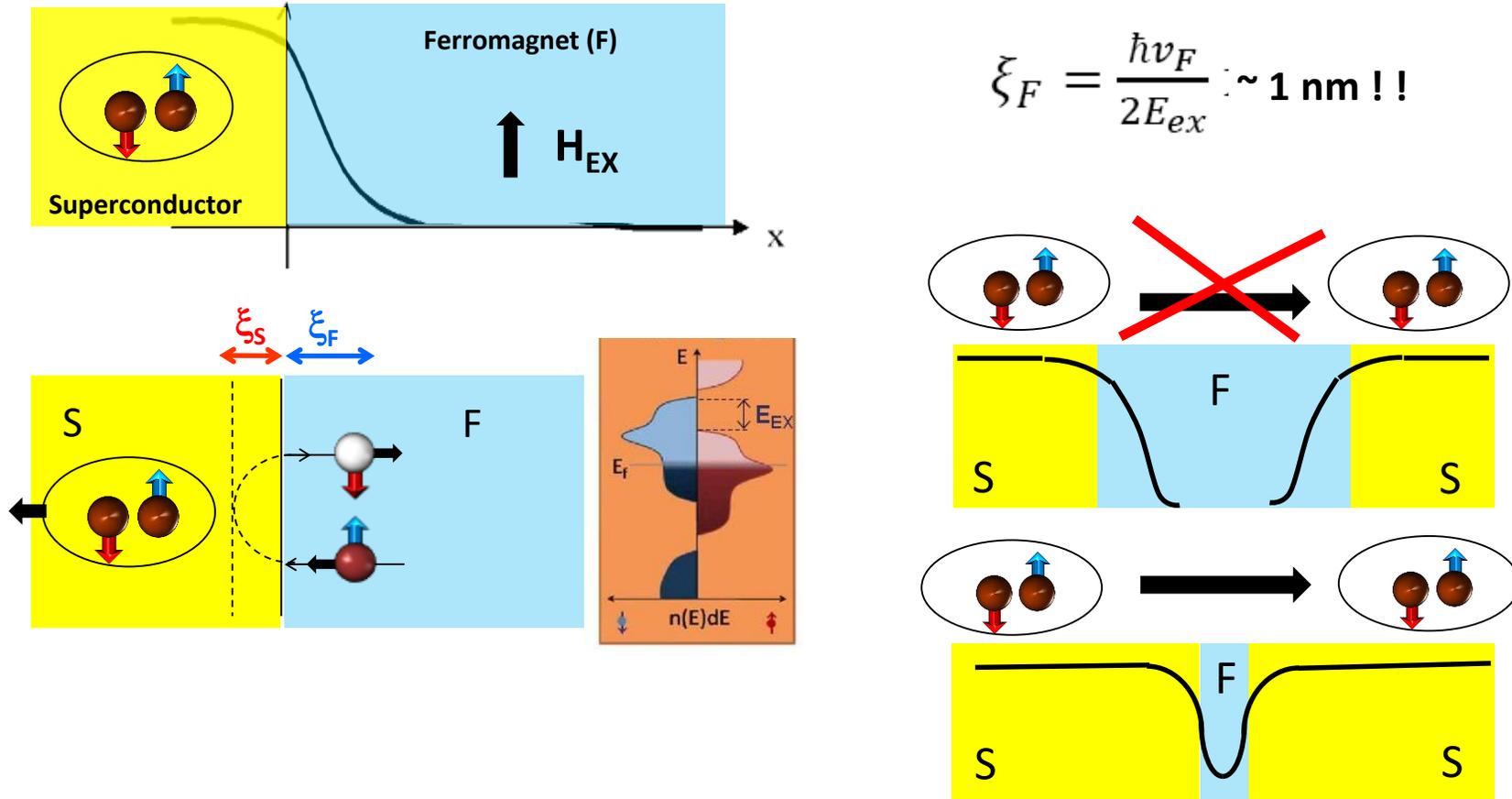
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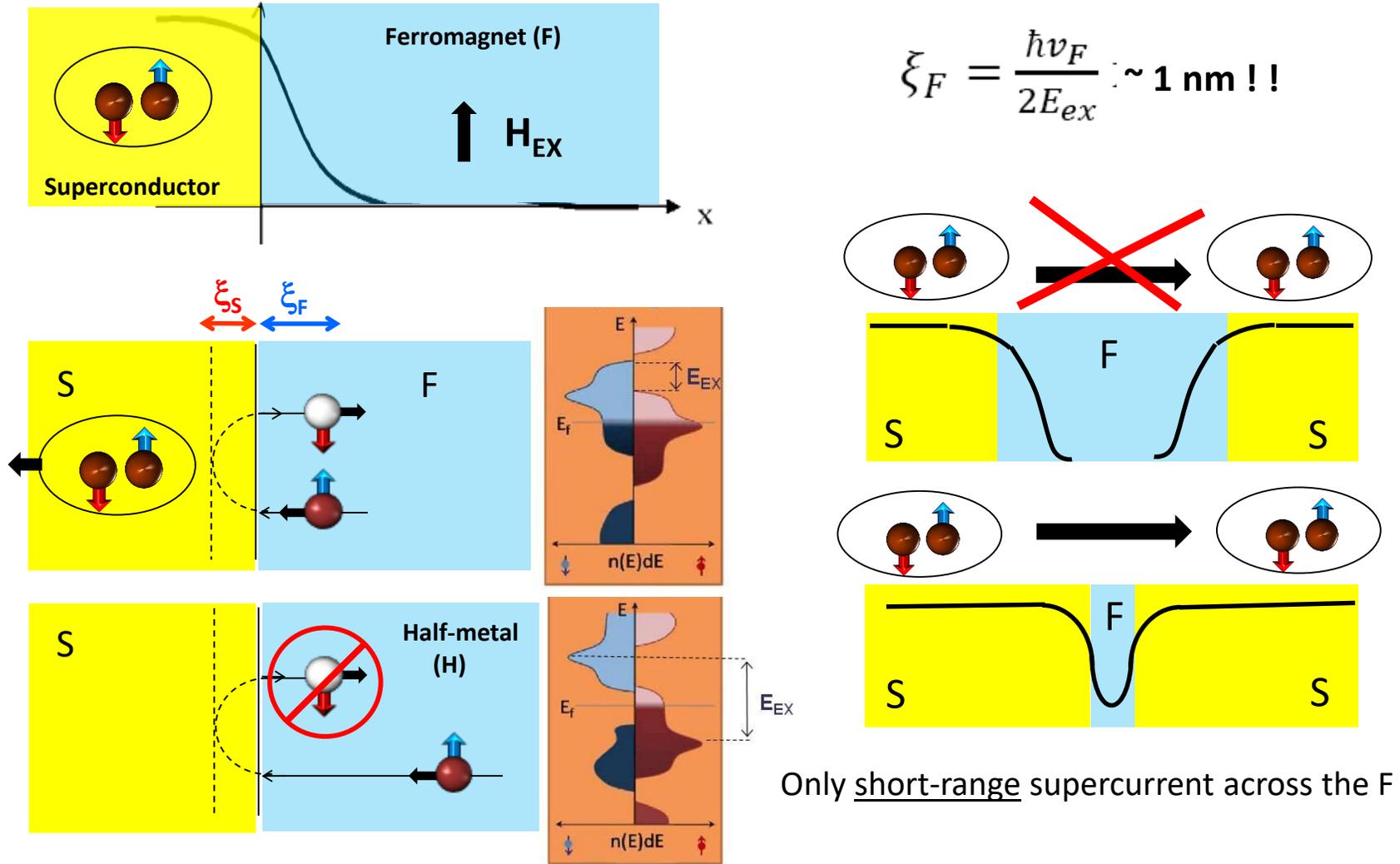


→ OPPORTUNITIES FOR HIGH- $T_c$  SUPERCONDUCTING SPINTRONICS 9

# Conventional proximity effect in ferromagnets

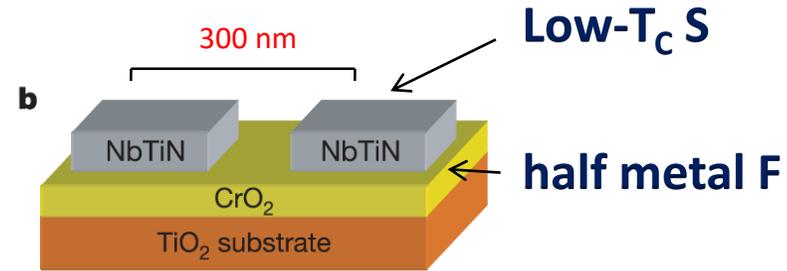
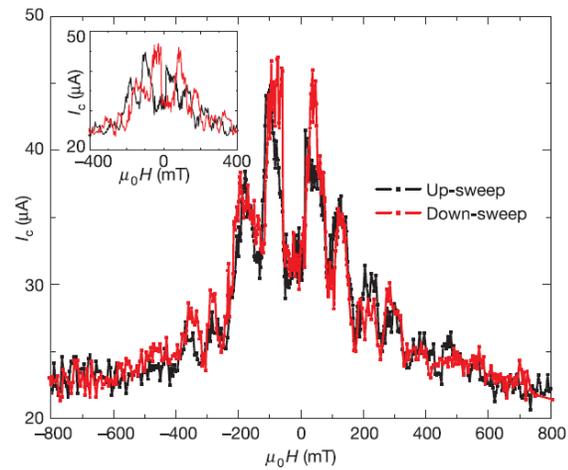


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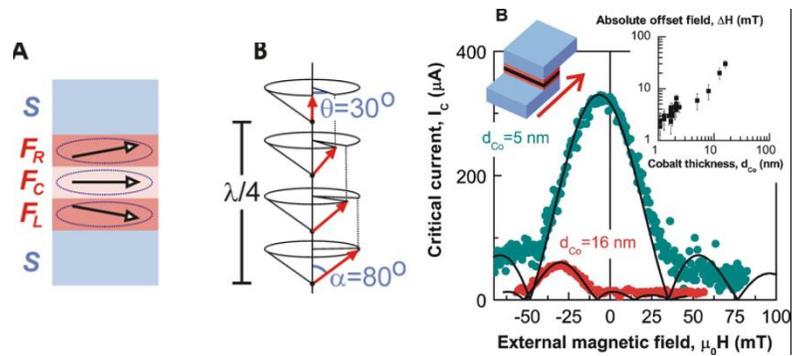


→ NO SINGLET SUPERCURRENTS ACROSS HALF-METALS

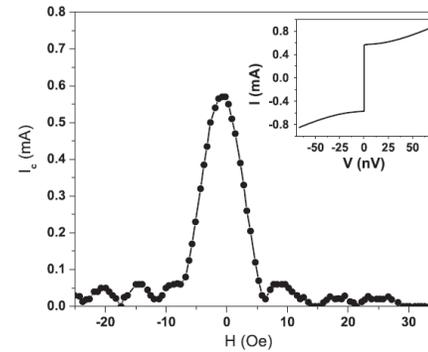
# Early experiments with low- $T_c$ superconductors



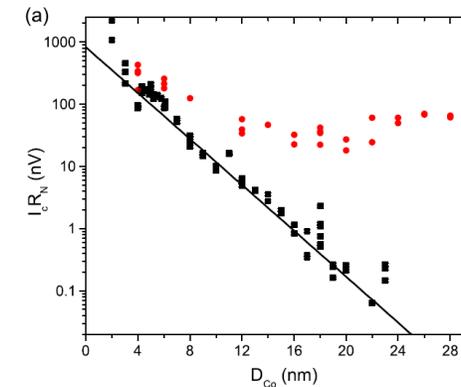
R. S. Keizer et al. Nature 439 (2006)



J. W. A. Robinson *et al*, Science (2010)



T. Khaire et al, PRL 104, 137002 (2010)

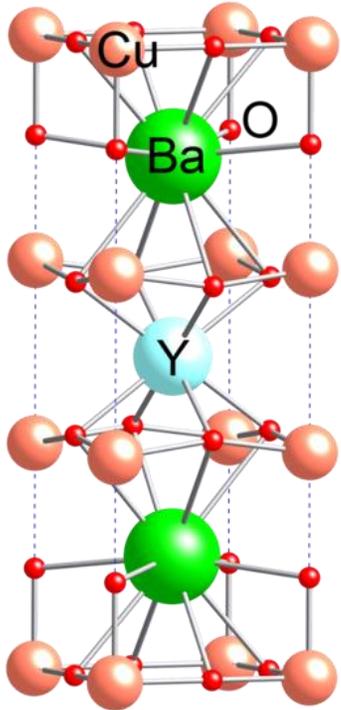


See also: Anwar et al. PRB (2010) & Sprungmann et al. PRB (2010)

→ LONG-RANGE JOSEPHSON COUPLING ACROSS STRONG F

# Aim: the case of high- $T_C$ superconductors

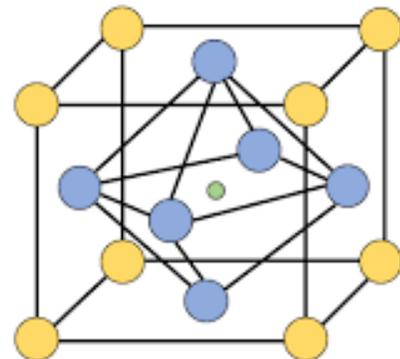
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Superconductor: YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> (YBCO)

- d-wave
- high- $T_C$

Ferromagnets: La<sub>0,7</sub>Sr<sub>0,3</sub>MnO<sub>3</sub> (LSMO) or La<sub>0,7</sub>Ca<sub>0,3</sub>MnO<sub>3</sub>

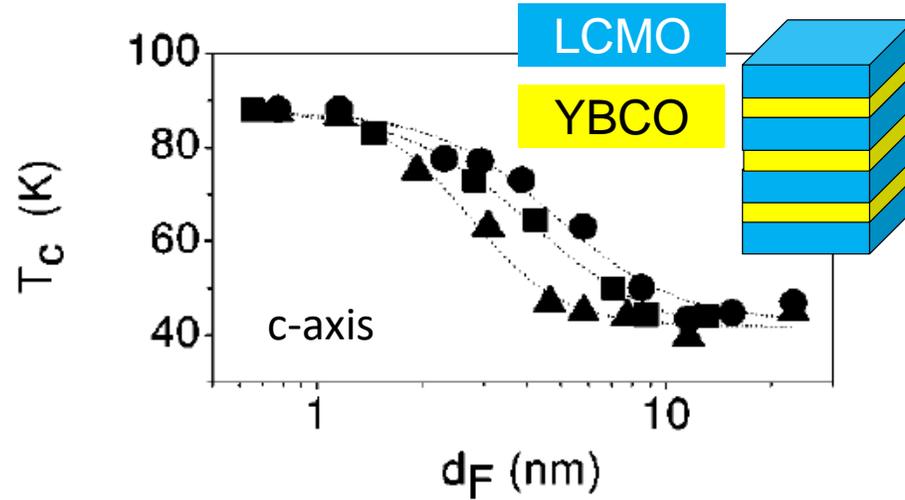


- La<sup>3+</sup>/ Sr<sup>2+</sup>
- O<sup>2-</sup>
- Mn<sup>3+</sup>/ Mn<sup>4+</sup>

- Half-metal  
(100% spin-polarized)

# Long-range proximity effect in $\text{YBa}_2\text{Cu}_3\text{O}_7/\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$

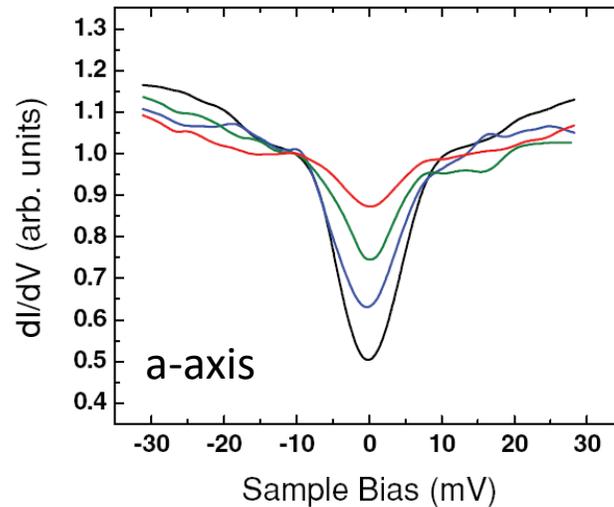
high-temperature superconductor / half-metal ferromagnet



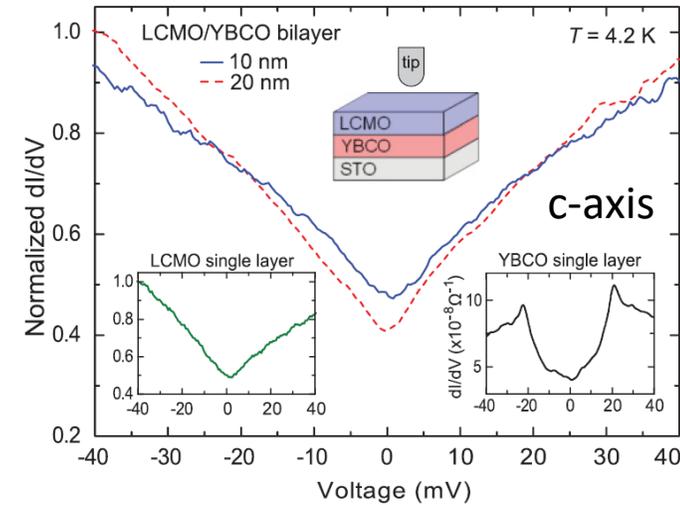
Sefrioui, JEV *et al.* PRB 67, 214511 (2003)

Peña *et al.* PRB 69, 224502 (2004)

$T_c$  DEPRESSION  $\rightarrow$  PROXIMITY EFFECT



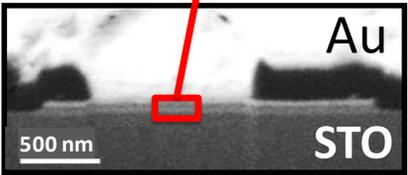
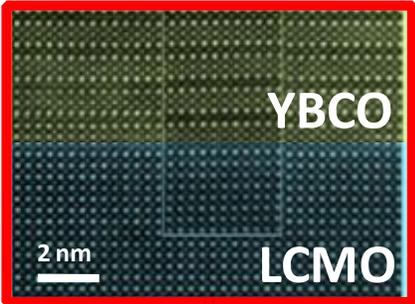
Kalcheim *et al.* Phys. Rev. B 83, 064510 (2011)



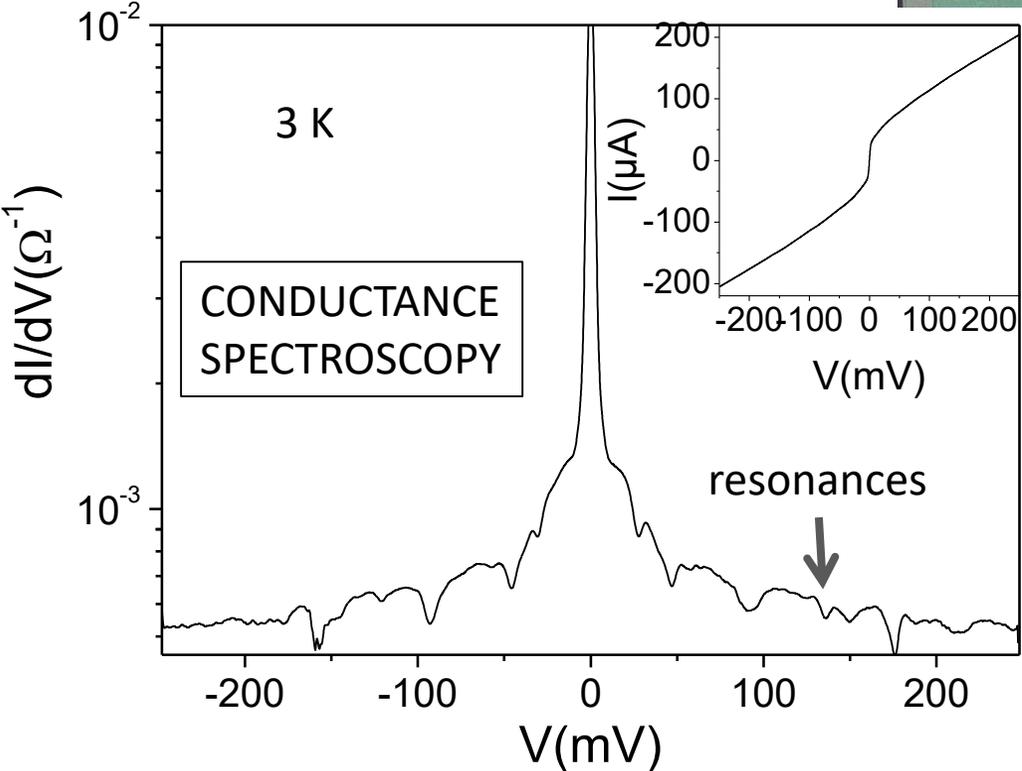
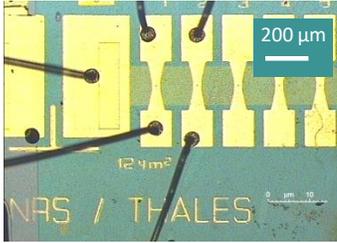
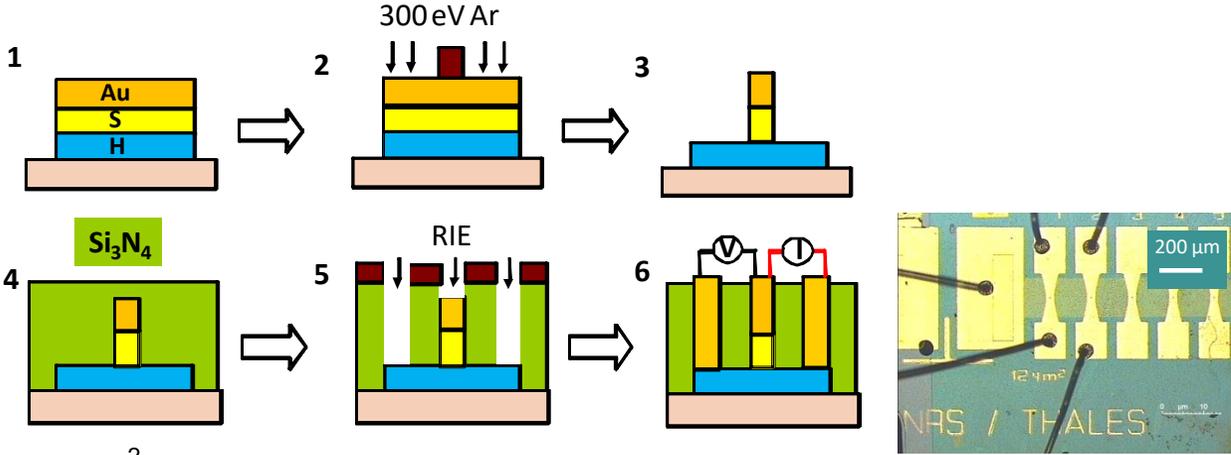
Fridman *et al.* Phys. Rev. B 84, 104522 (2011)

# High- $T_C$ /half-metal junctions

$\text{YBa}_2\text{Cu}_3\text{O}_7$   
 $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$

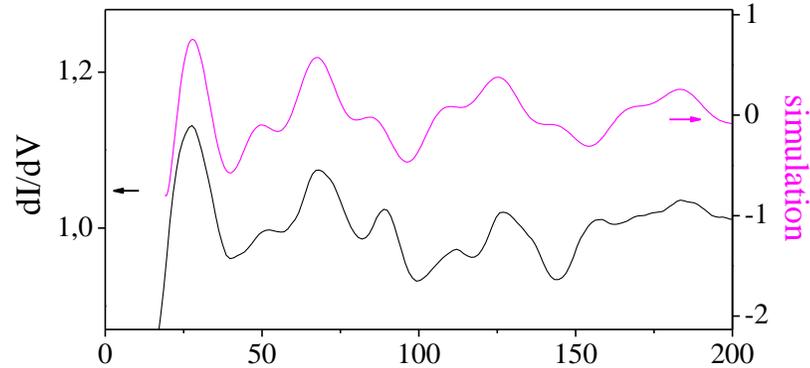


grown by  
 Santamaria's group  
 (UCM Spain)

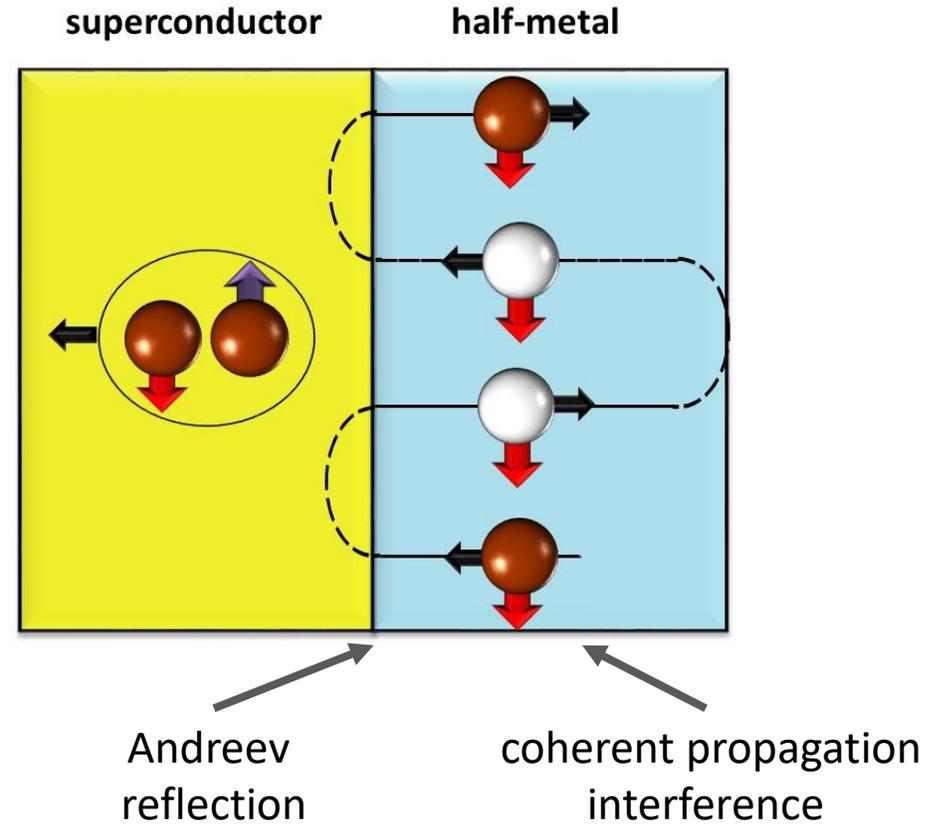


# Equal-spin Andreev reflection

Tomasch & McMillan-Rowell resonances



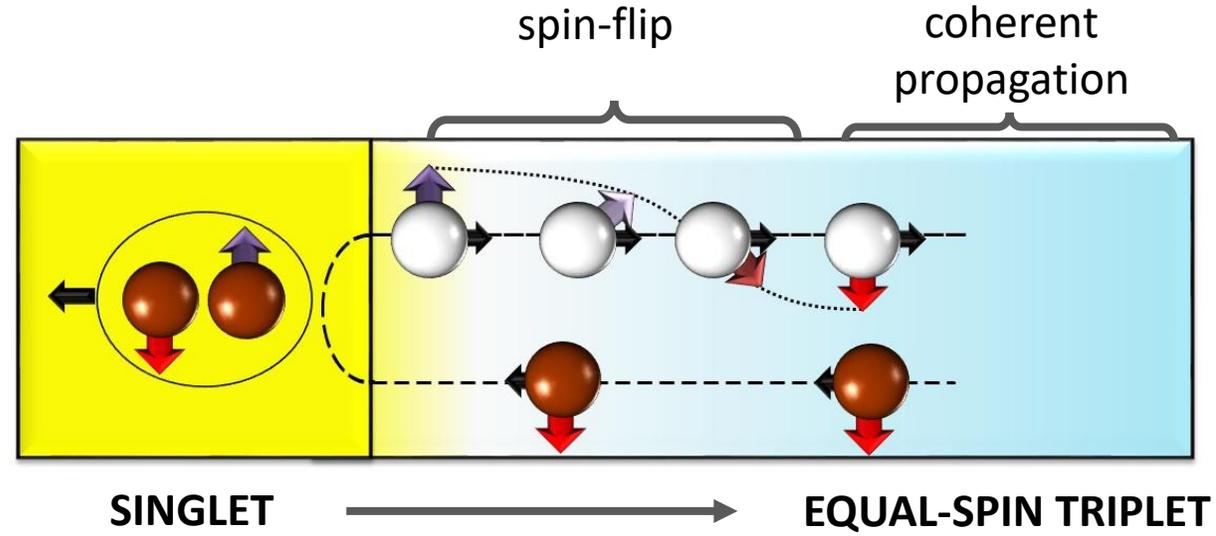
$$V_m = V_0 + \frac{m\hbar v_F^N}{4d_N}$$



→ ANDREEV REFLECTION

→ PHASE COHERENT, SPIN-POLARIZED CHARGE TRANSPORT

# Equal-spin triplets: physical origin



**ARTICLES** *Nat. Phys.* **4**, 138-143 (2008).

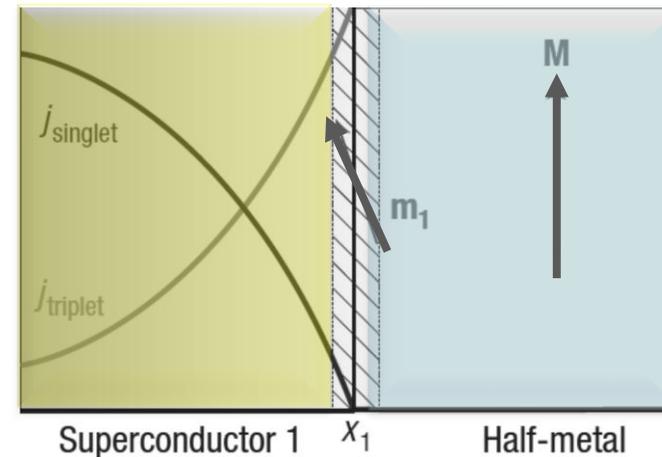
Triplet supercurrents in clean and disordered half-metallic ferromagnets

MATTHIAS ESCHRIG\* AND TOMAS LÖFWANDER†

Bergeret et al. *Rev. Mod. Phys.* **77**, 1321-1373 (2005)

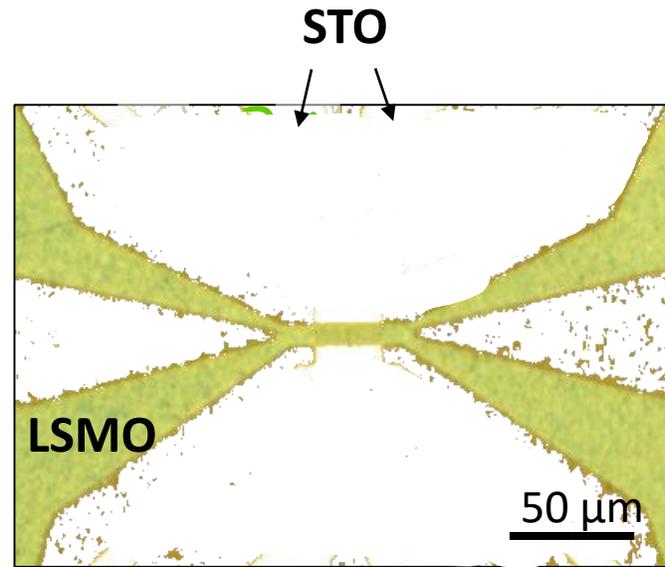
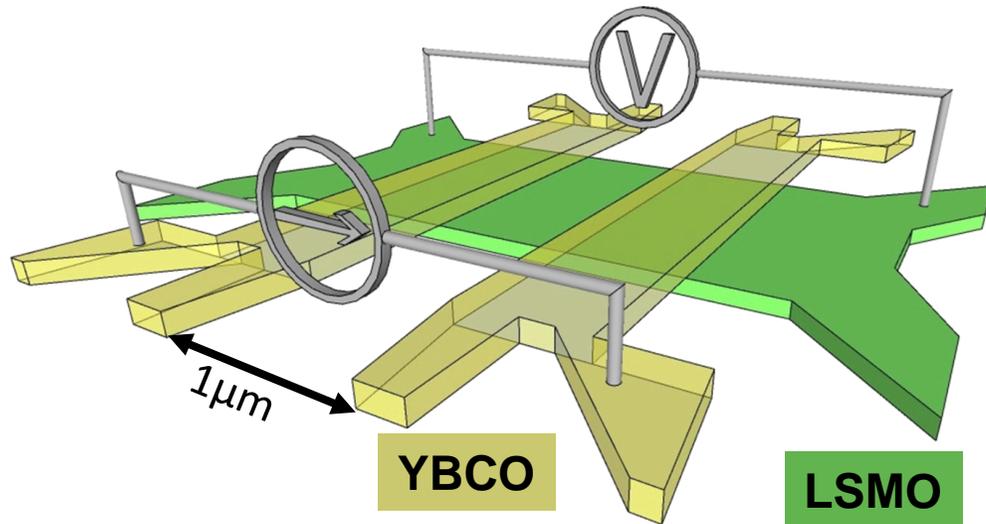
Buzdin et al. *Phys. Rev. B* **76** 060504 (2007)

Linder et al. *Phys. Rev. B* **81**, 174526 (2010)



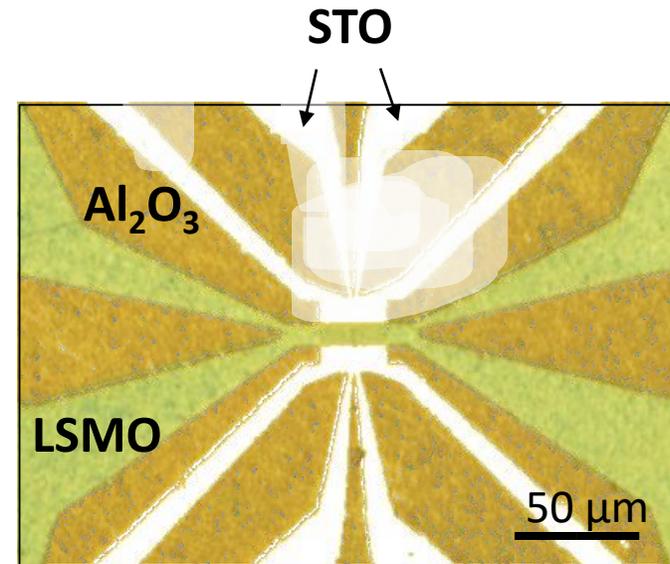
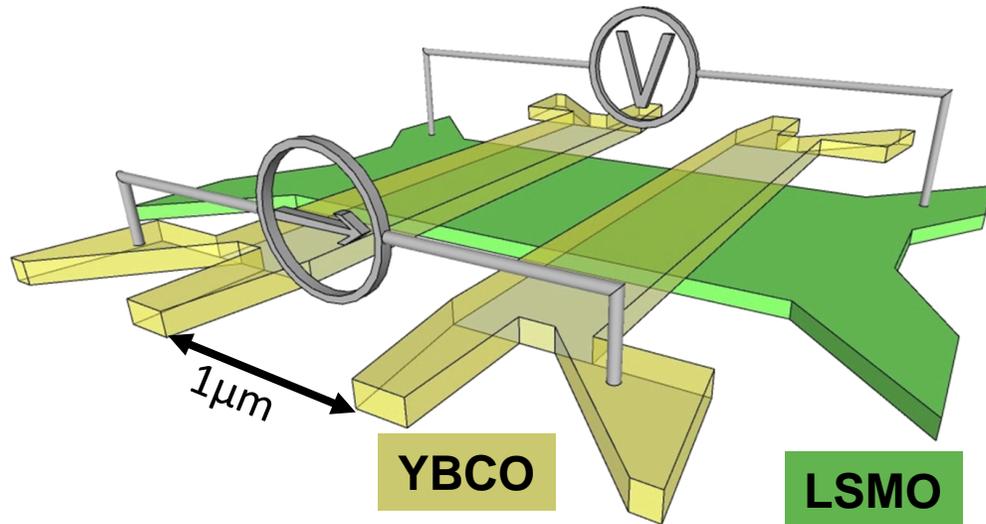
→ **MAGNETIC INHOMOGENEITY YIELDS SPIN-FLIP**

# Fabrication of planar Josephson junctions



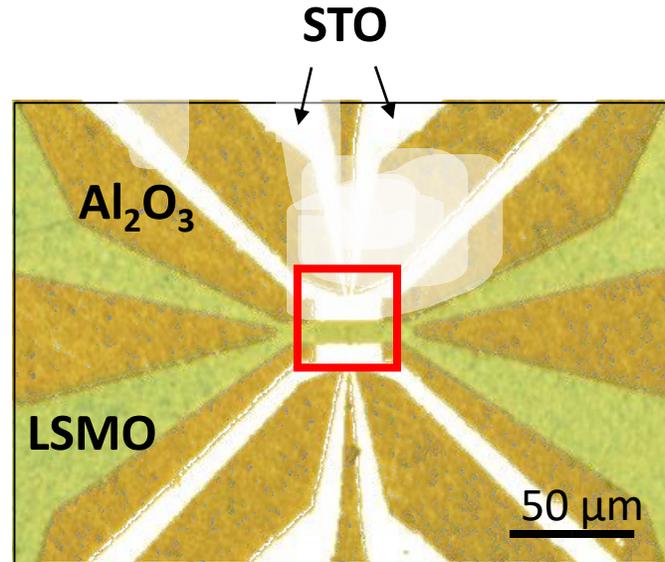
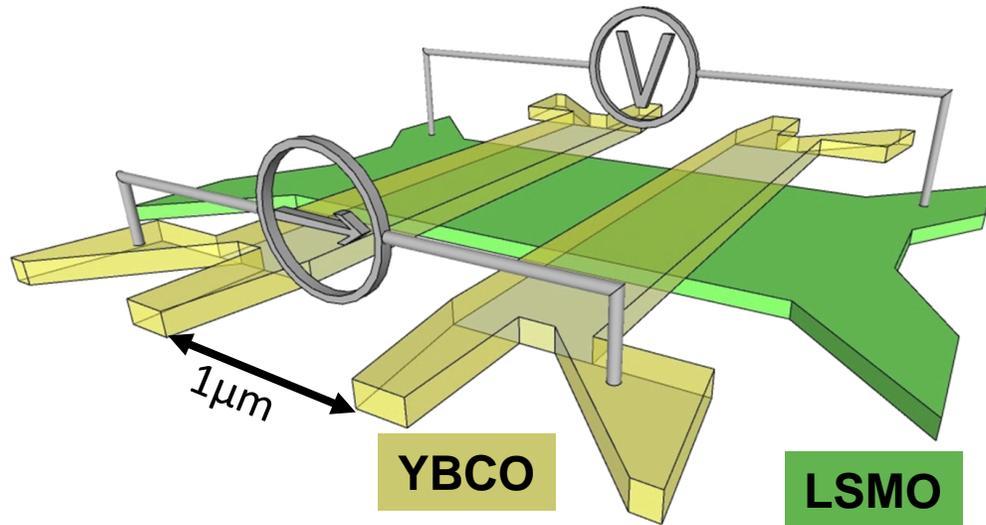
→ PLD GROWTH OF LSMO AND LITHOGRAPHY

# Fabrication of planar Josephson junctions

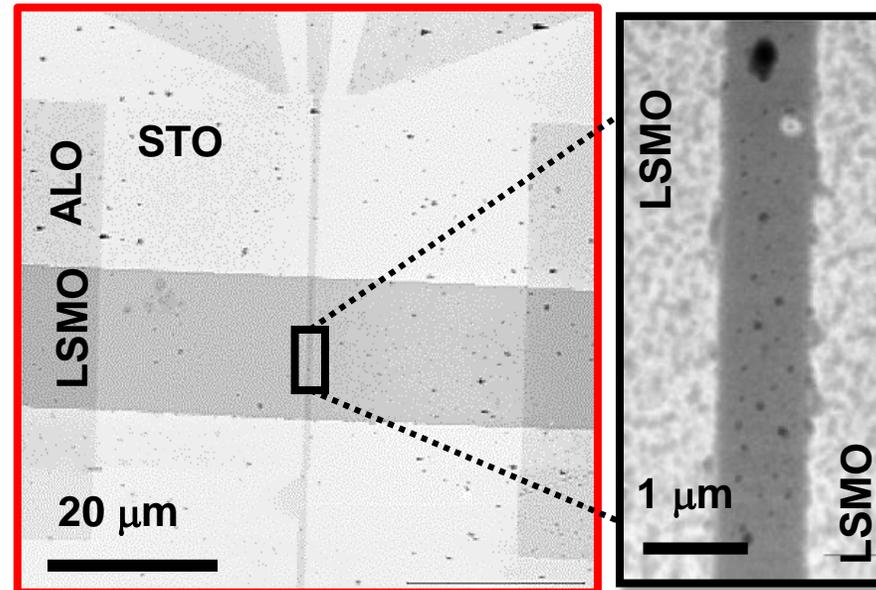


→ E-BEAM LITHOGRAPHY  
→ DEPOSITION OF AMORPHOUS ALUMINA

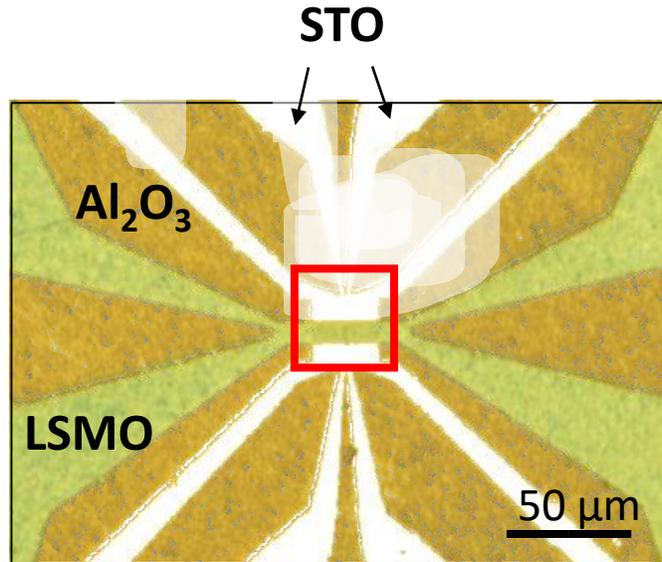
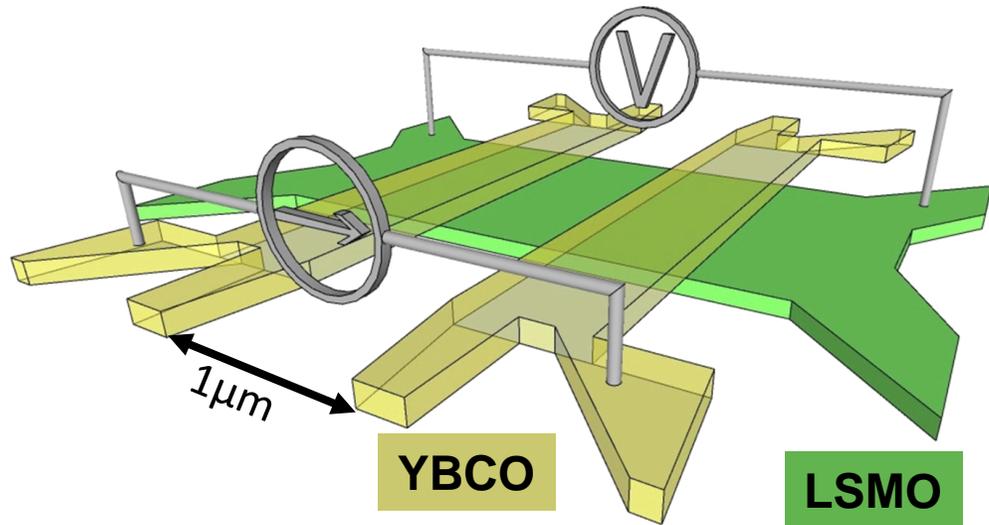
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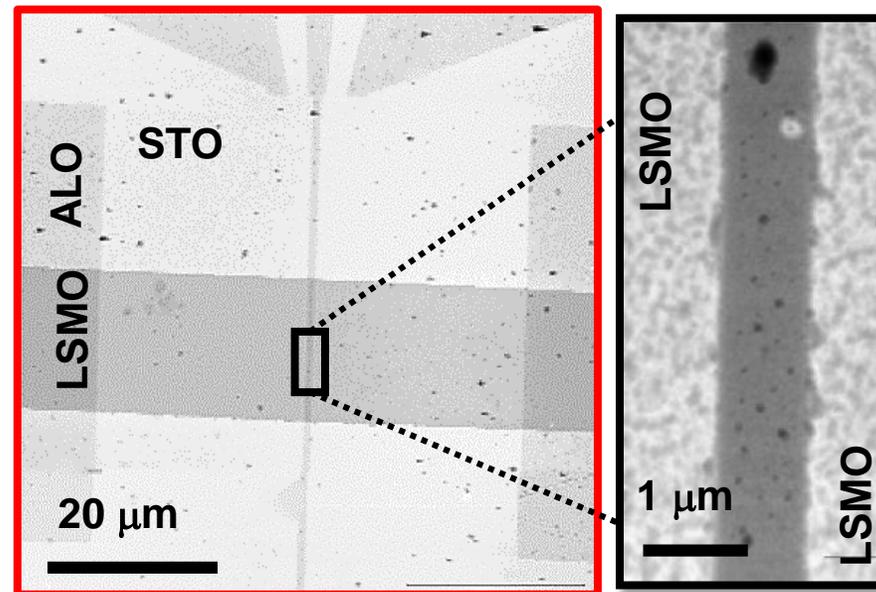
→ AMORPHOUS ALUMINA MASK



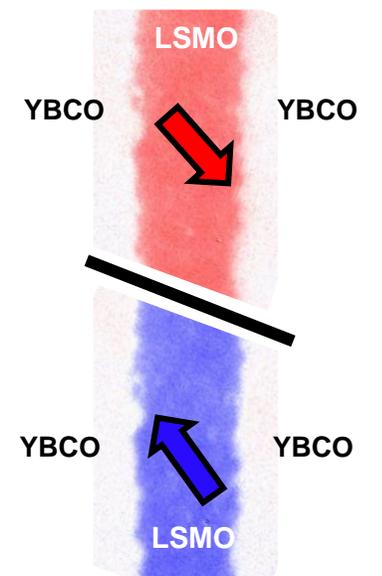
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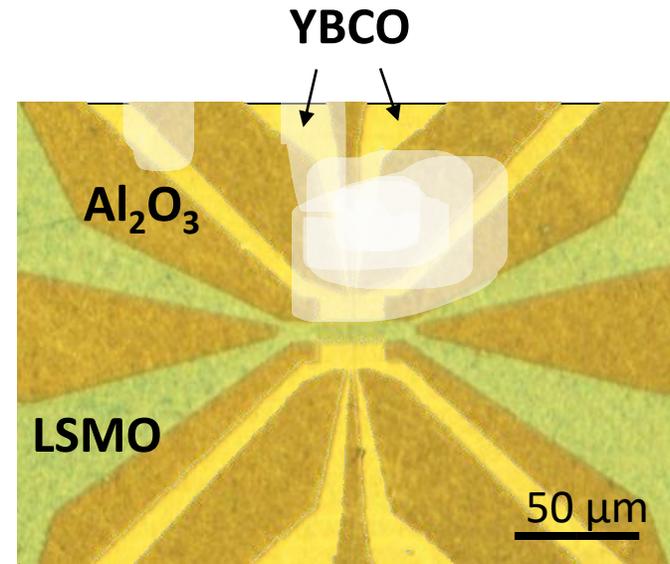
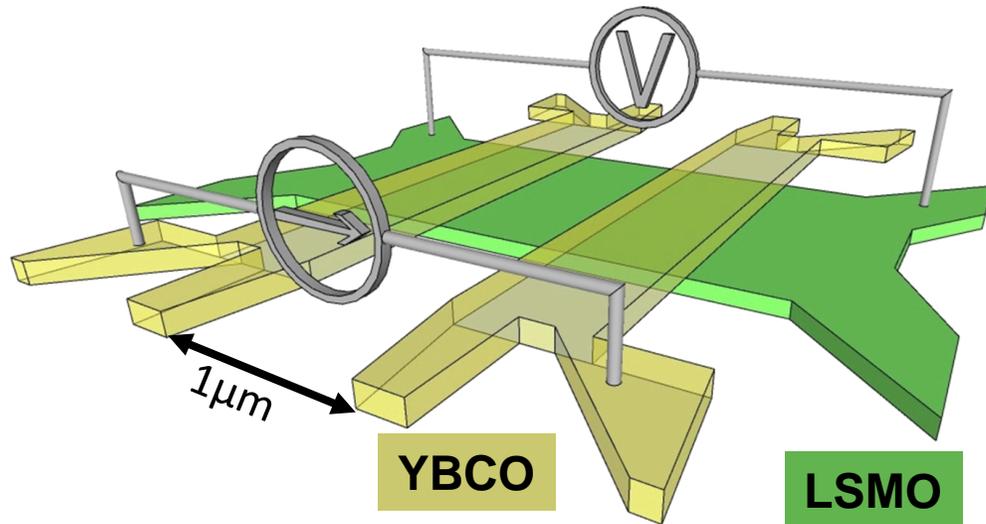
→ AMORPHOUS ALUMINA MASK  
→ LSMO REMAINS MAGNETIC



XMCD-PEEM



# Fabrication of planar Josephson junctions

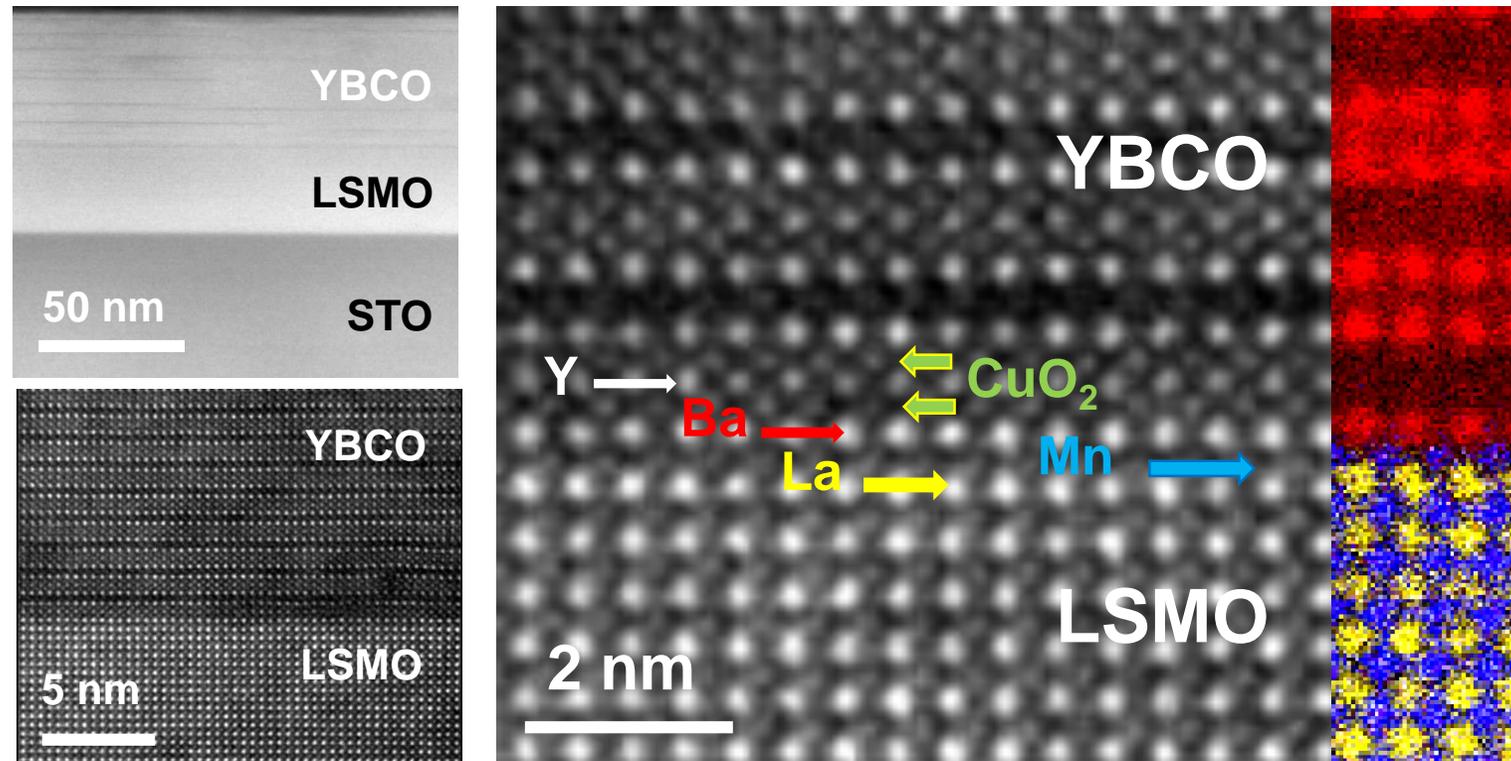


→ PLD GROWTH OF EPITAXIAL YBCO

# Interface quality

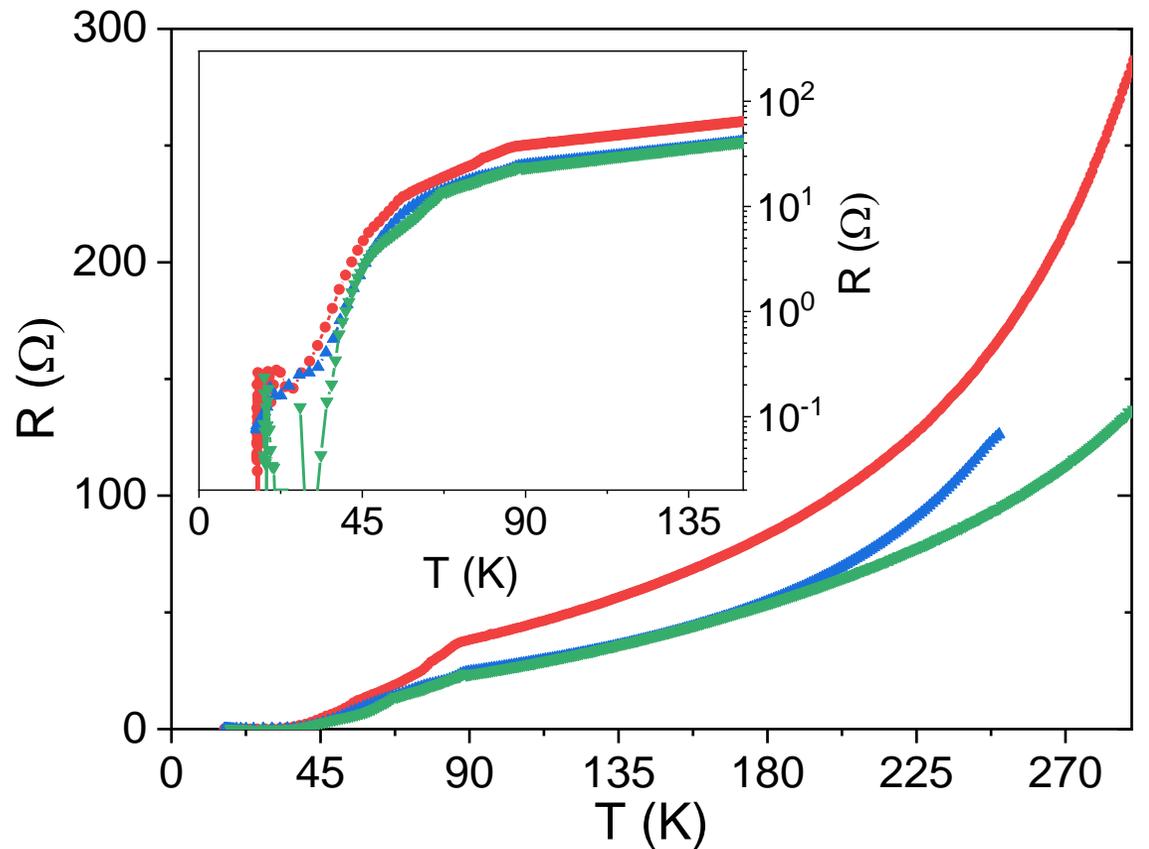
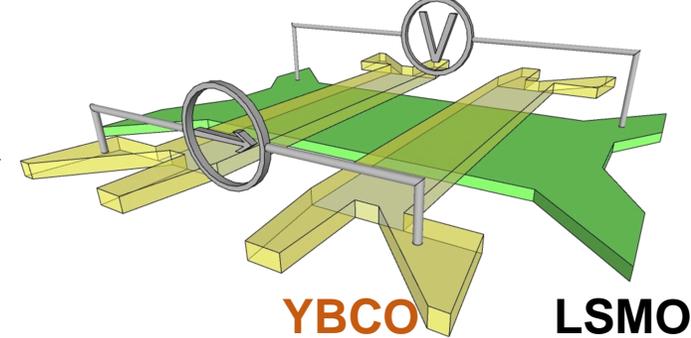
Z-contrast

EELS

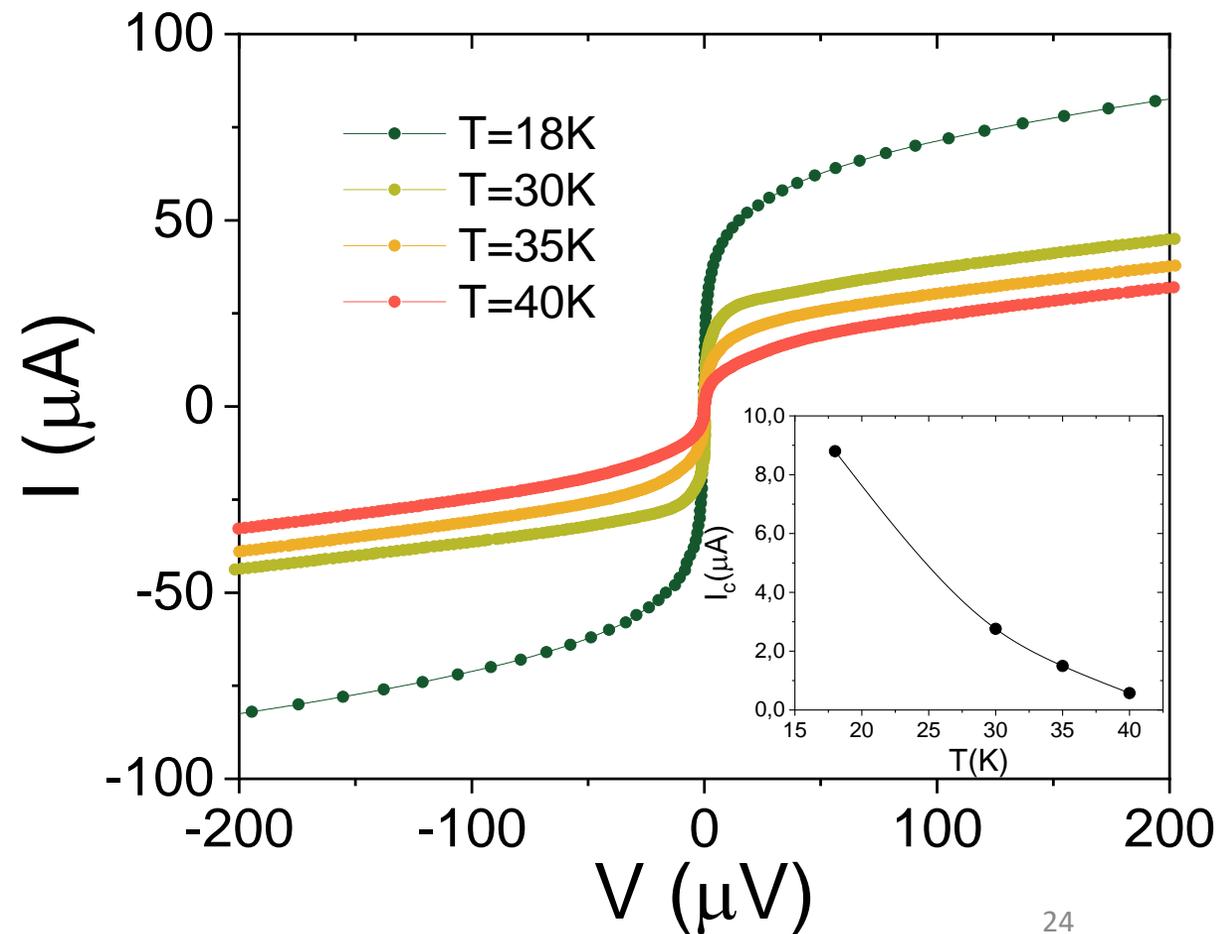


→ HIGH QUALITY INTERFACES DESPITE EX-SITU GROWTH

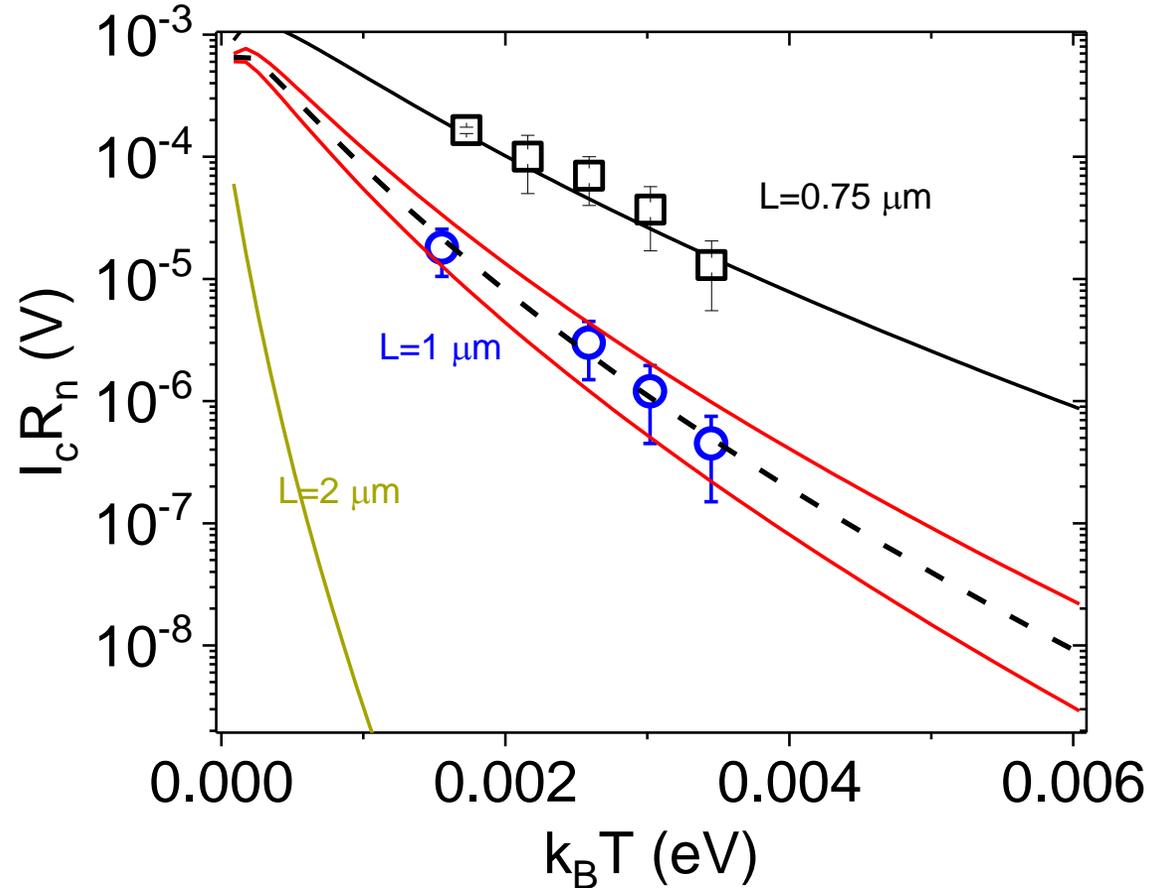
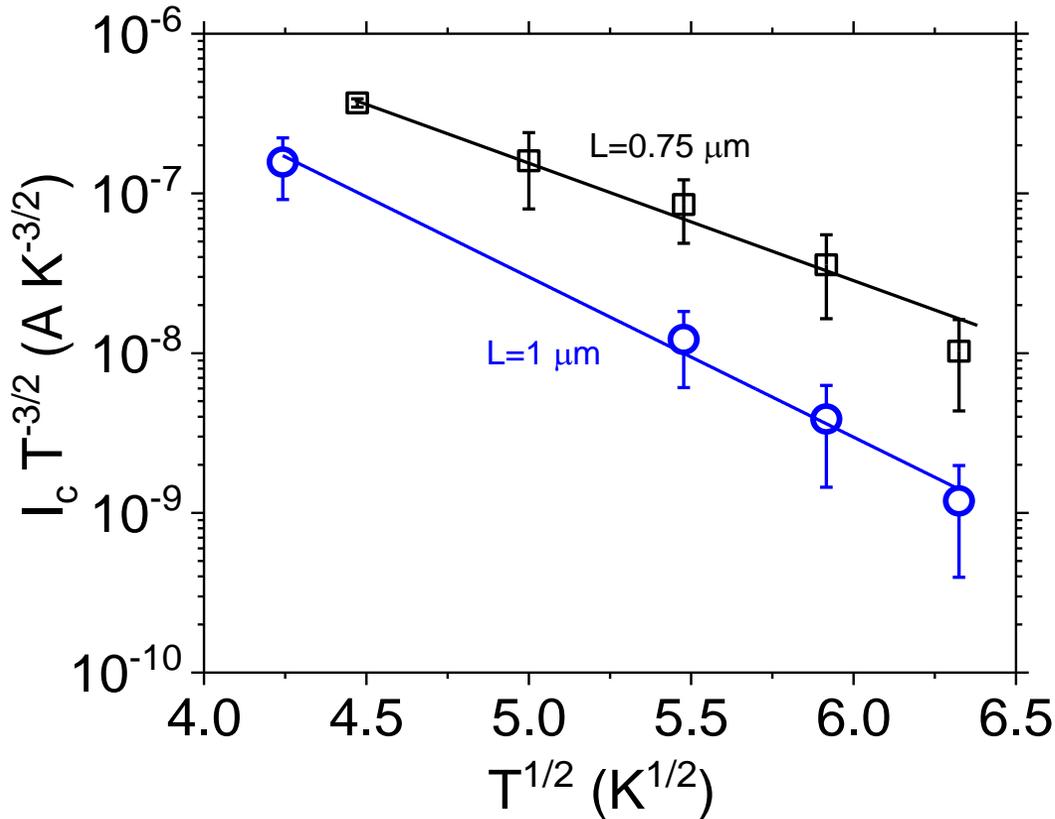
# Supercurrents across the device



→ ZERO RESISTANCE STATE AT ~ 40K



# Size of critical current



$$I_c \propto T^{3/2} \exp(\sqrt{2\pi k_B T / E_{Th}}) \rightarrow \ln(I_c) - \frac{3}{2} \ln(T) \text{ vs } \sqrt{T}$$

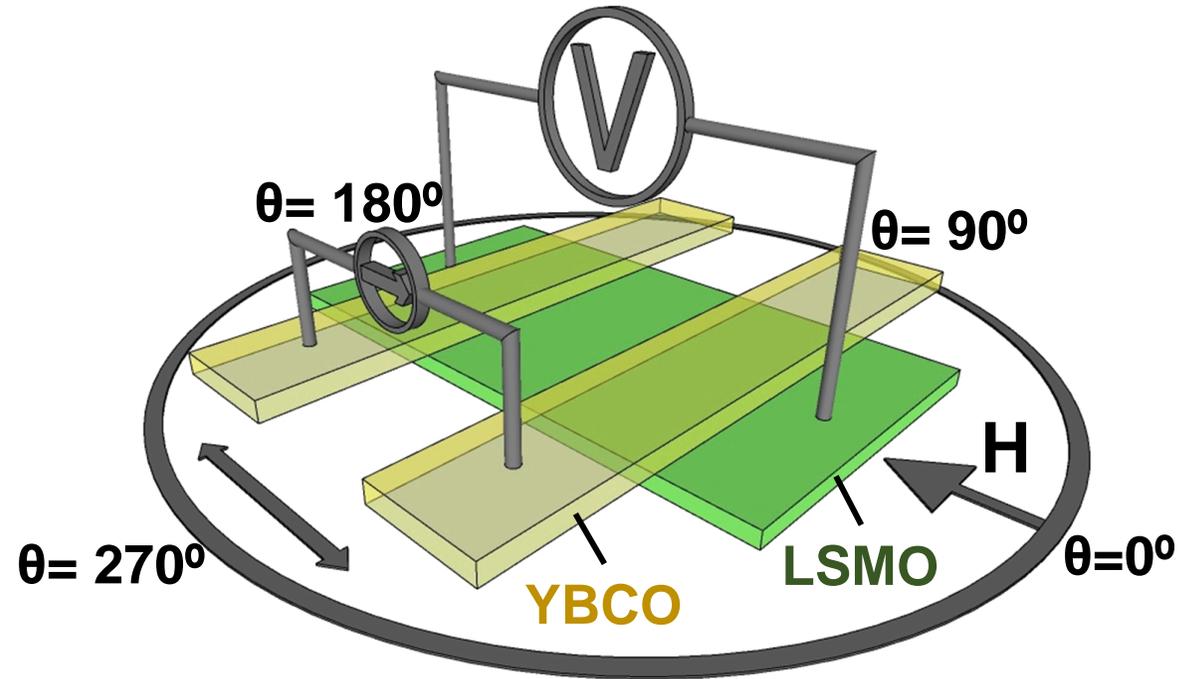
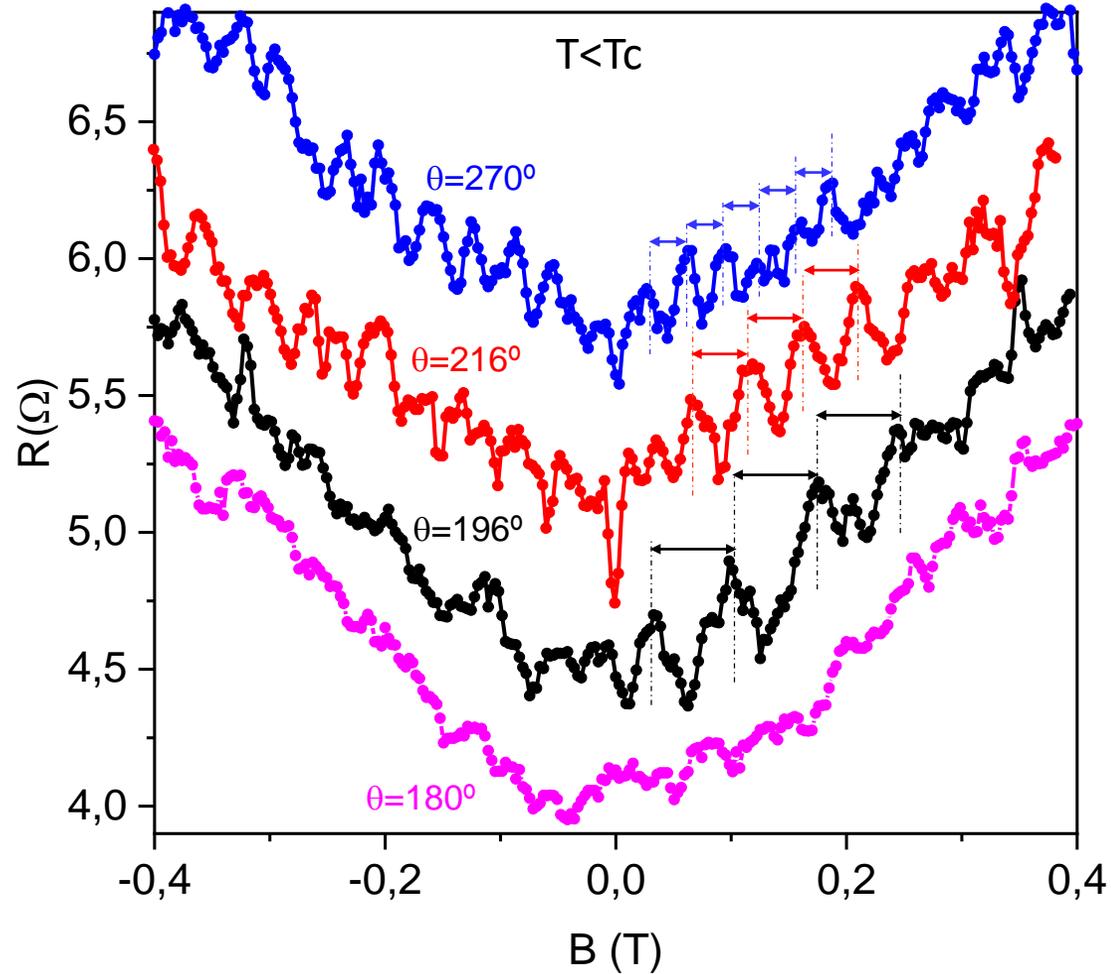
$$E_{Th} = 100 \pm 10 \mu\text{eV} \text{ (for } 1 \mu\text{m)}$$

From: Anwar et al, Appl. Phys. Lett. 100, 052602 (2012)

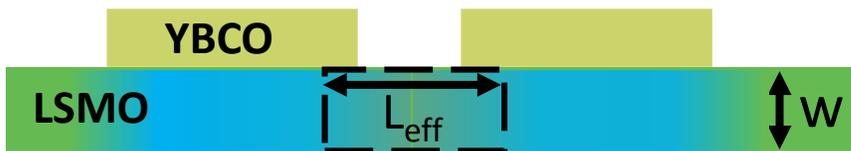
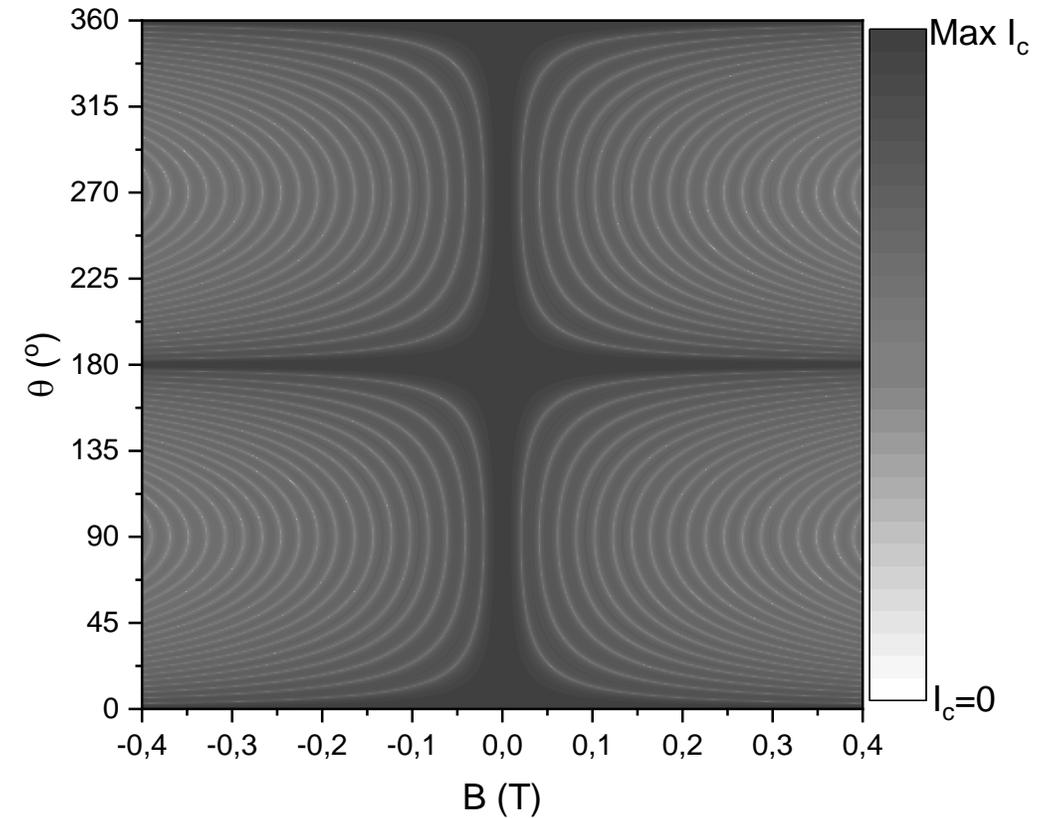
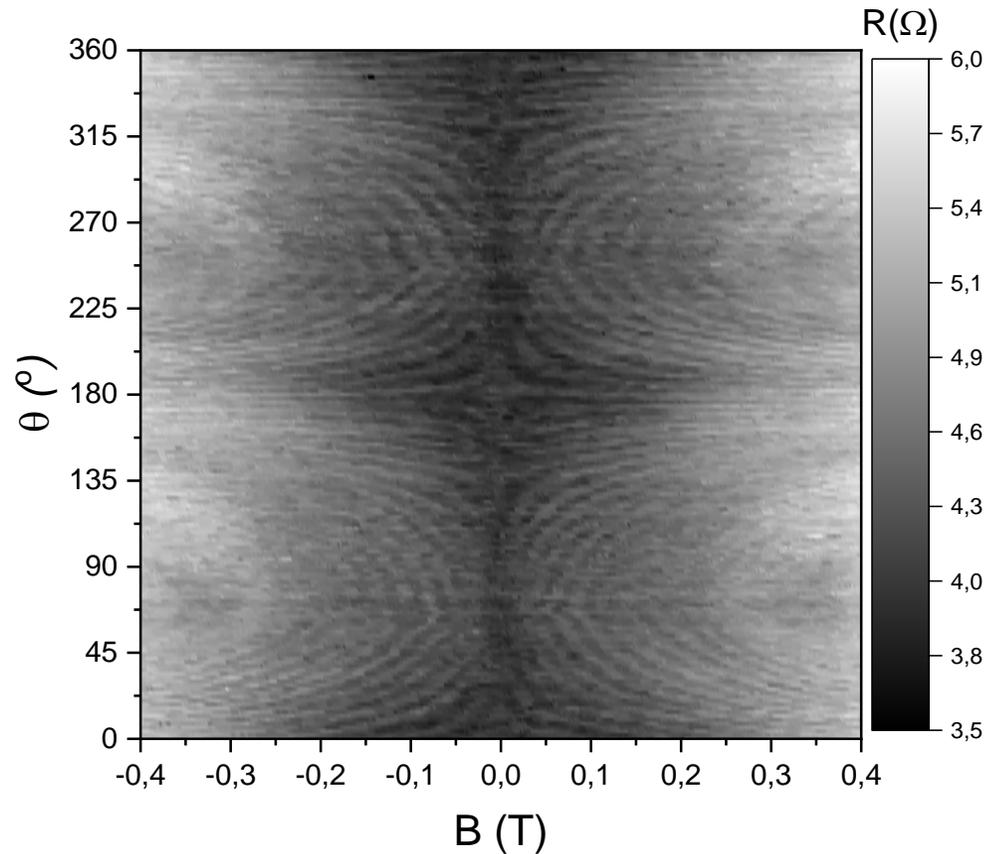
$$eI_c R_n = \frac{32}{3 + 2\sqrt{2}} E_{Th} \left[ \frac{2\pi k_B T}{E_{Th}} \right]^{3/2} e^{-[2\pi k_B T / E_{Th}]^{1/2}}$$

From: Dubos et al, Phys. Rev. B 63, 064502 (2001)

# Magnetic flux quantization: angular dependence



# Magnetic flux quantization: angular dependence

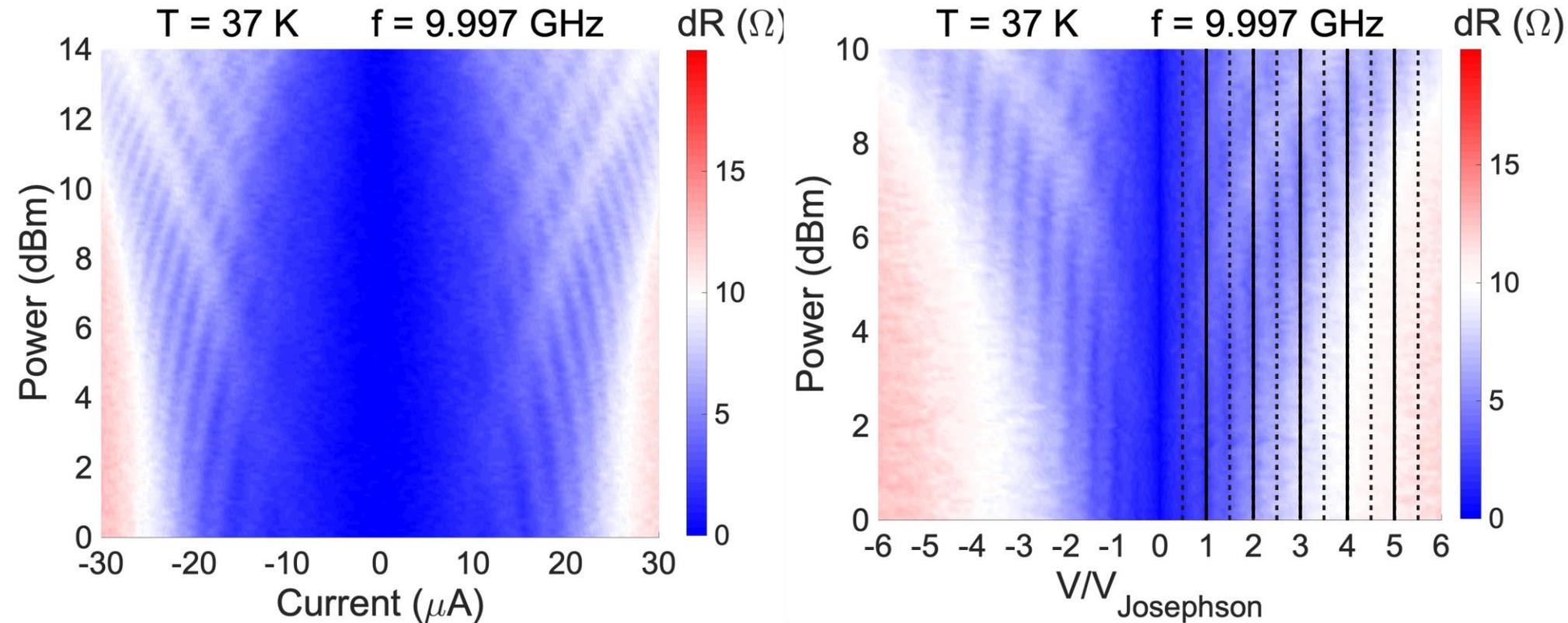


$$I_c = I_0 \left| \frac{\sin\left(\frac{\pi\phi}{\phi_0}\right)}{\left(\frac{\pi\phi}{\phi_0}\right)} \right| = I_0 \left| \frac{\sin\left(\frac{\pi H_{\perp} w L}{\phi_0}\right)}{\left(\frac{\pi H_{\perp} w L}{\phi_0}\right)} \right| = I_0 \left| \frac{\sin\left(\frac{\pi H \sin\theta w L}{\phi_0}\right)}{\left(\frac{\pi H \sin\theta w L}{\phi_0}\right)} \right|$$

$$L_{\text{eff}} = 3,4\mu\text{m}$$

$$L_{\text{eff}} = L_{\text{junction}} + 2\lambda_{\text{YBCO}}$$

# Shapiro steps: quantum interference



$$V_{\text{Josephson}} = n \frac{\hbar\omega}{4e}$$

$$I_c = I_0 \left| \frac{\sin\left(\frac{2\pi\phi}{\phi_0}\right)}{\left(\frac{2\pi\phi}{\phi_0}\right)} \right|$$

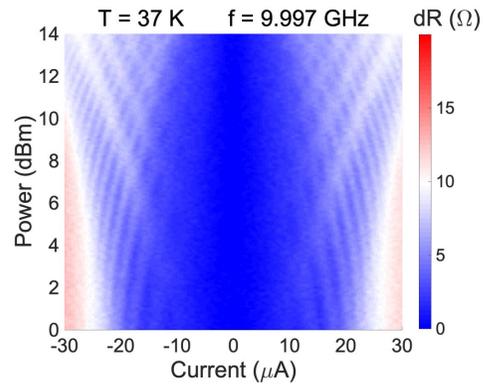
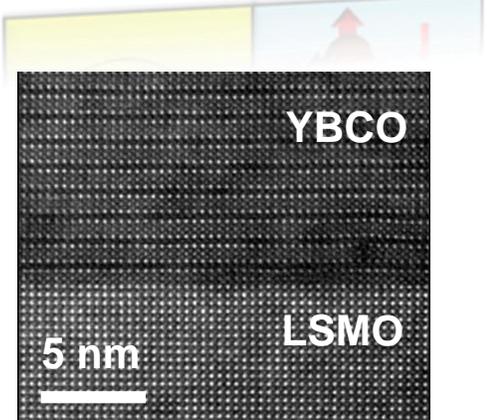
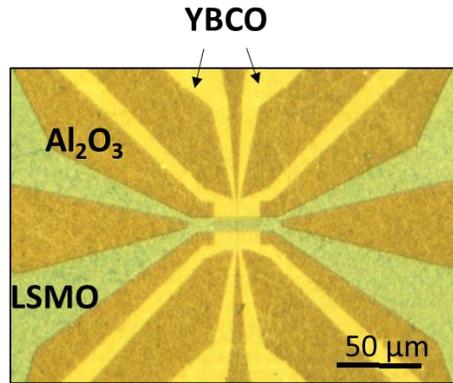
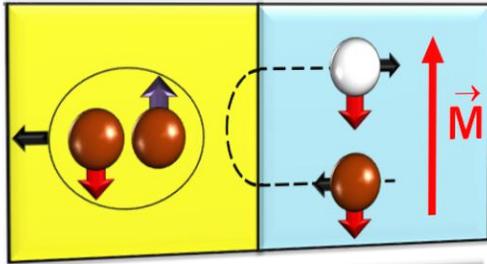
Half integer Shapiro steps → Second harmonic component

Richard, C., Houzet, M. & Meyer, J. S. *Phys. Rev. Lett.* **110**, 1–5 (2013).

Can explain  $L_{\text{eff}} = 2 \times (L_{\text{junction}} + 2\lambda_{\text{YBCO}})$

# The talk in a nutshell

## Proximity effect

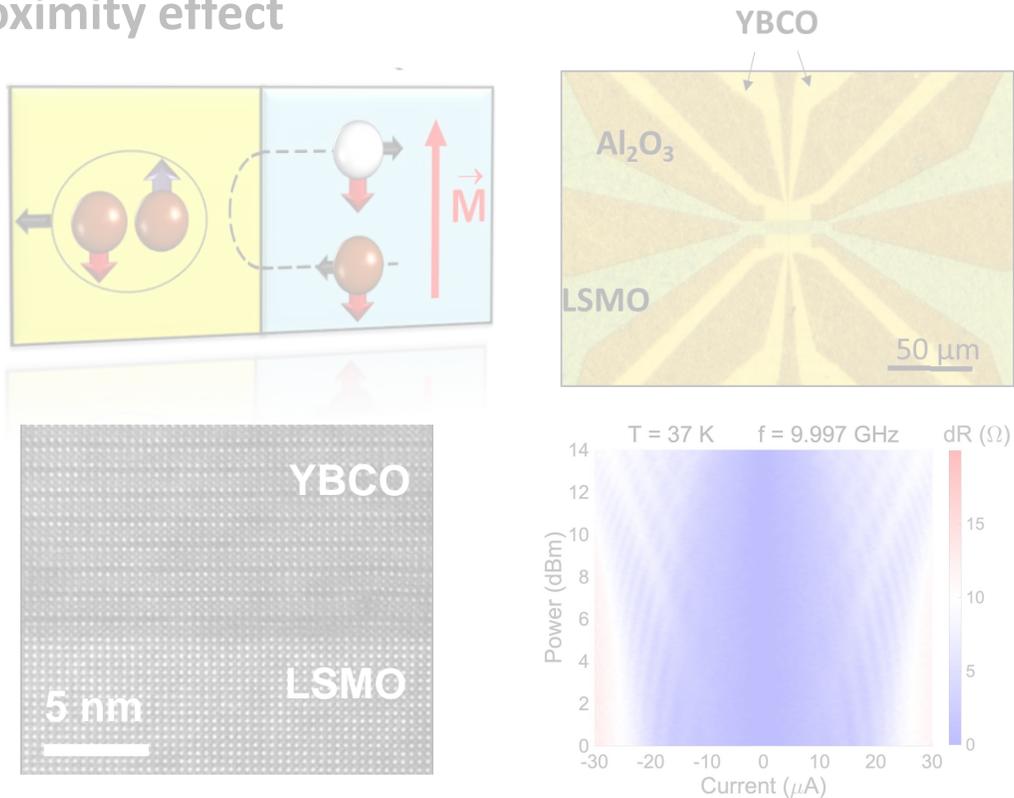


**Fully spin-polarized, high- $T_c$  Josephson supercurrents**  
**Quantum phase coherence effects**

**Sanchez-Manzano *et al.* Nature Materials (2022)**

# The talk in a nutshell

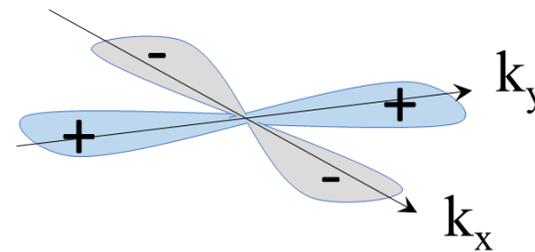
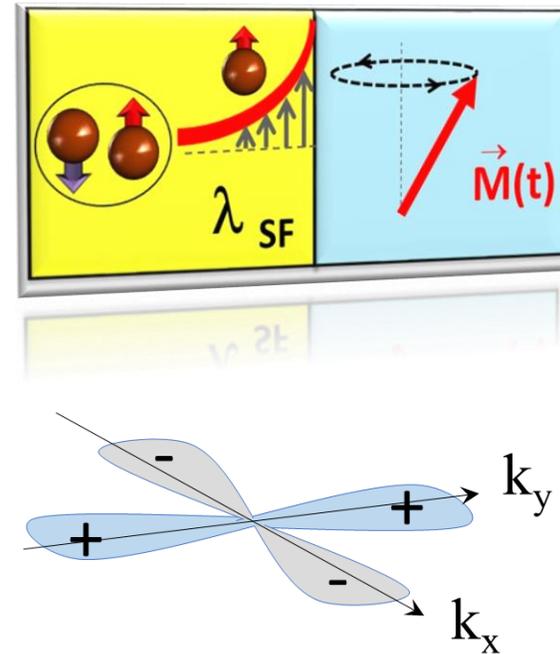
## Proximity effect



Fully spin-polarized, high- $T_C$  Josephson supercurrent  
Quantum phase coherence effects

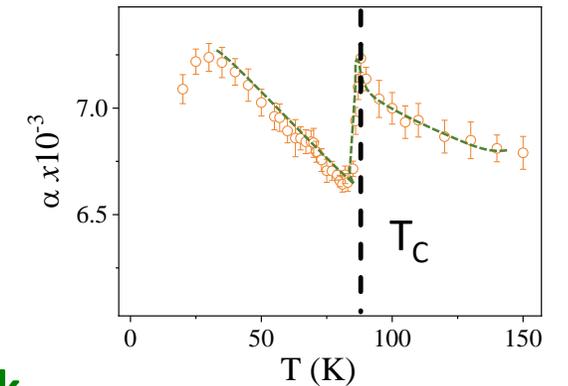
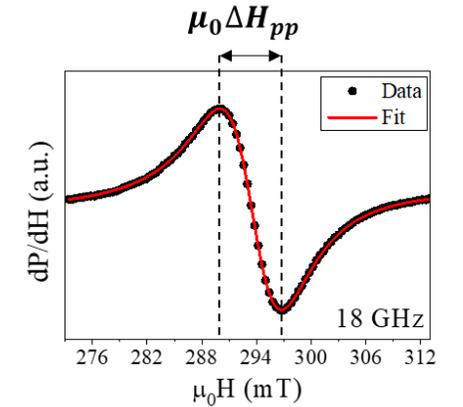
Sanchez-Manzano *et al.* Nature Materials (2022)

## Dynamic coupling



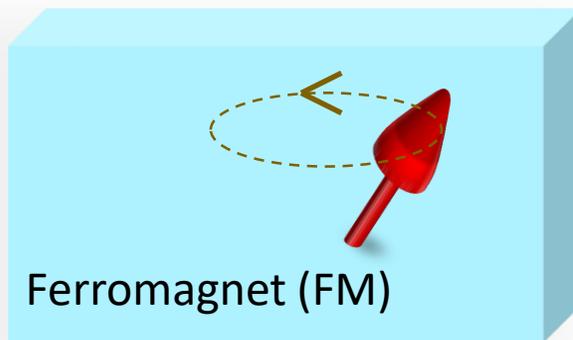
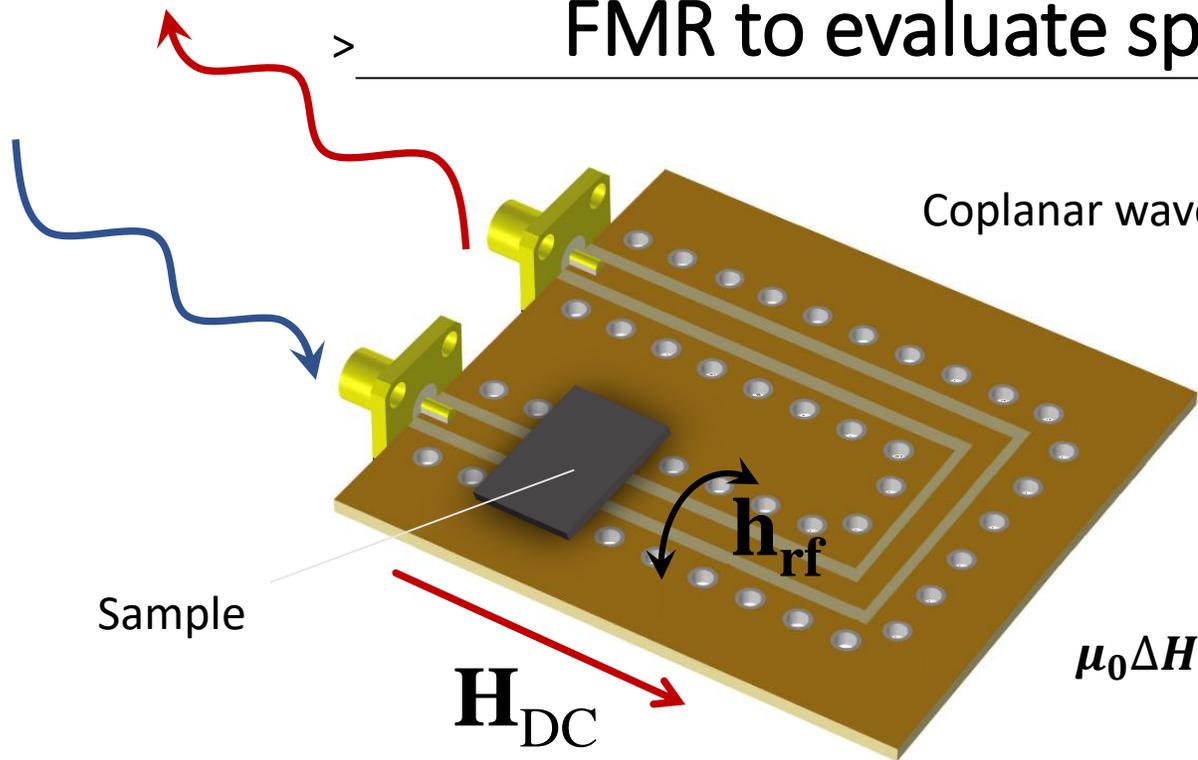
Tunable spin sink  
d-wave effects

Carreira *et al.* PRB (2021)



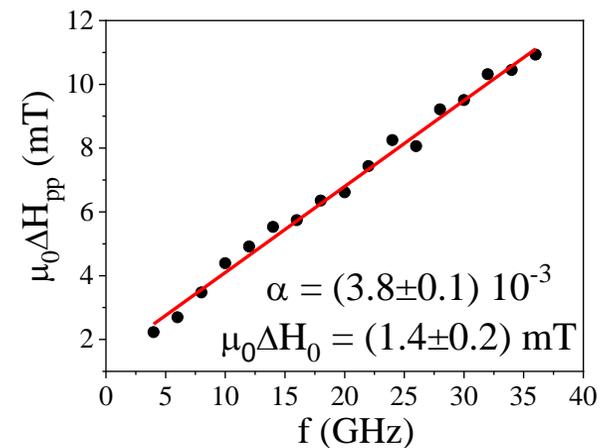
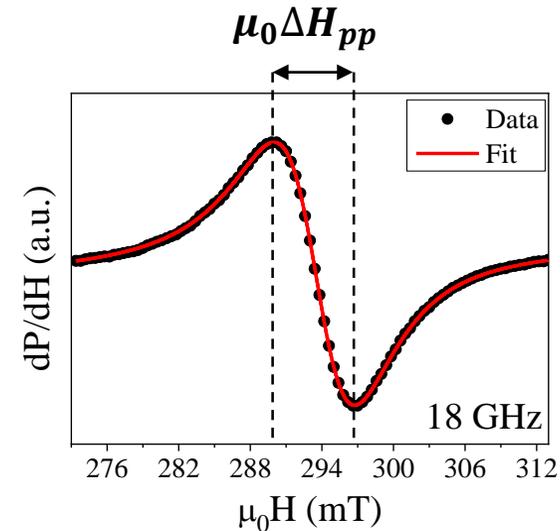
→ OPPORTUNITIES FOR HIGH- $T_C$  SUPERCONDUCTING SPINTRONICS 30

# FMR to evaluate spin absorption by SC

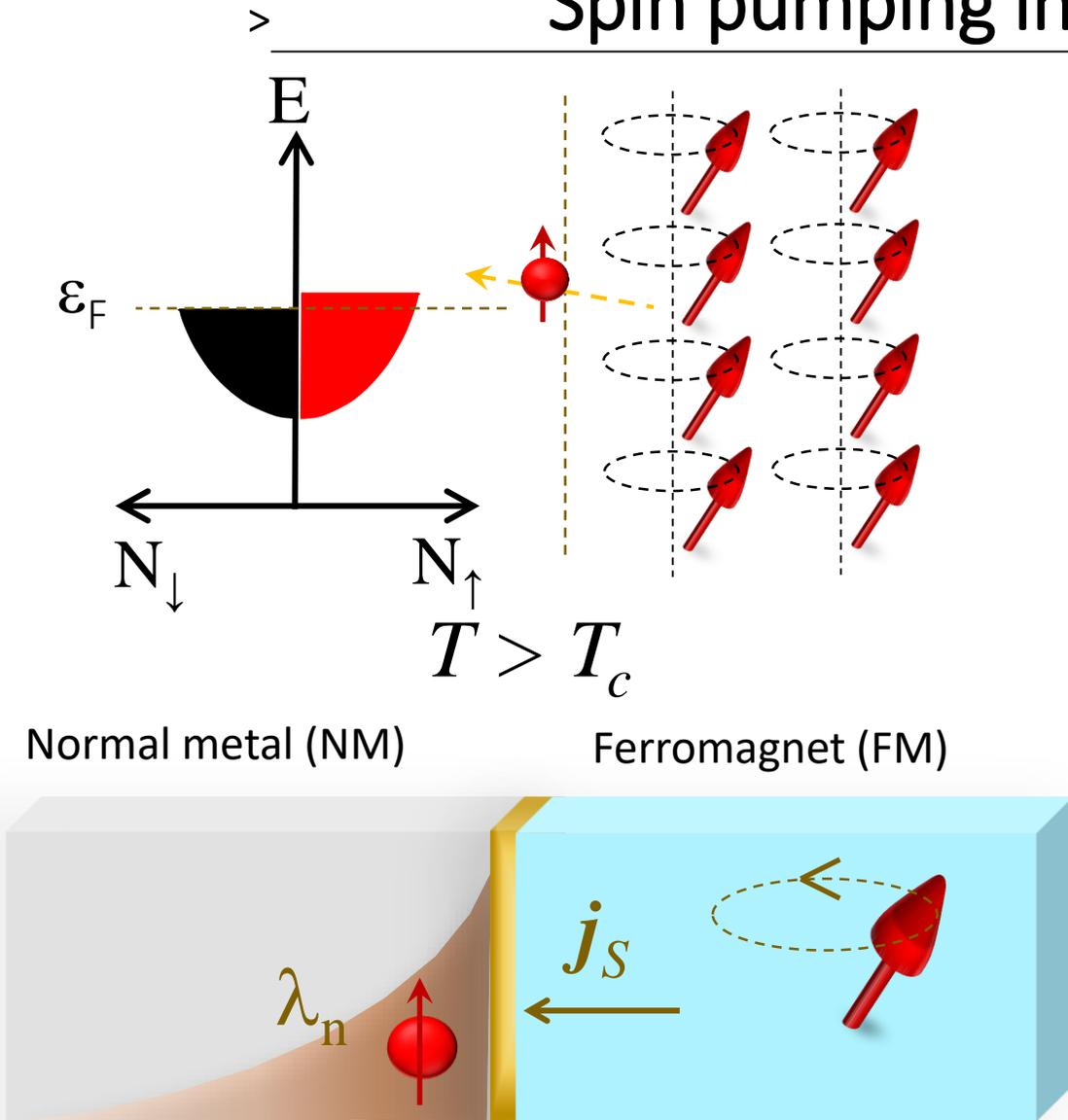


Damping      Inhomogeneous broadening

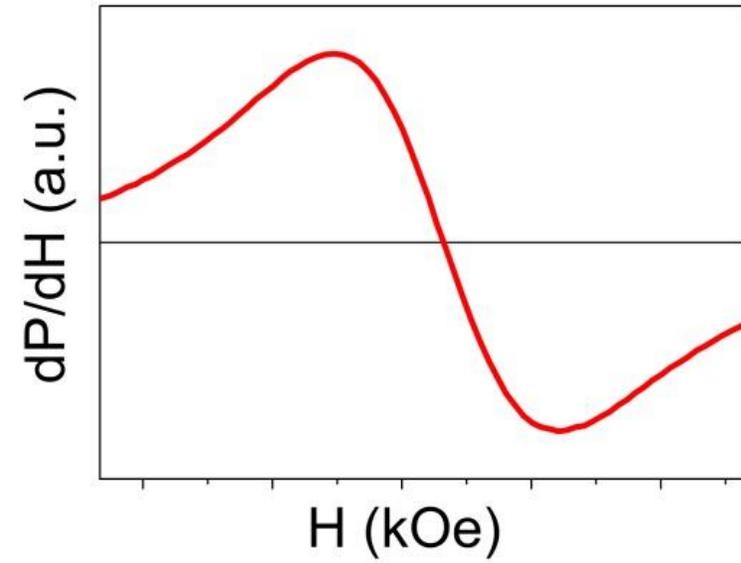
$$\mu_0 \Delta H_{pp} = \frac{2\alpha}{\sqrt{3}\gamma} f + \mu_0 \Delta H_0$$



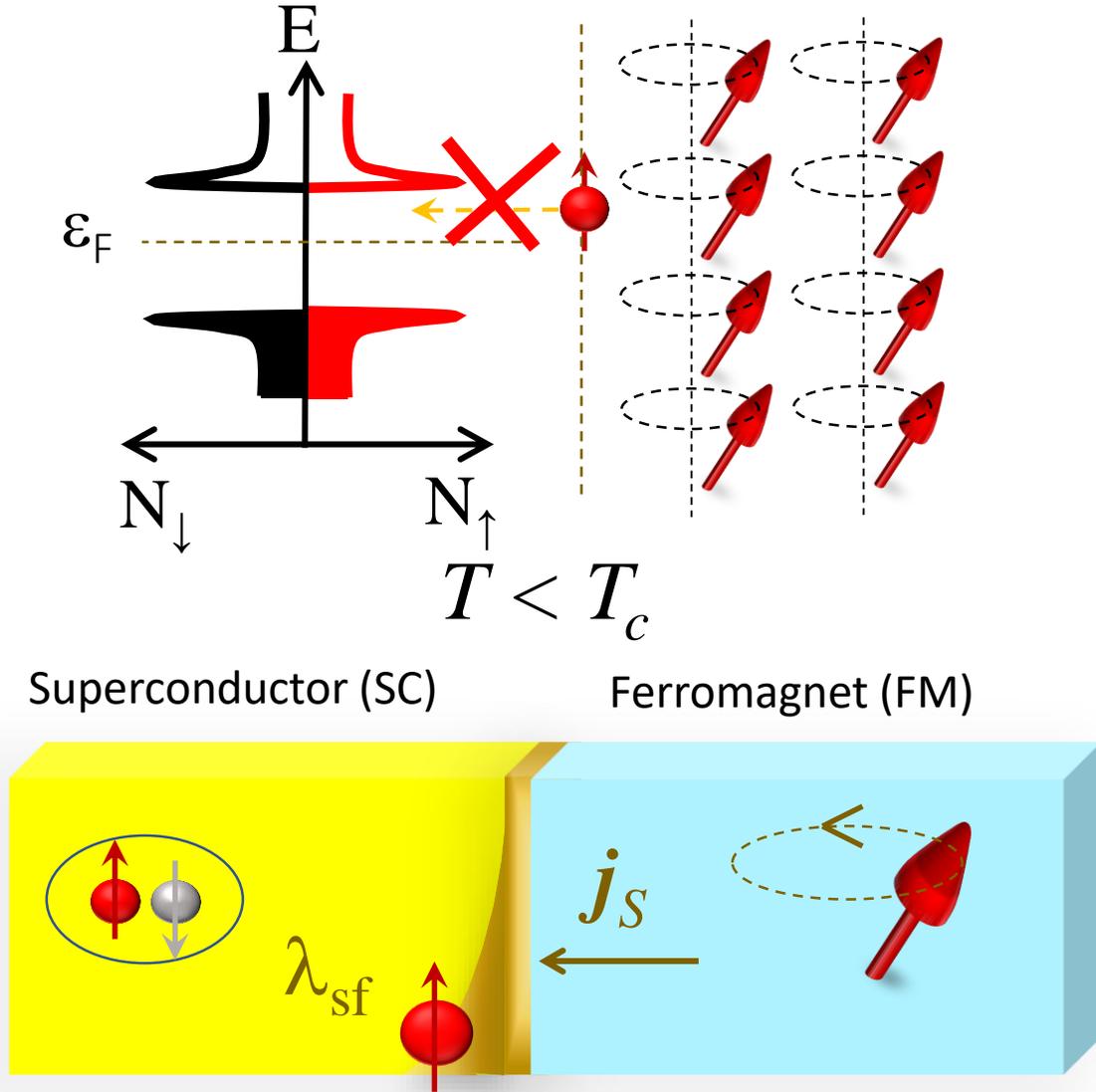
# Spin pumping into normal-metal



Spin sinking  
  
 linewidth broadening

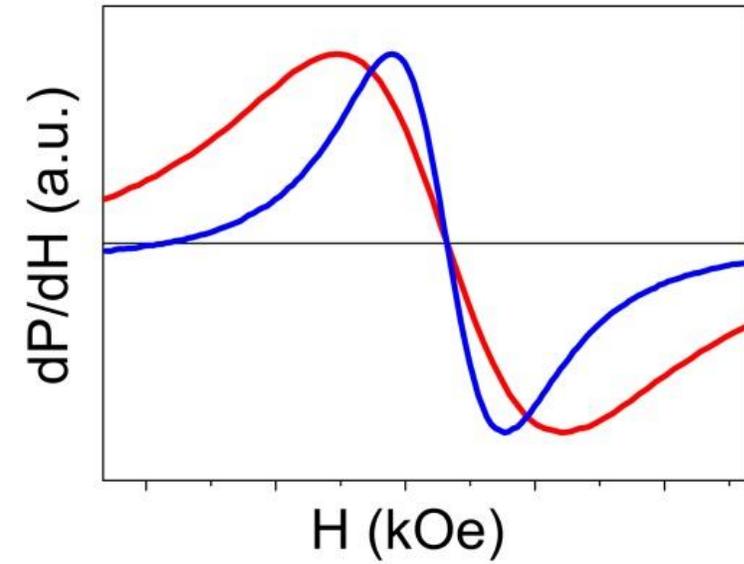


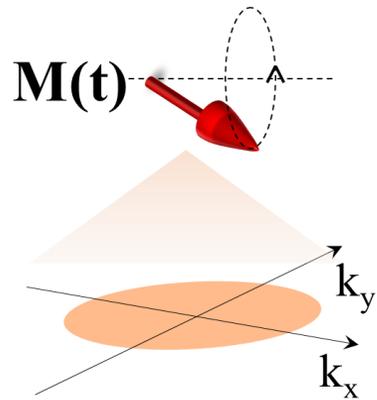
# Spin pumping into a superconductor



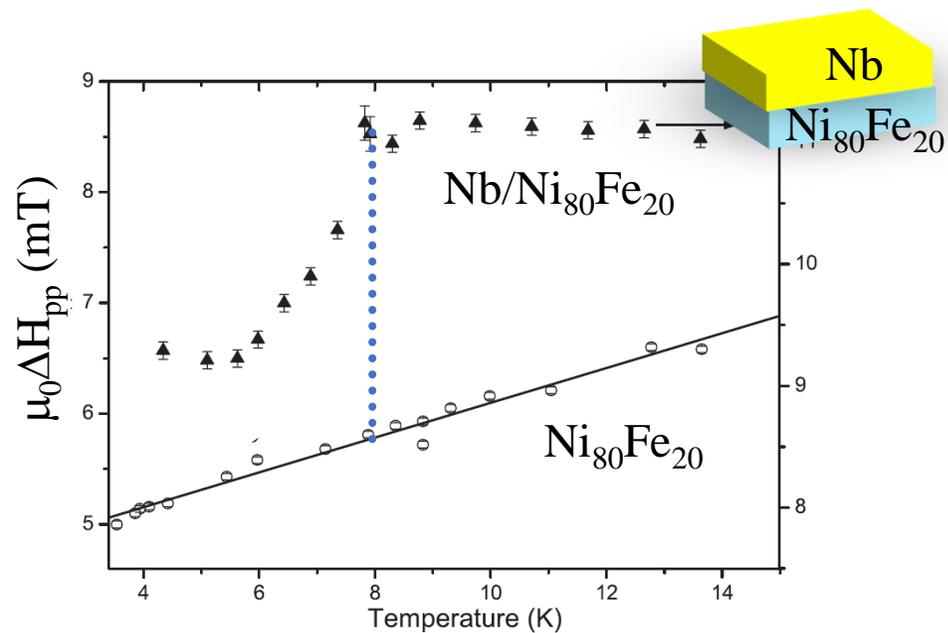
Vanishing DOS reduces spin sinking

linewid~~th~~ narrowing

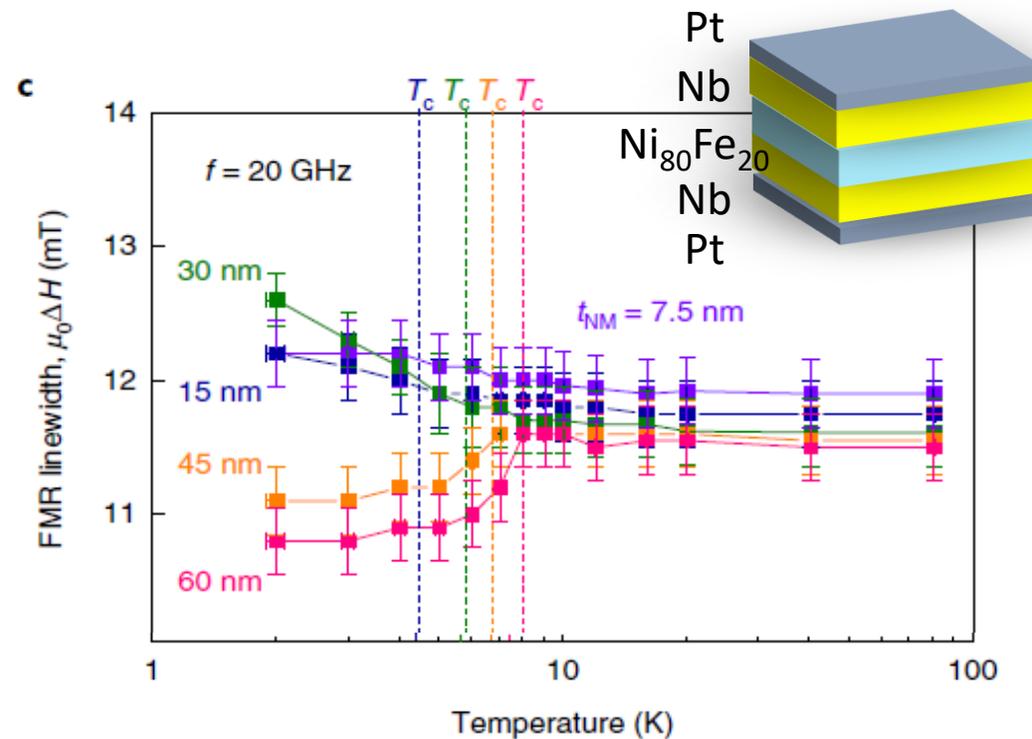




# Experiments with s-wave superconductors

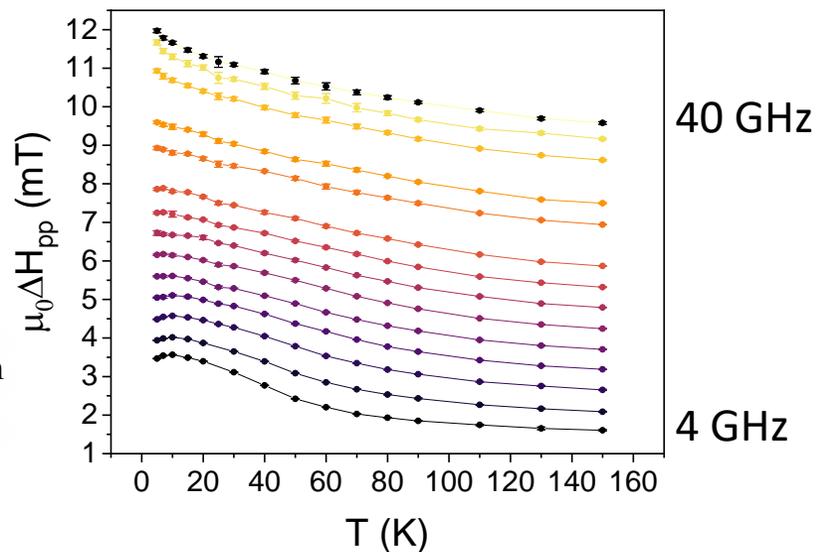
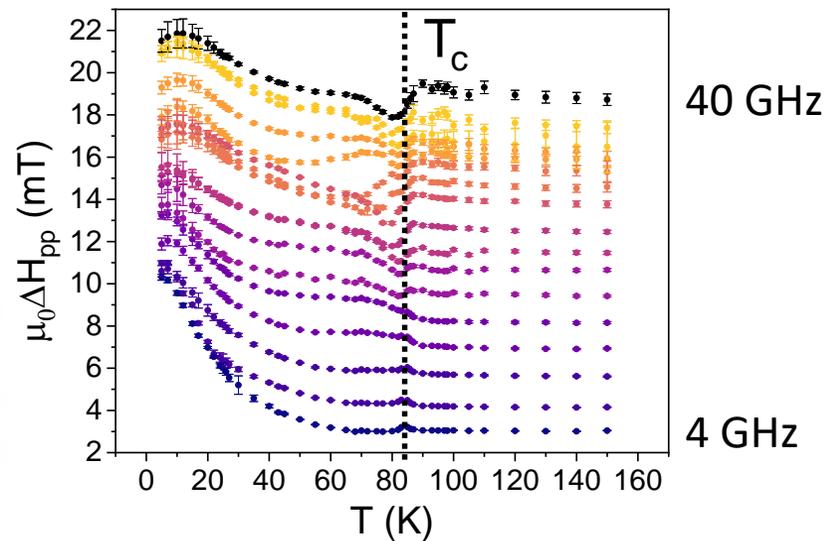
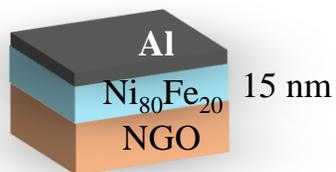
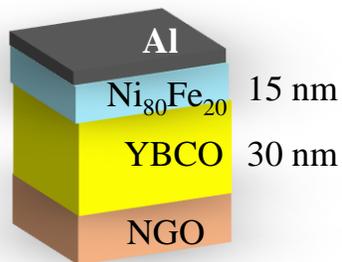
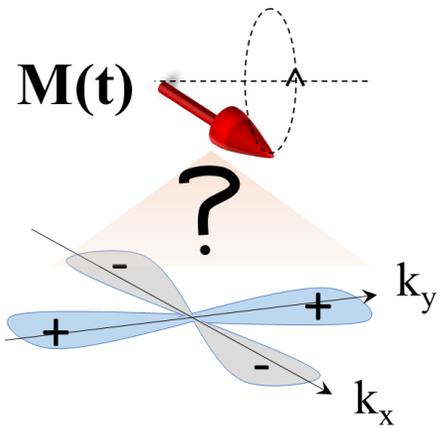


C. Bell et al, *Phys. Rev. Lett.* **100**, 047002 (2008)



K. Jeon et al. *Nat. Mater.* **17**, 499–503 (2018)

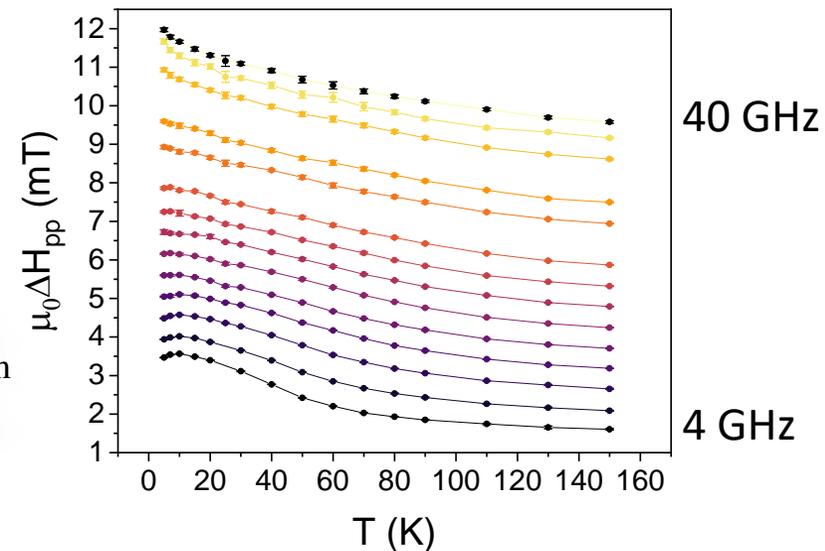
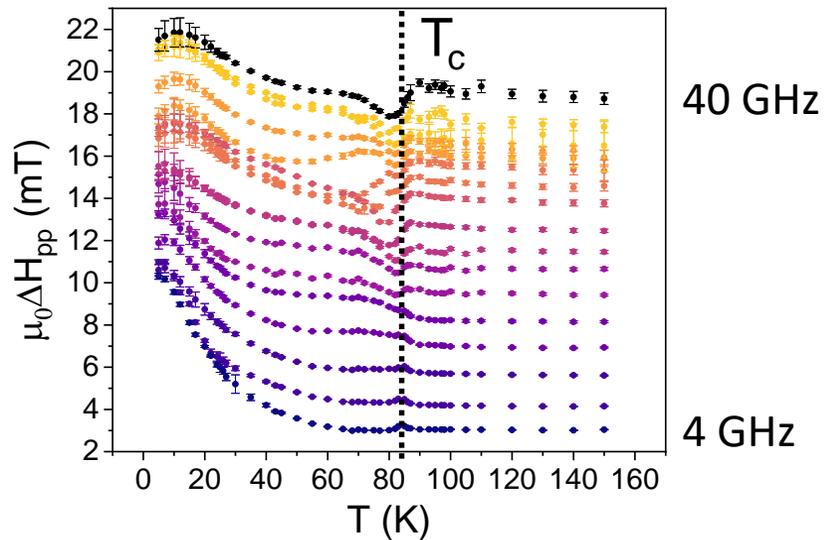
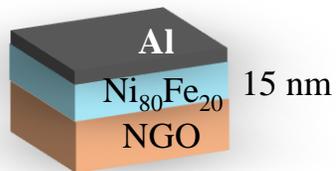
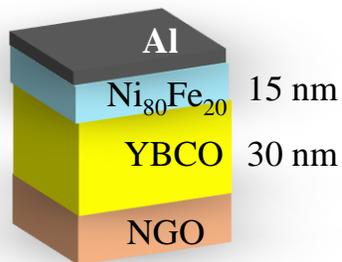
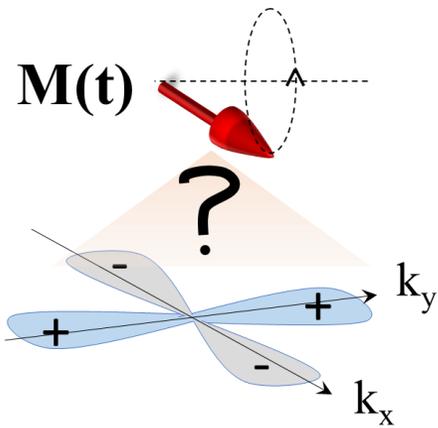
# Experiments with d-wave superconductors



→ Drastic change below  $T_c$

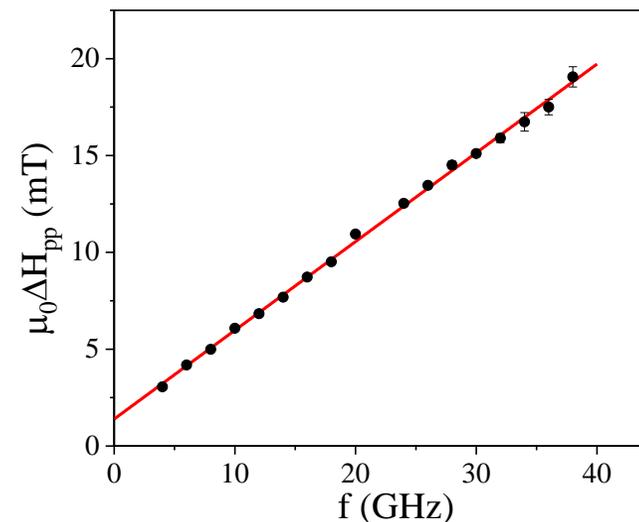
→ Frequency dependent

# Experiments with d-wave superconductors

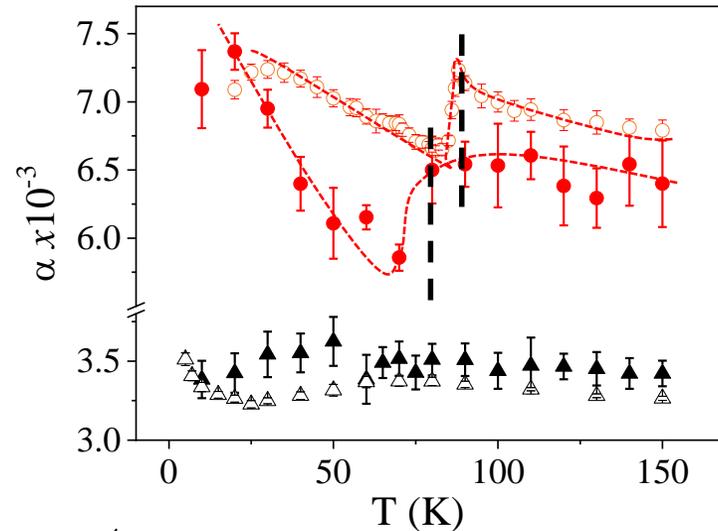
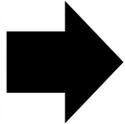
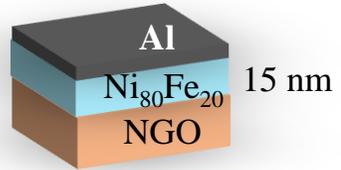
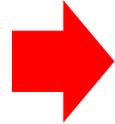
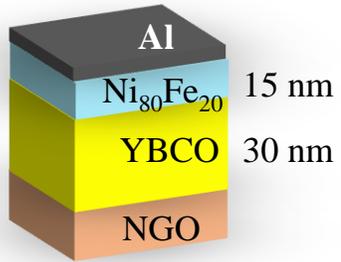


→ Drastic change below  $T_C$   
 → Frequency dependent

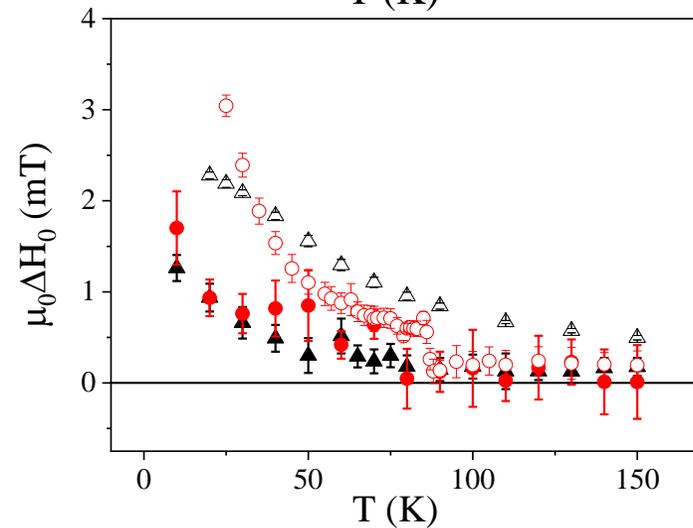
$$\Delta H_{pp} = \frac{2\alpha}{\sqrt{3}\gamma\mu_0} f + \Delta H_0$$



# Experiments with d-wave superconductors

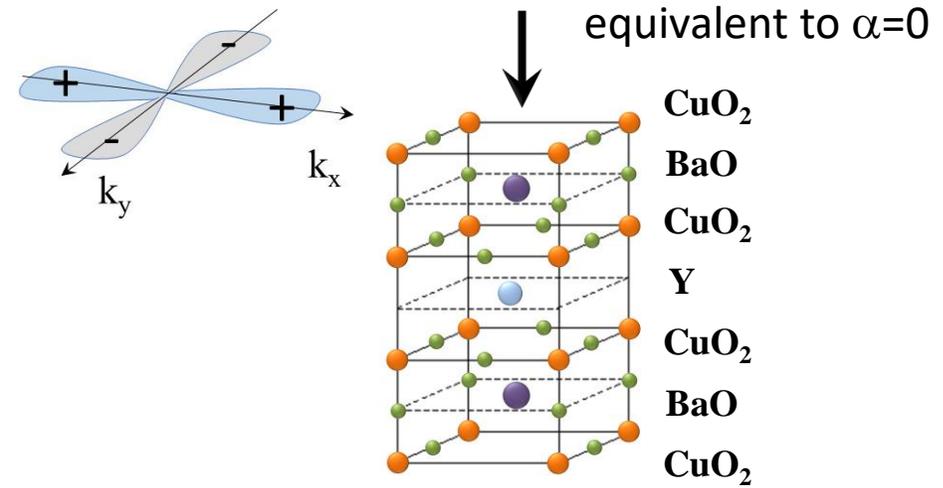
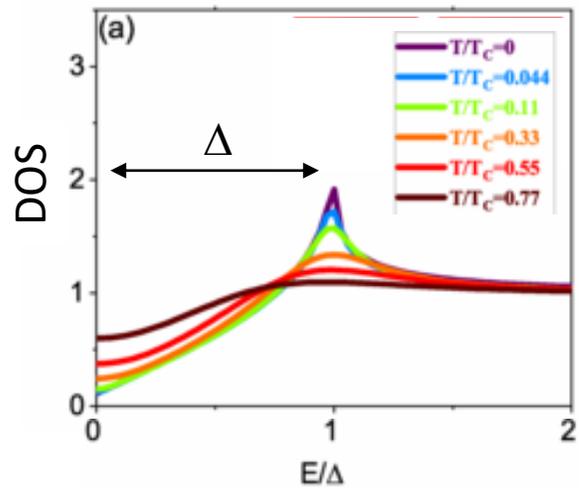


- $\alpha$  drops at  $T_c$
- unexpected upturn further below

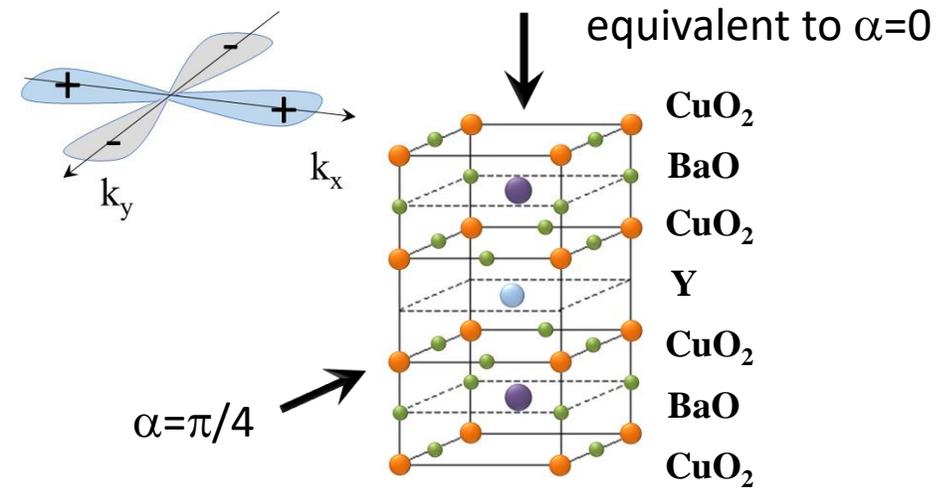
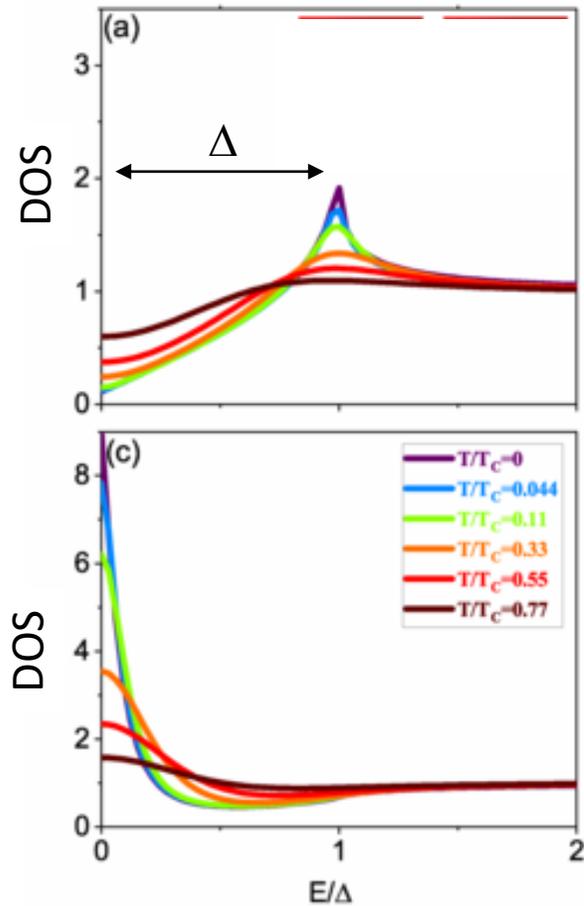


- $\Delta H_0$  not affected by SC

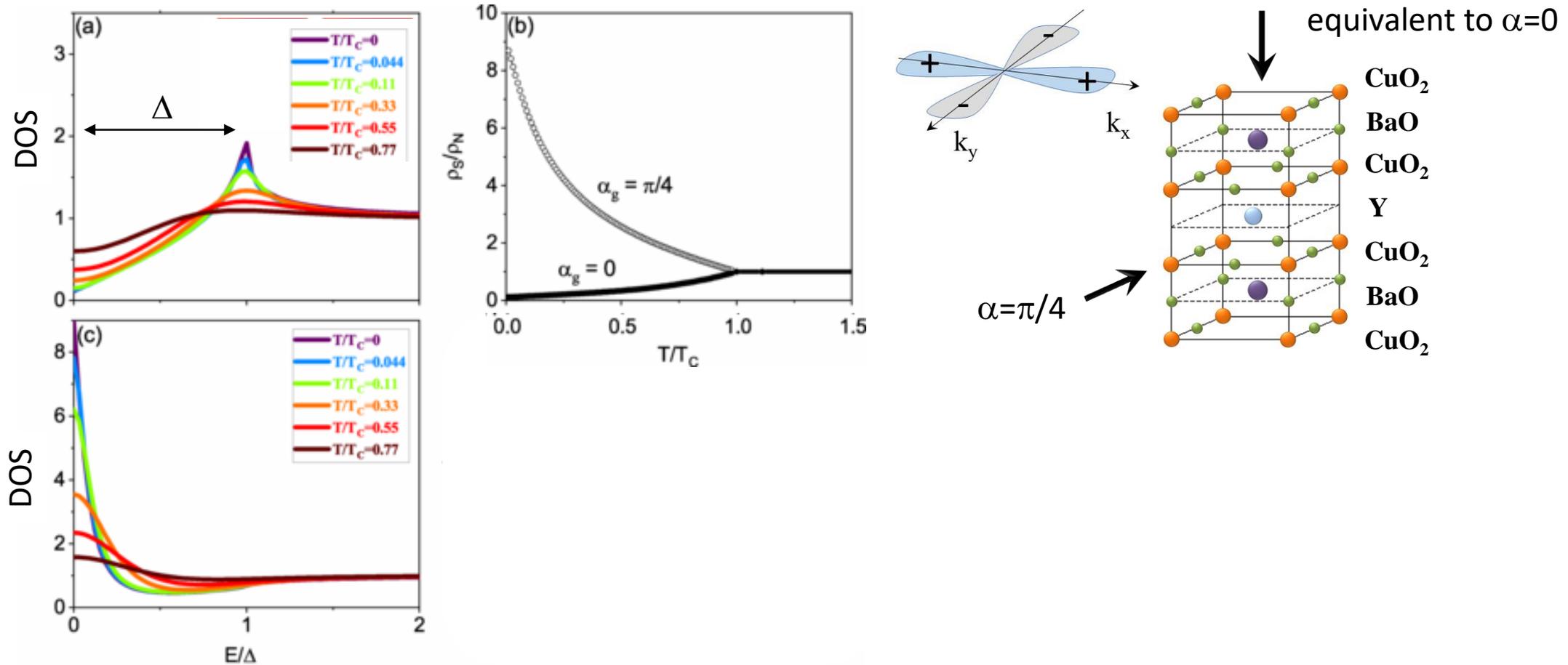
# Anisotropic DOS as a function of temperature



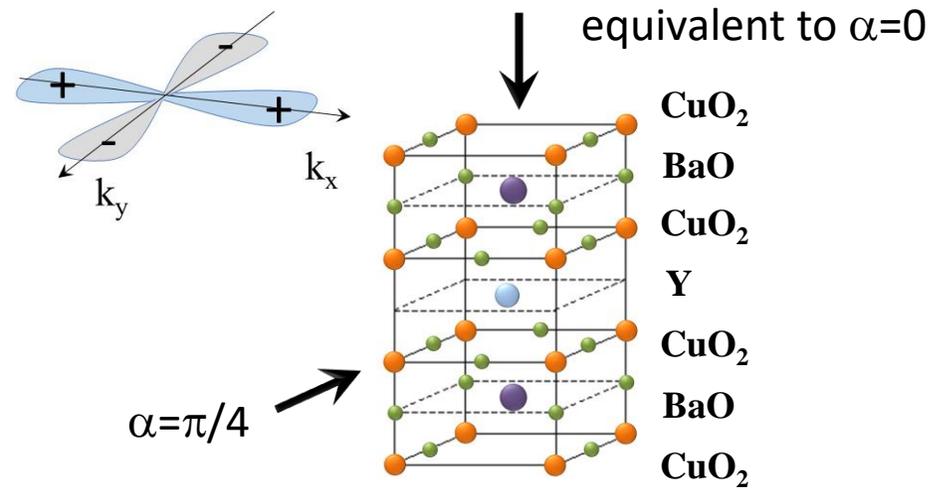
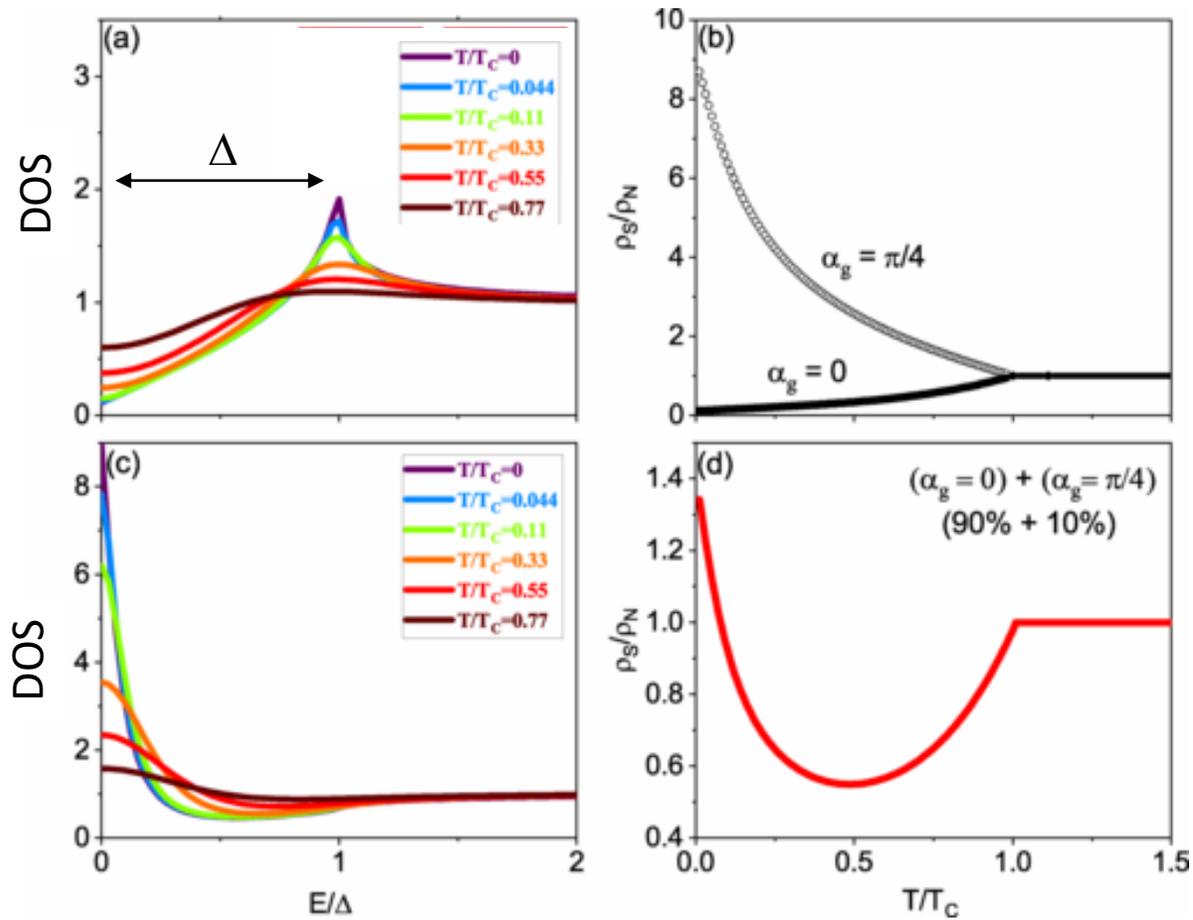
# Anisotropic DOS as a function of temperature



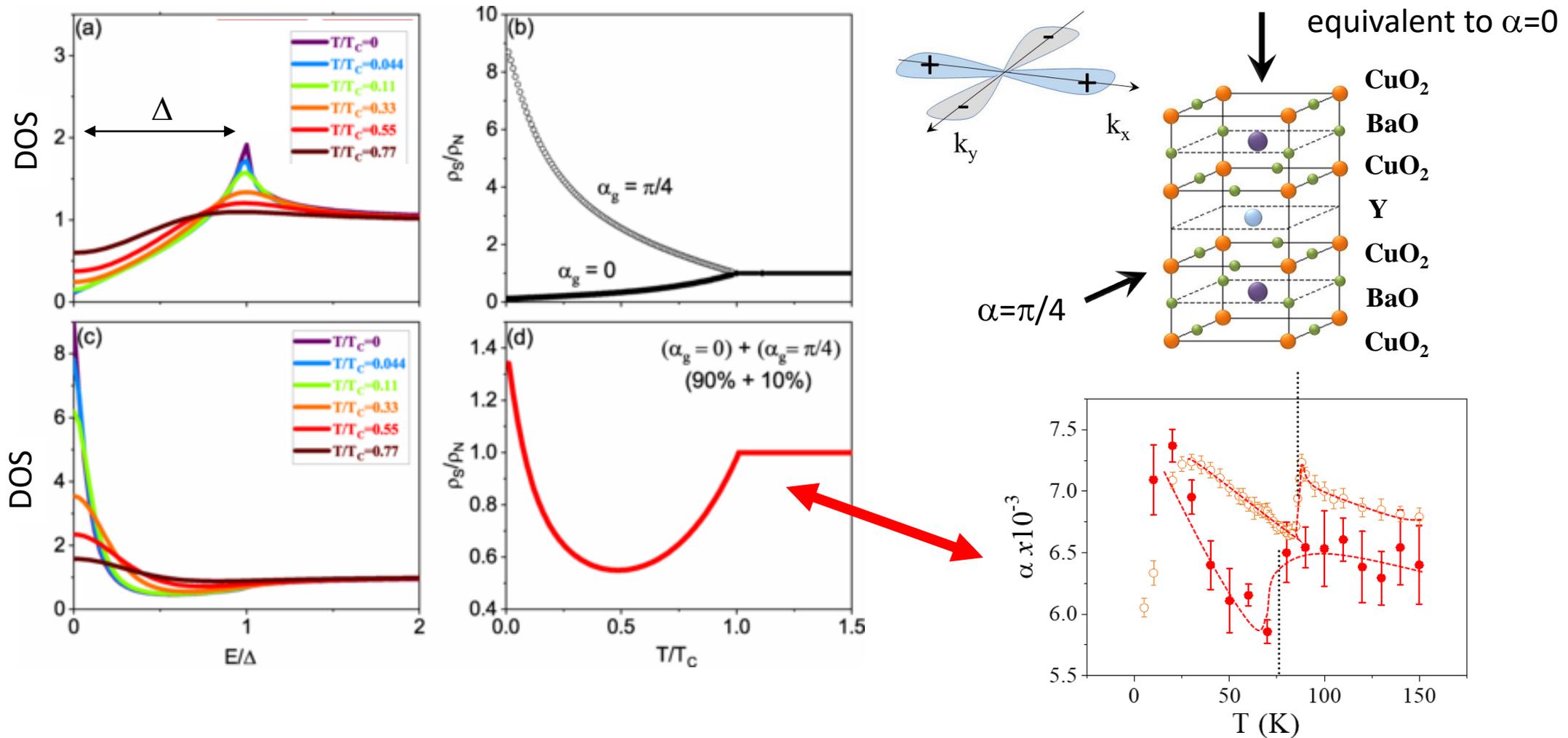
# Anisotropic DOS as a function of temperature



# Anisotropic DOS as a function of temperature

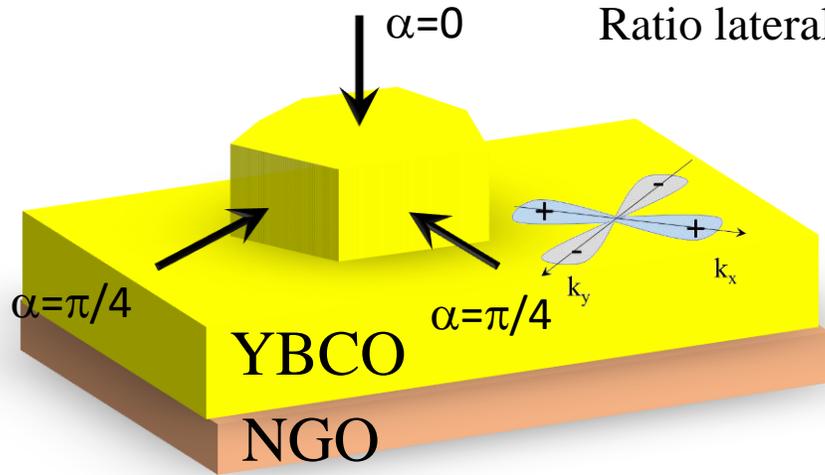
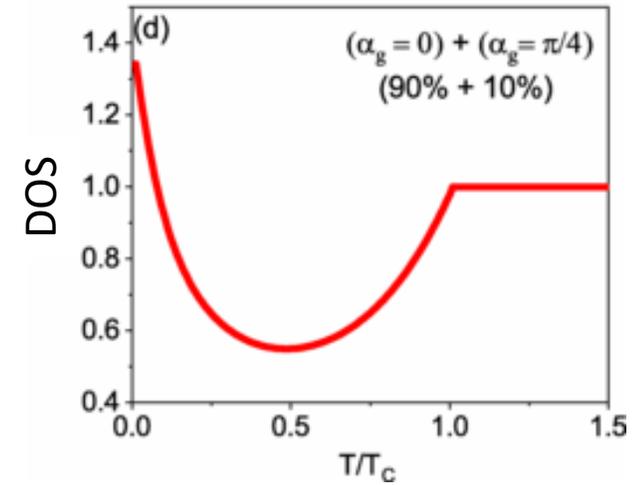
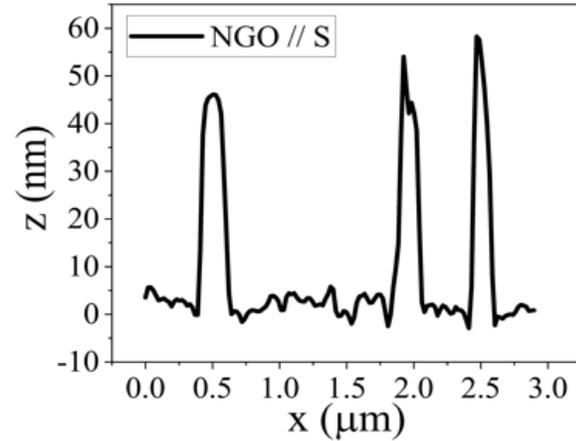
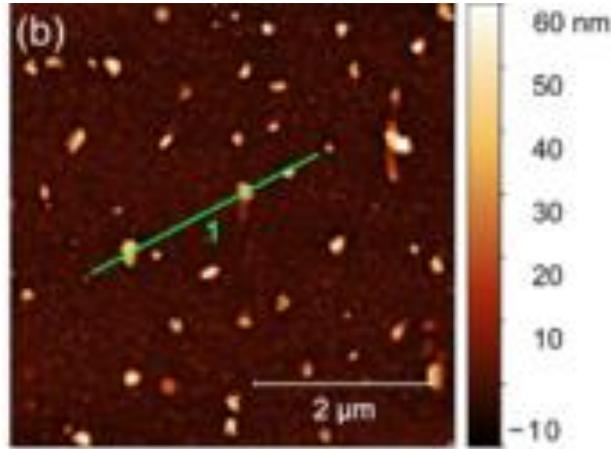


# Anisotropic DOS as a function of temperature

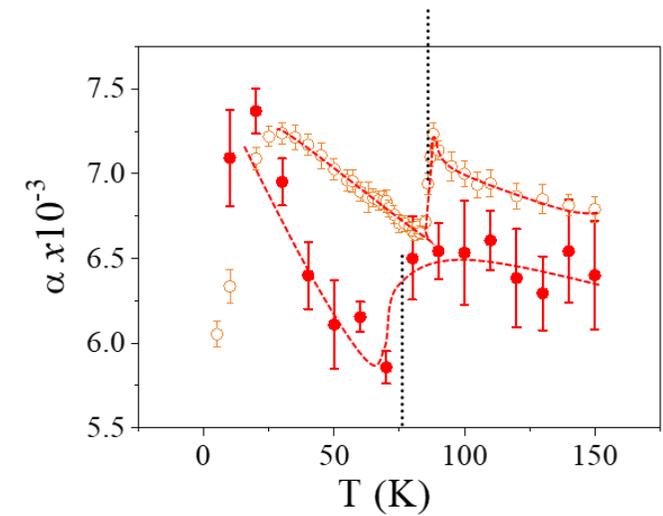
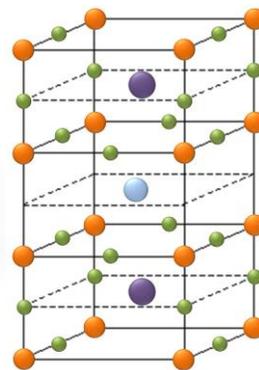


→ TEMPERATURE DEPENDENT QUASIPARTICLE DENSITY EXPLAINS  $\alpha(T)$

# Surface topography and effective DOS

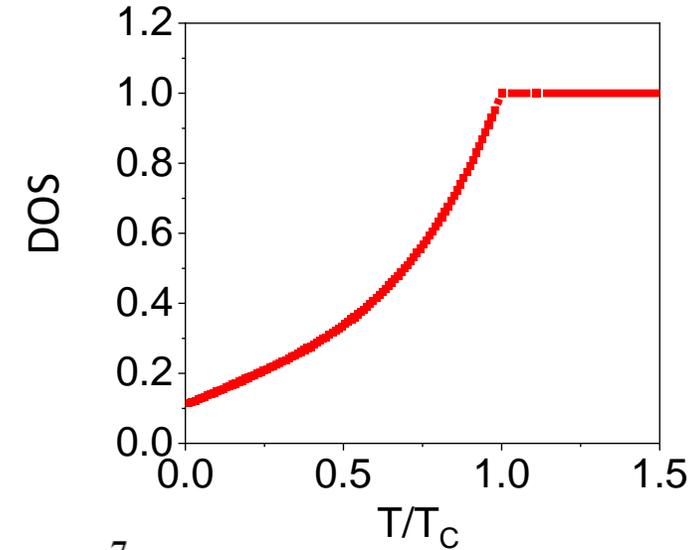
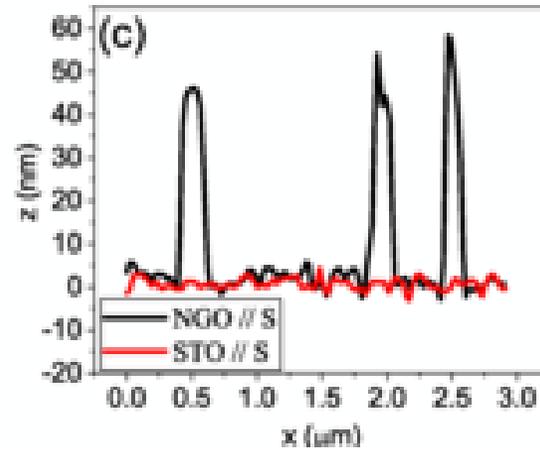
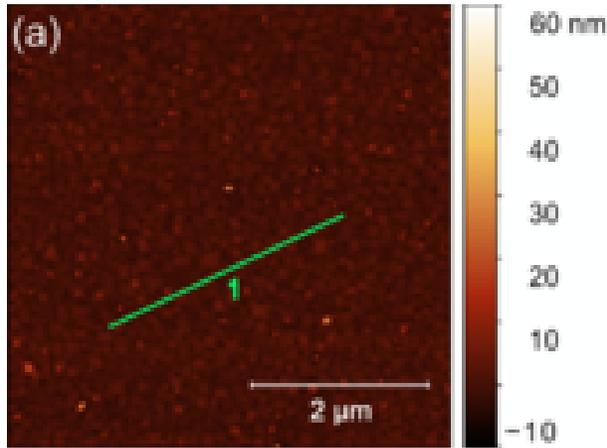


Ratio lateral/vertical surface  $\approx 10\%$

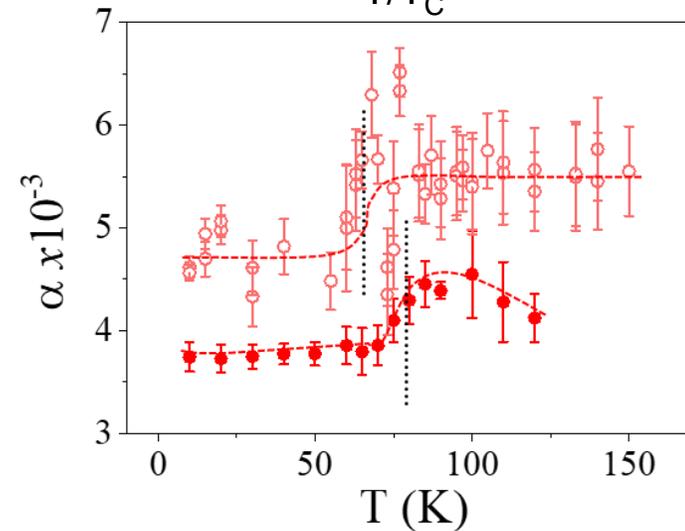
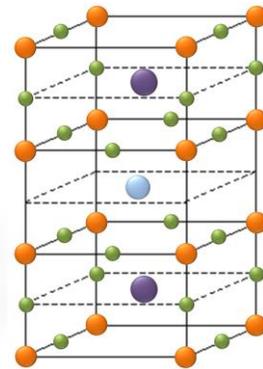
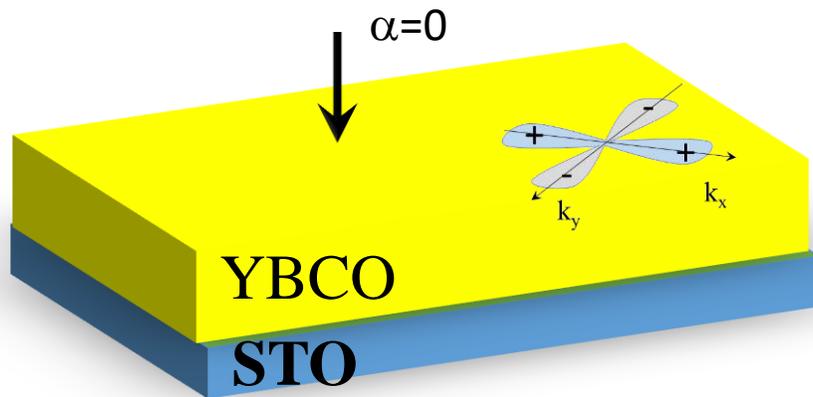


**→ TOPOGRAPHY ALLOWS ACCESS TO LARGE QUASIPARTICLE DOS**

# Surface topography and effective DOS



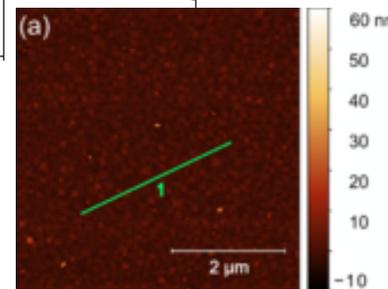
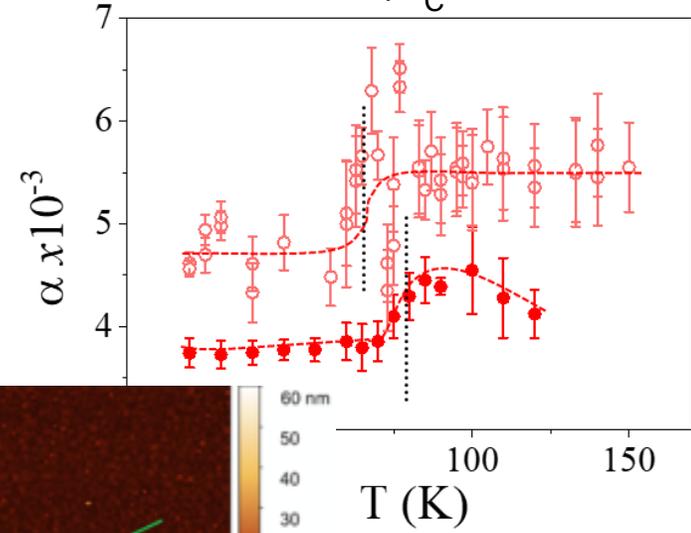
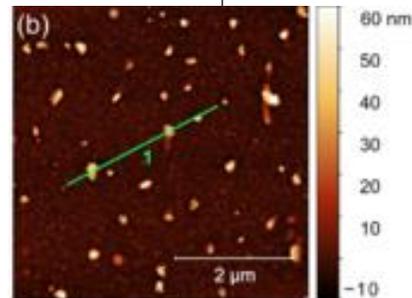
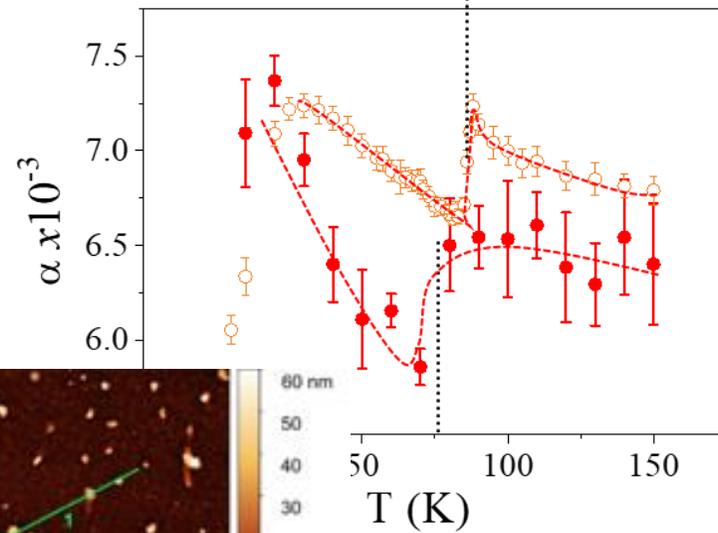
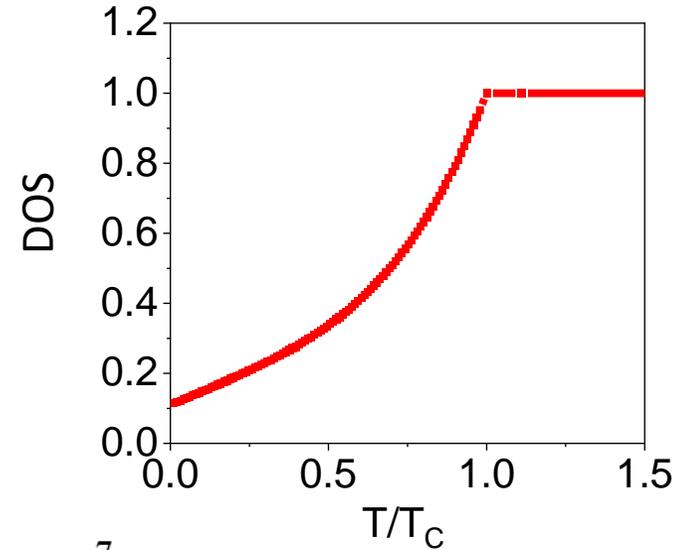
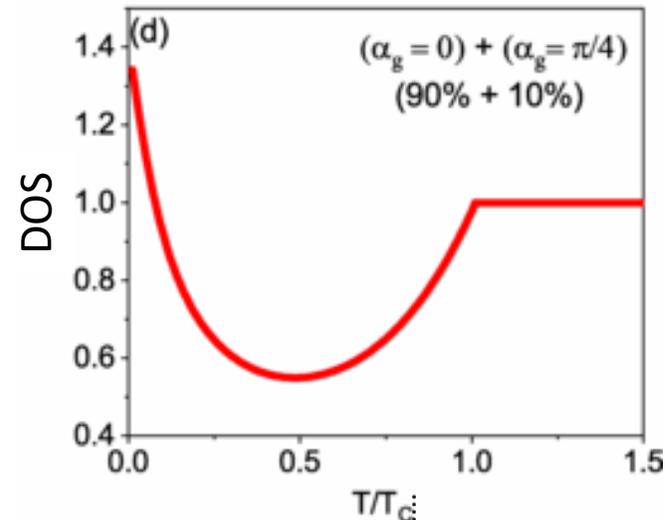
vertical surface  $\approx 100\%$



→ FLAT SURFACE IMPEDES LIMITS ACCES TO QP BOUND STATES  
 → BEHAVIOR AS WITH S-WAVE SUPERCONDUCTOR

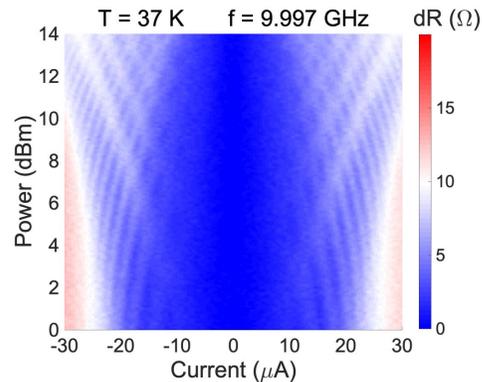
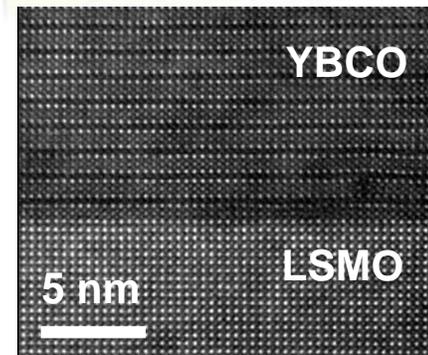
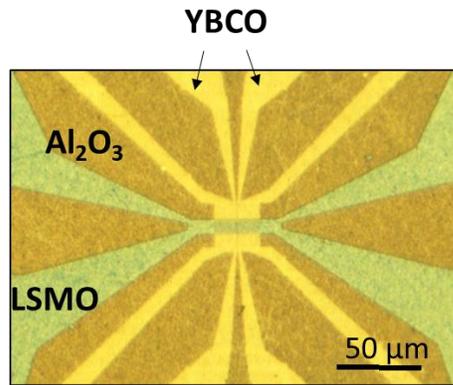
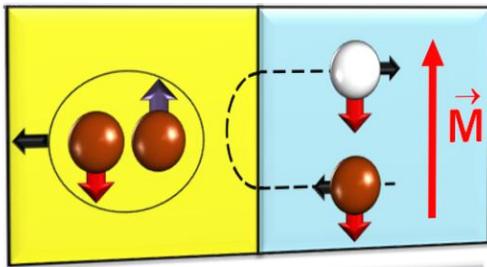
# Surface topography and effective DOS

→ UNUSUAL  
UPTURN  
EXPLAINED BY  
QP BOUND  
STATES

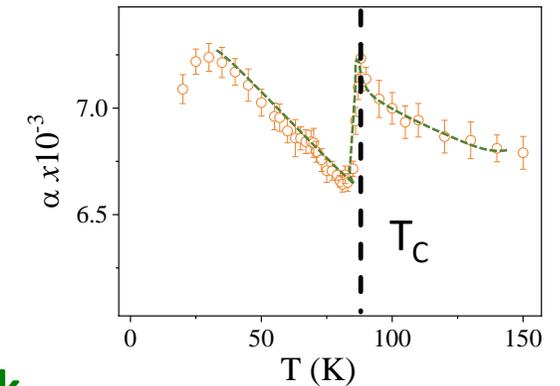
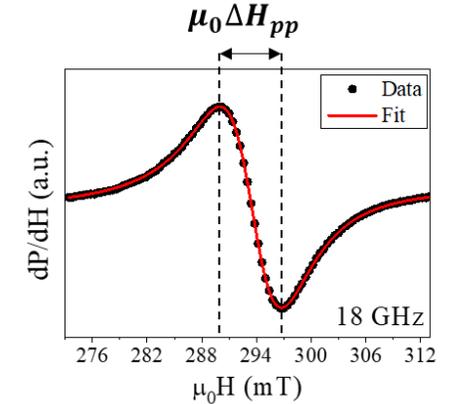
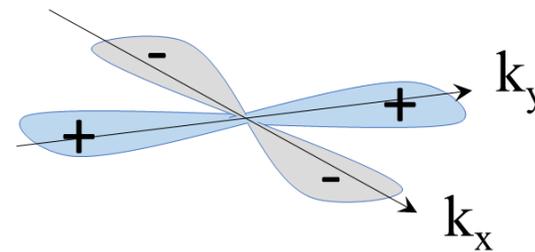
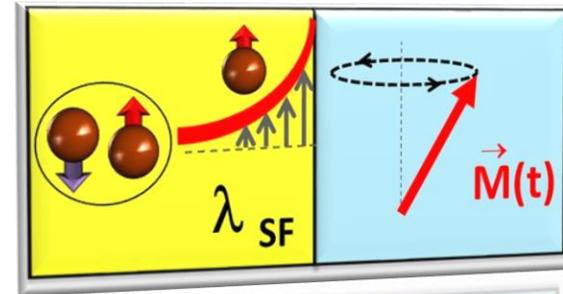


# Conclusions

## Proximity effect



## Dynamic coupling



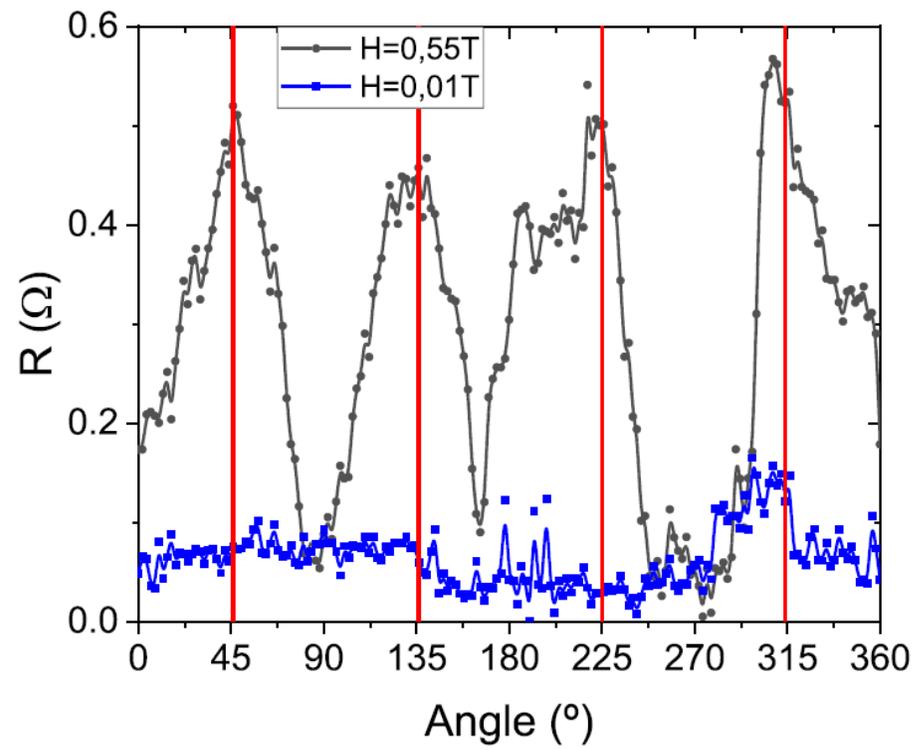
Fully spin-polarized, high- $T_C$  Josephson supercurrents  
Quantum phase coherence effects

Tunable spin sink  
d-wave effects

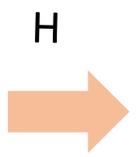
Sanchez-Manzano *et al.* Nature Materials (2022)

Carreira *et al.* PRB (2021)

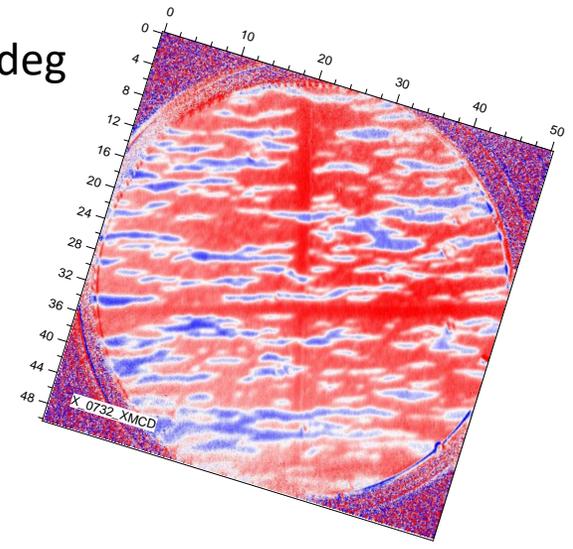
→ OPPORTUNITIES FOR HIGH- $T_C$  SUPERCONDUCTING SPINTRONICS 46



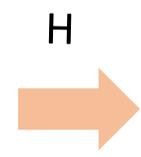
$\theta=45, 135, 225, 315$  deg



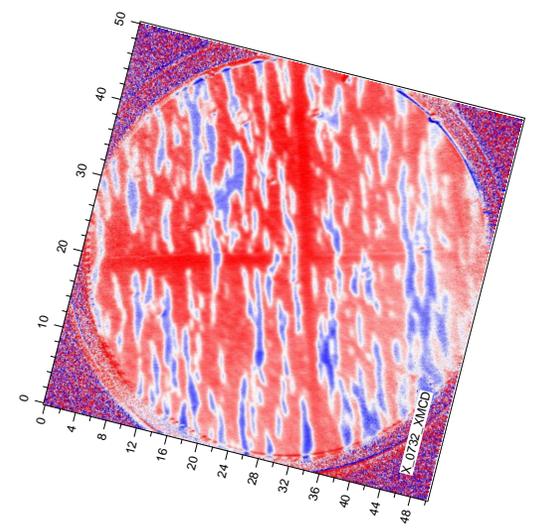
|| easy axis



$\theta=90, 180, 270, 360$  deg

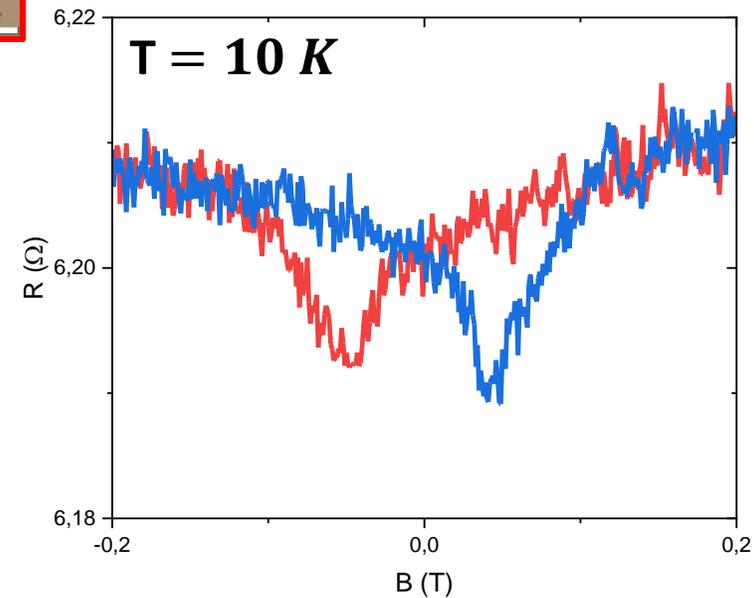
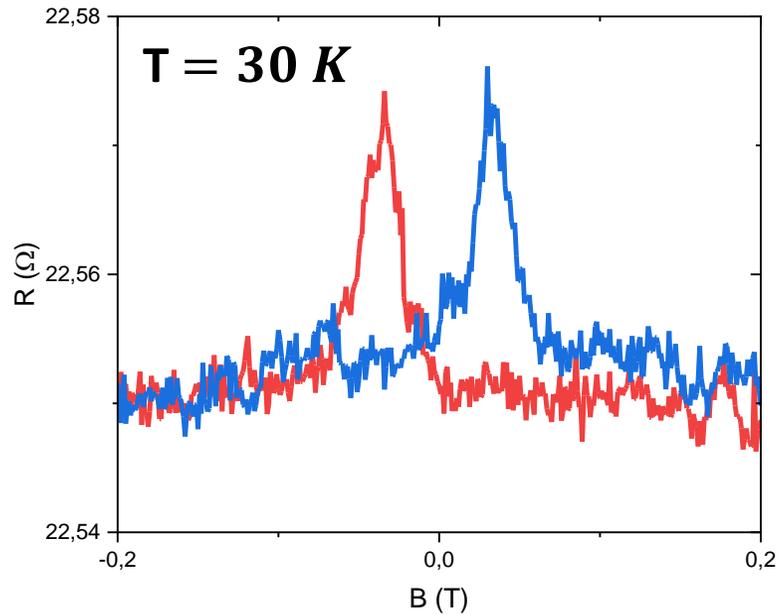
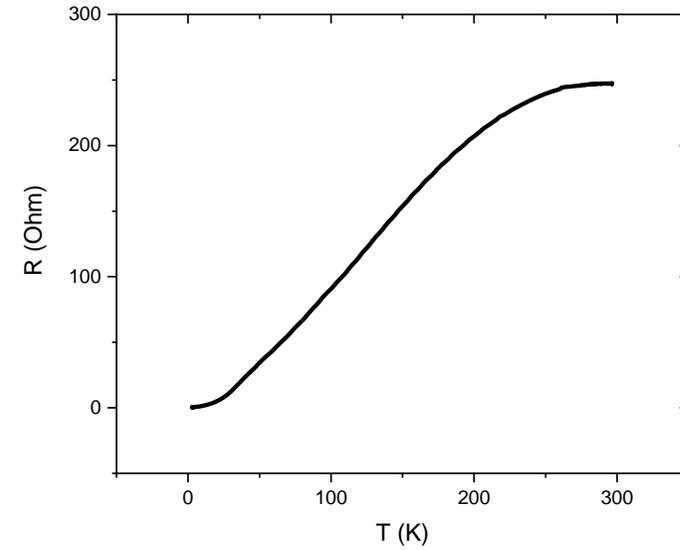
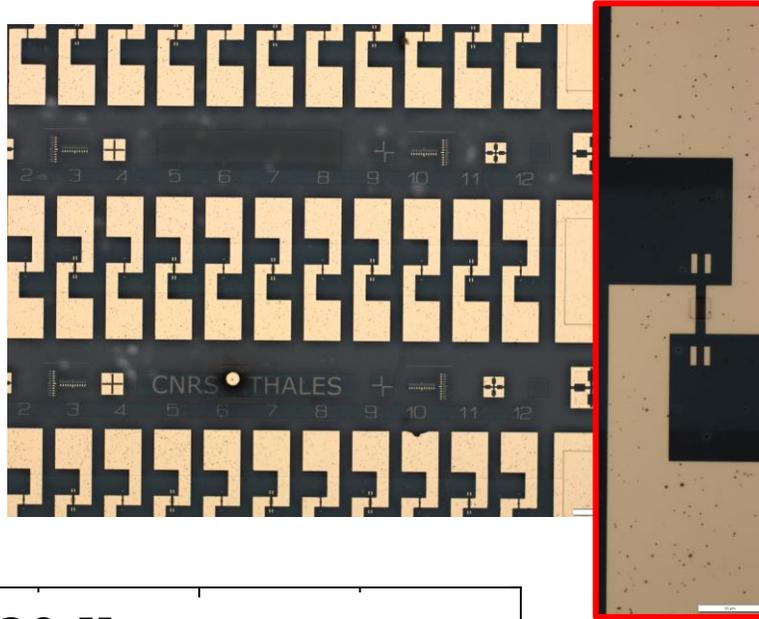
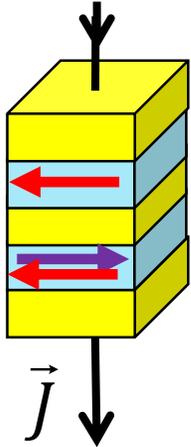


⊥ easy axis



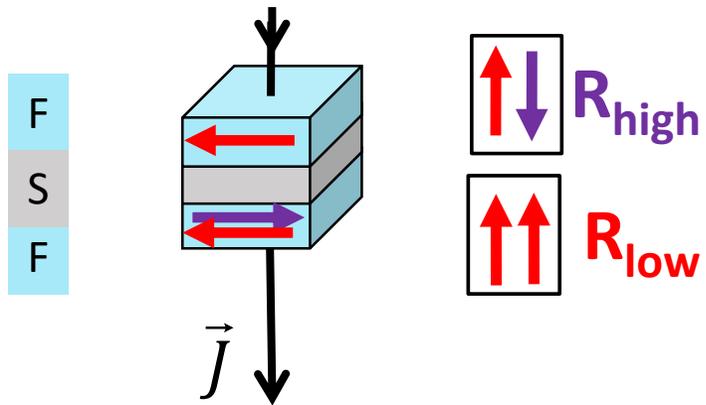
# High- $T_C$ Josephson spin valves: preliminary results

S  
F  
S  
F  
S

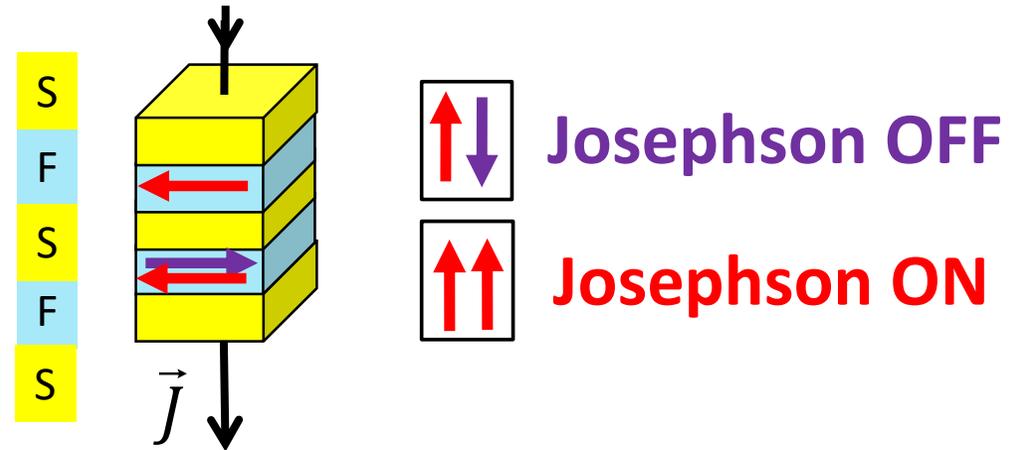


# High- $T_c$ Josephson spin valves

conventional spin valve

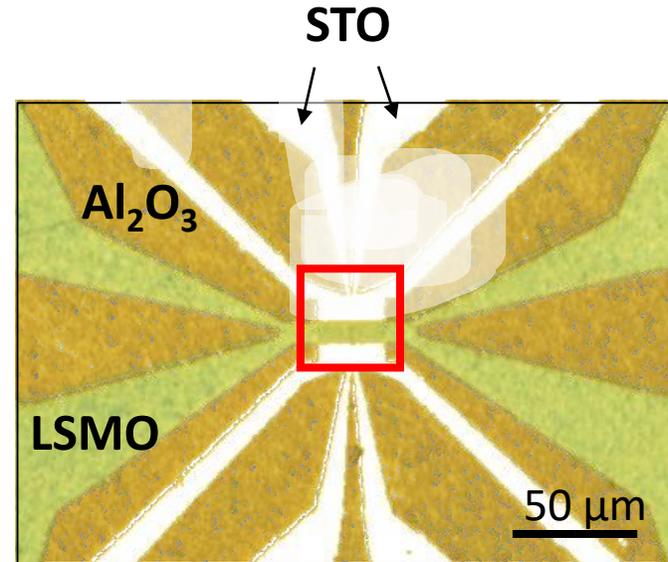
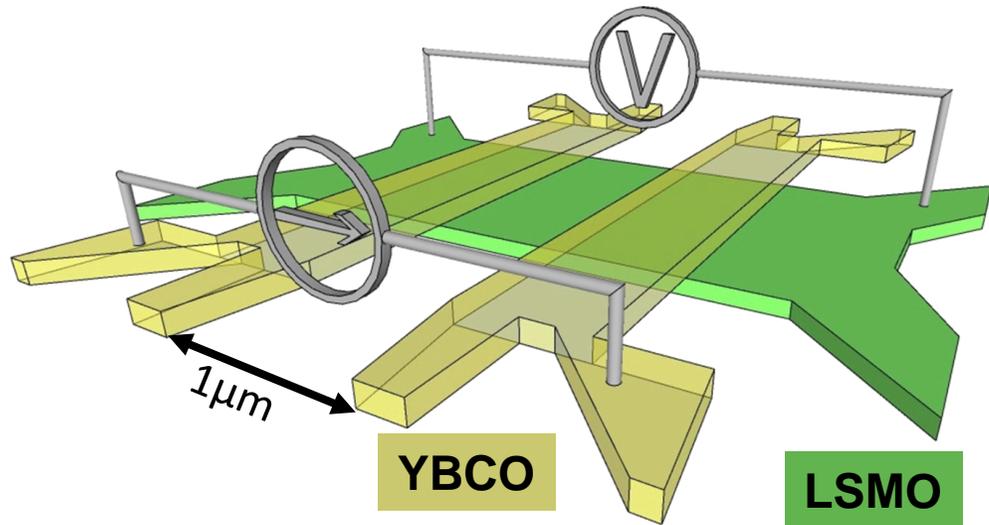


Josephson spin valve

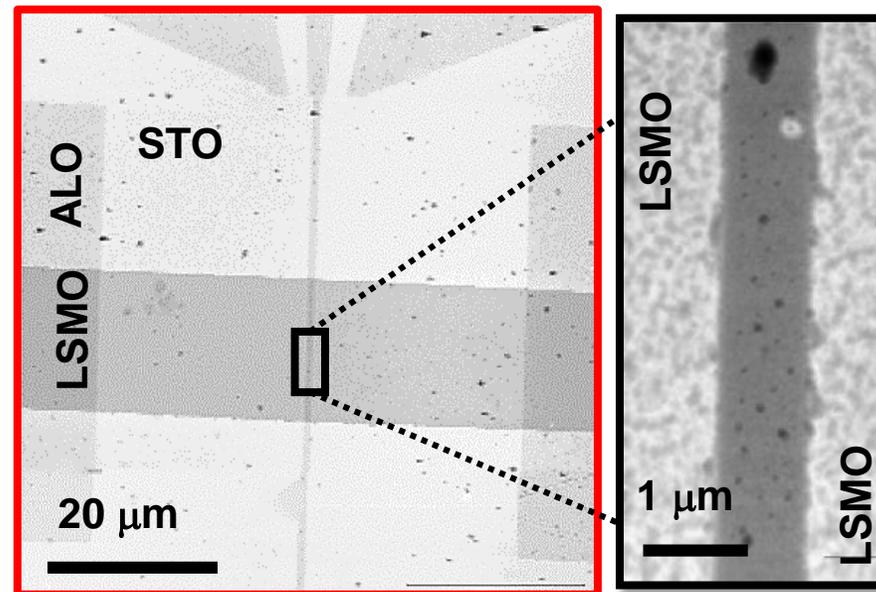


→ JOSEPHSON SPIN-VALVES... SPIN TORQUES, AND BEYOND?

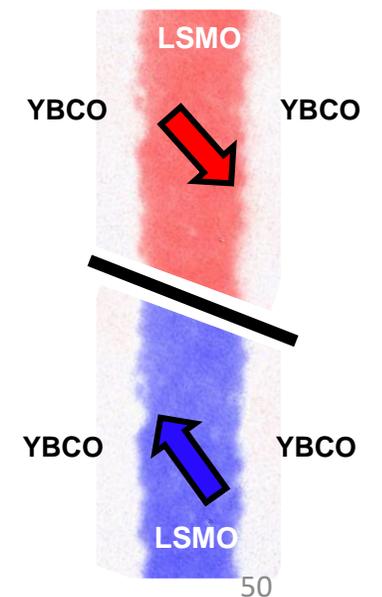
# Fabrication of planar Josephson junctions



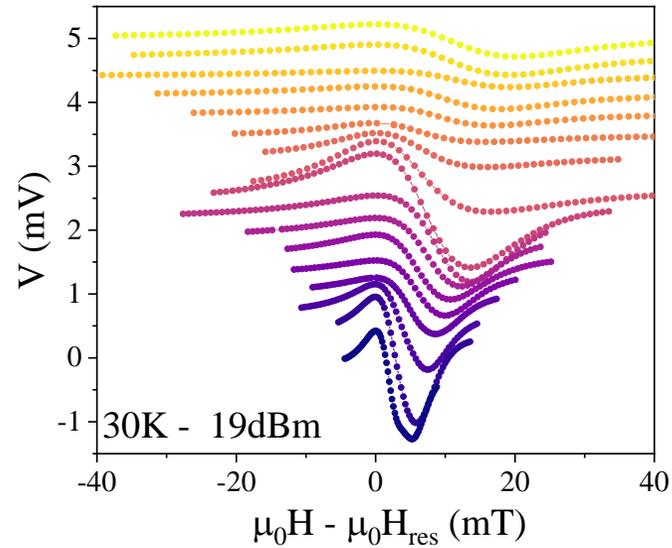
→ AMORPHOUS ALUMINA MASK  
→ LSMO REMAINS MAGNETIC



XMCD-PEEM



# FMR experiments – Damping and inhomogeneous broadening

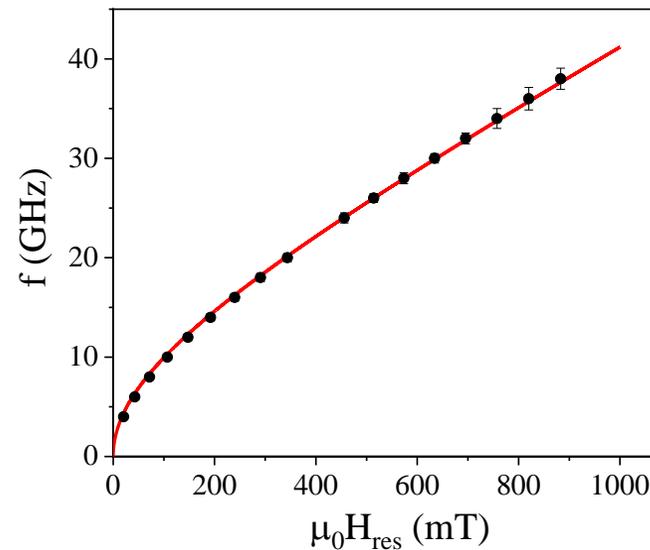
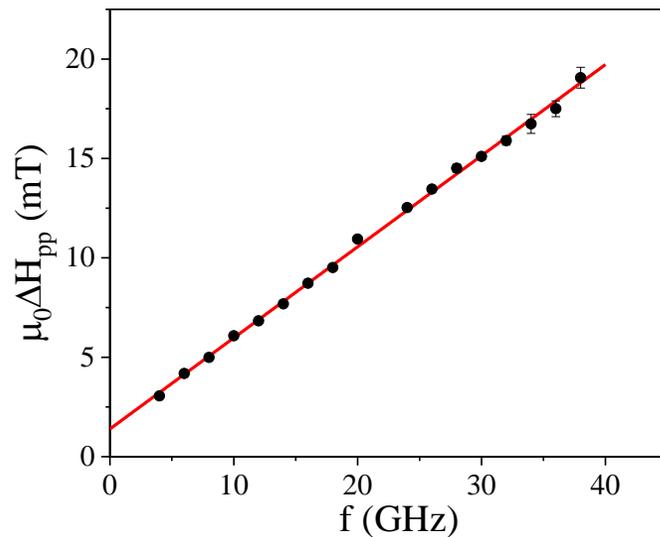


$$\Delta H_{pp} = \frac{2\alpha}{\sqrt{3}\gamma\mu_0} f + \Delta H_0$$

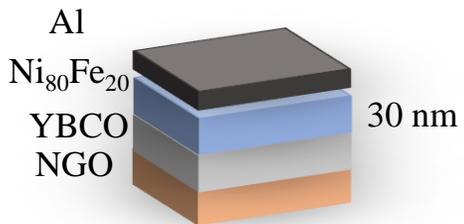
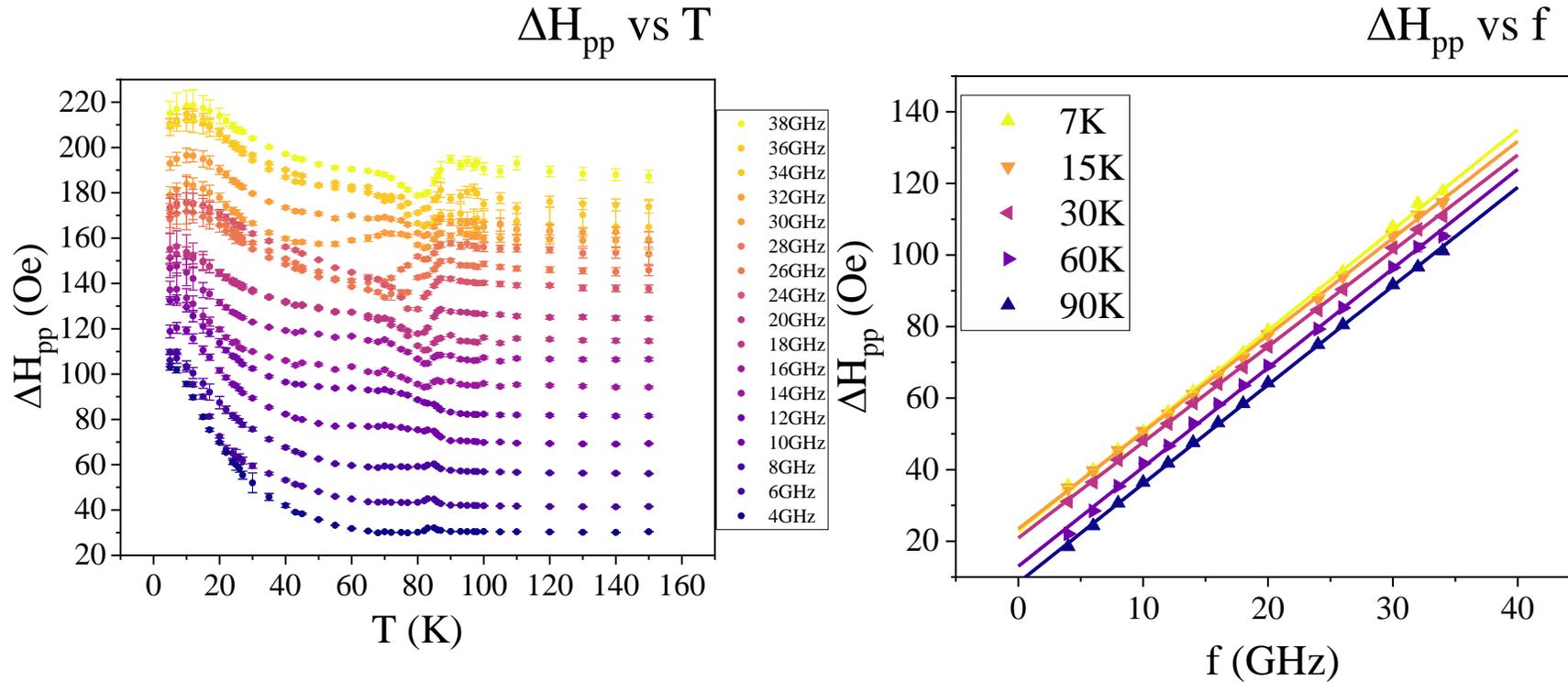
$$f = \gamma\mu_0 \sqrt{H_{res}(H_{res} + M_{eff})}$$

$\alpha$  : Gilbert damping

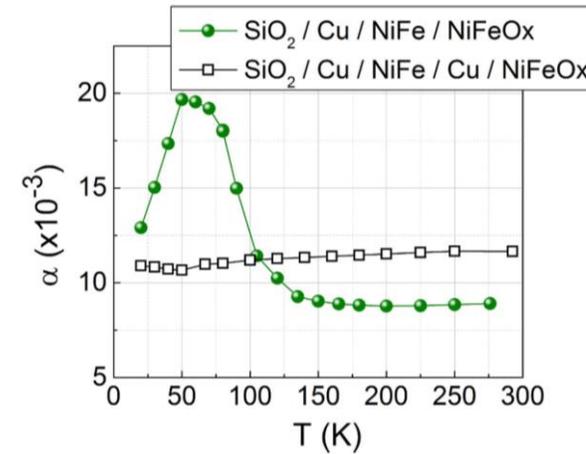
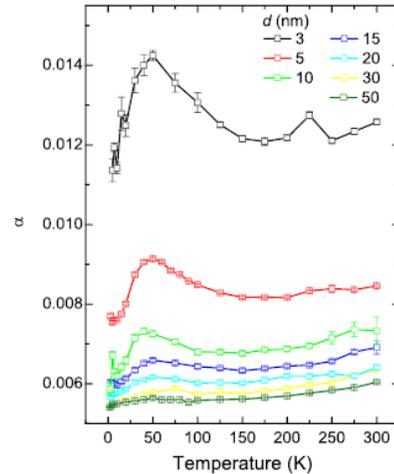
$\Delta H_0$ : inhomogeneous broadening



# Linewidth vs T and vs f for the sample grown on NGO



# Intrinsic broadening of the linewidth at low temperatures in NiFe thin films



Y. Zhao et al, *Sci. Rep.* 6, 22890 (2016)

L. Frangou et al. *Phys. Rev. B* **95**, 054416 (2017)

Magnetic inhomogeneities at the interface due to the oxidation of the Py



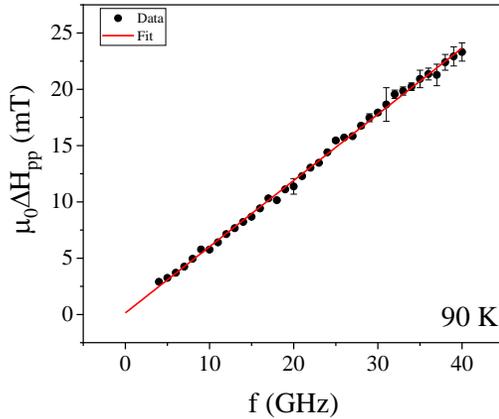
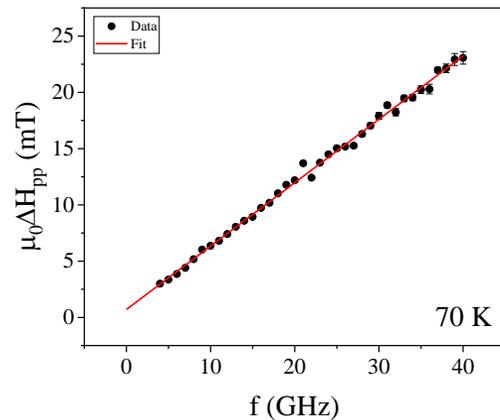
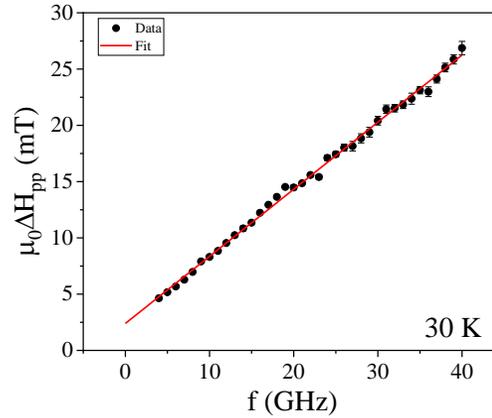
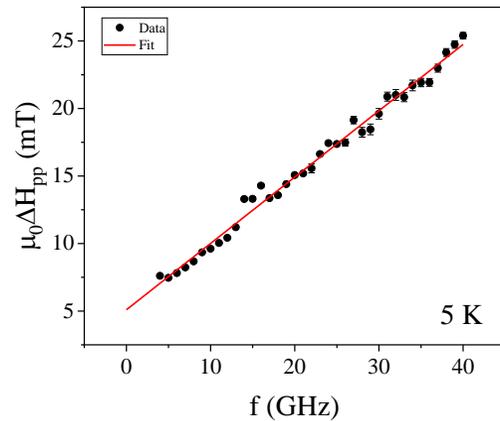
Opening of extrinsic relaxation channels



Linewidth increases at low temperatures



# Ferromagnetic resonance experiments – Damping calculation

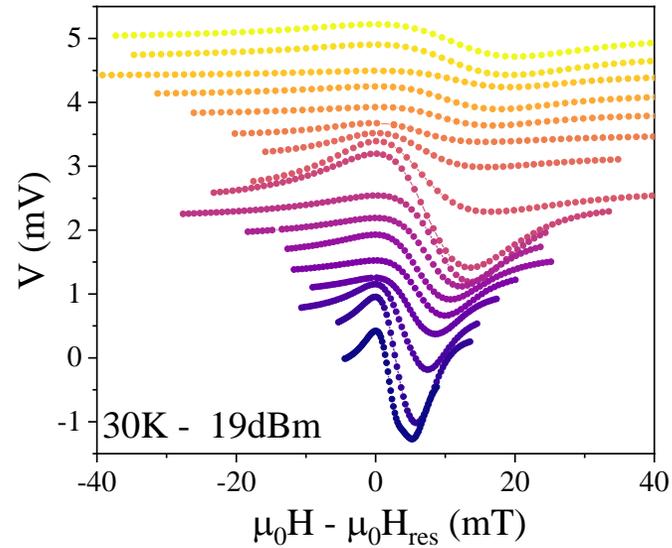


$$\Delta H_{pp} = \frac{2\alpha}{\sqrt{3}\gamma\mu_0} f + \Delta H_0$$

$\alpha$  : Gilbert damping

$\Delta H_0$ : inhomogeneous broadening

# FMR experiments – Damping and inhomogeneous broadening

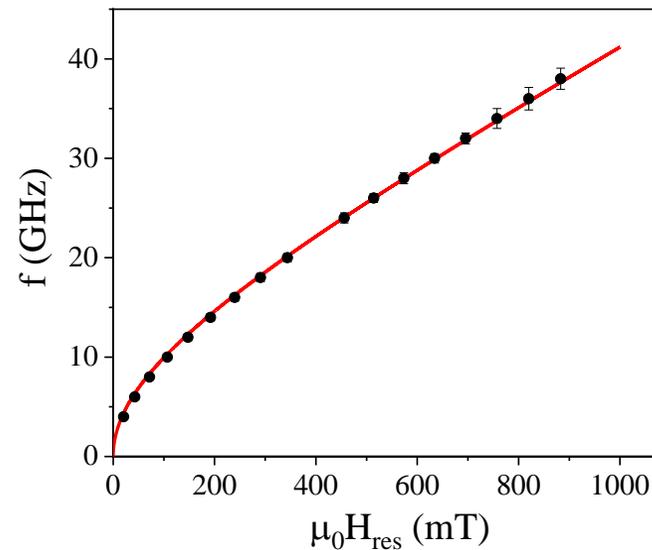
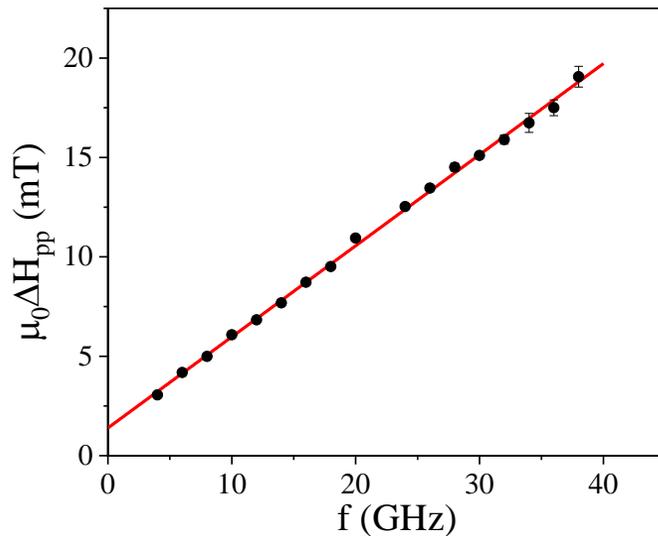


$$\Delta H_{pp} = \frac{2\alpha}{\sqrt{3}\gamma\mu_0} f + \Delta H_0$$

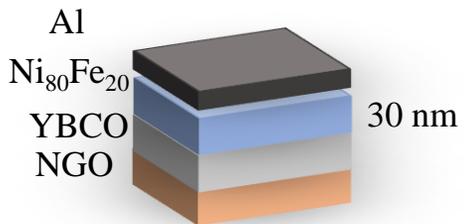
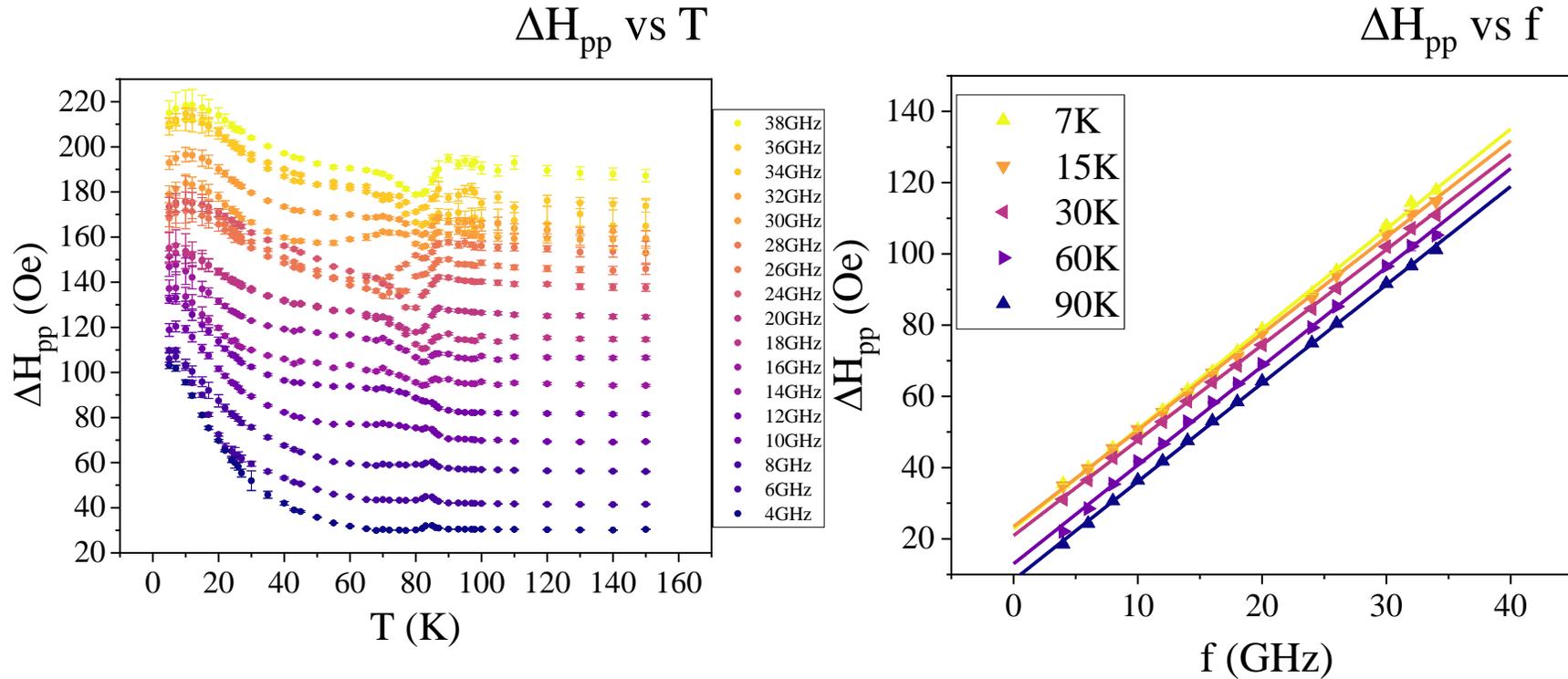
$$f = \gamma\mu_0 \sqrt{H_{res}(H_{res} + M_{eff})}$$

$\alpha$  : Gilbert damping

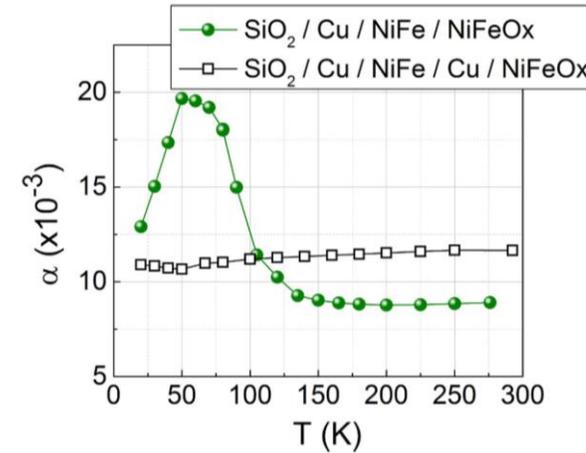
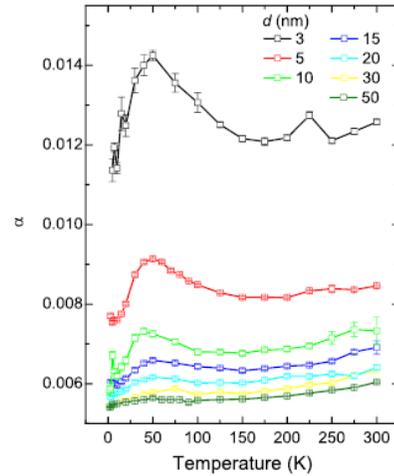
$\Delta H_0$ : inhomogeneous broadening



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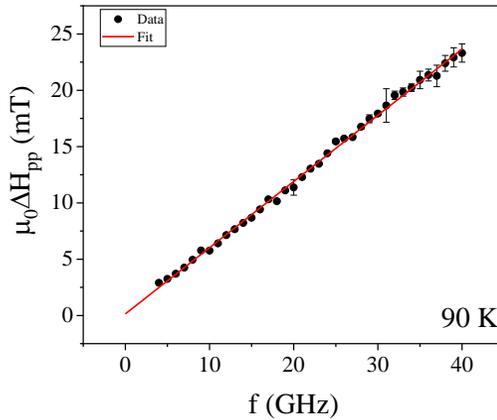
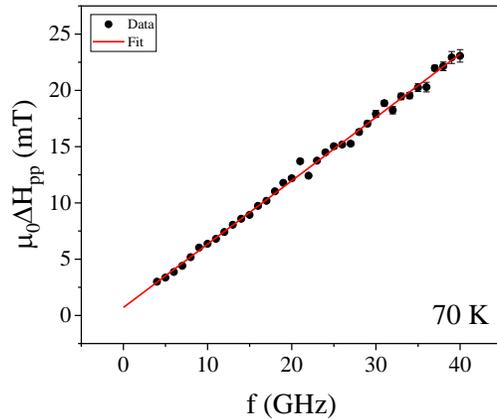
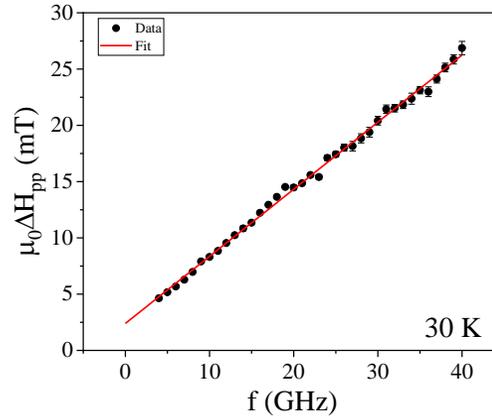
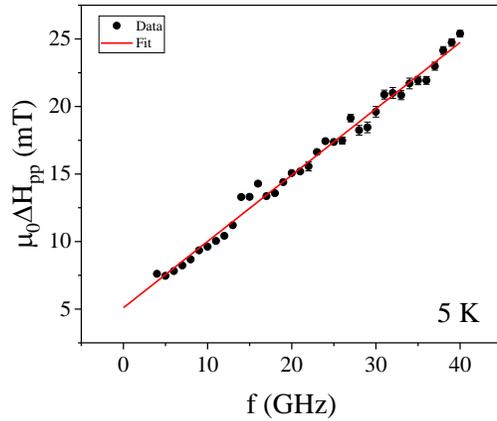
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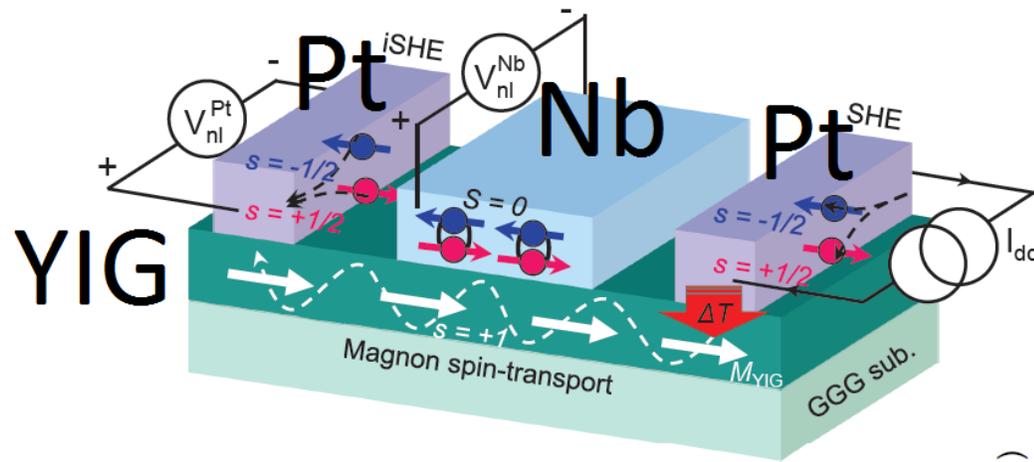


$$\Delta H_{pp} = \frac{2\alpha}{\sqrt{3}\gamma\mu_0} f + \Delta H_0$$

$\alpha$  : Gilbert damping

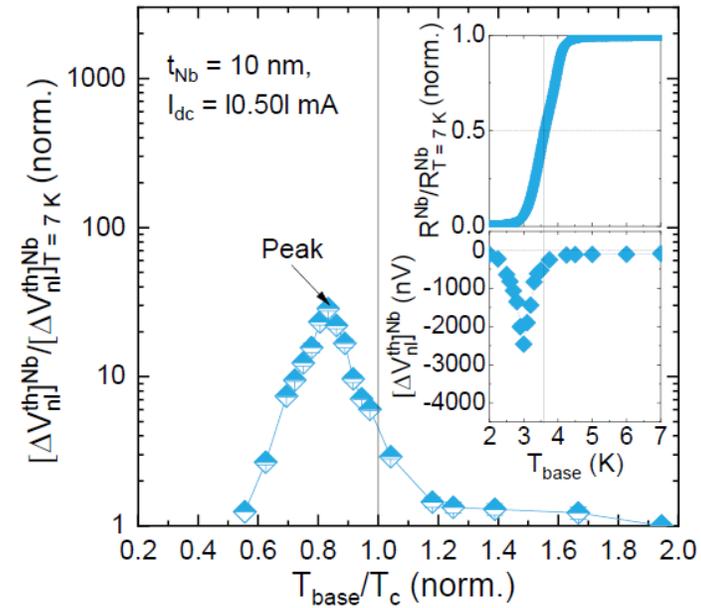
$\Delta H_0$ : inhomogeneous broadening

# Different designs to study spin injection in S/F hybrids



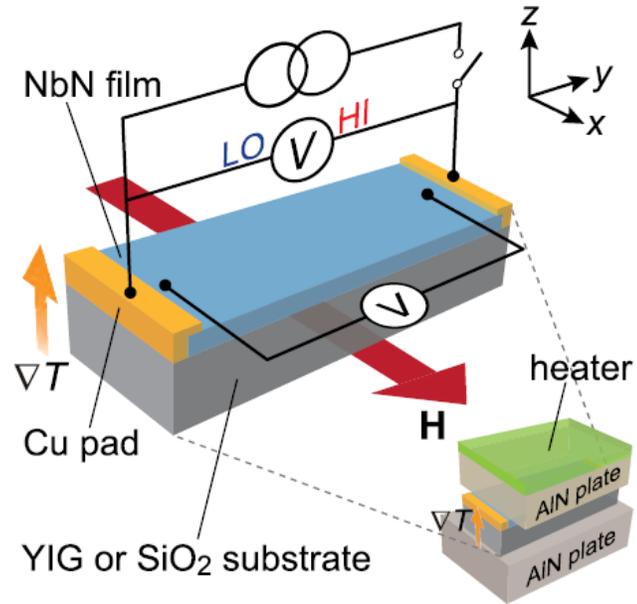
Spin injection efficiency in Nb increases below  $T_c$ !

Non-local spin injection devices.  
Inject angular momentum using magnons instead of spin polarized electrons

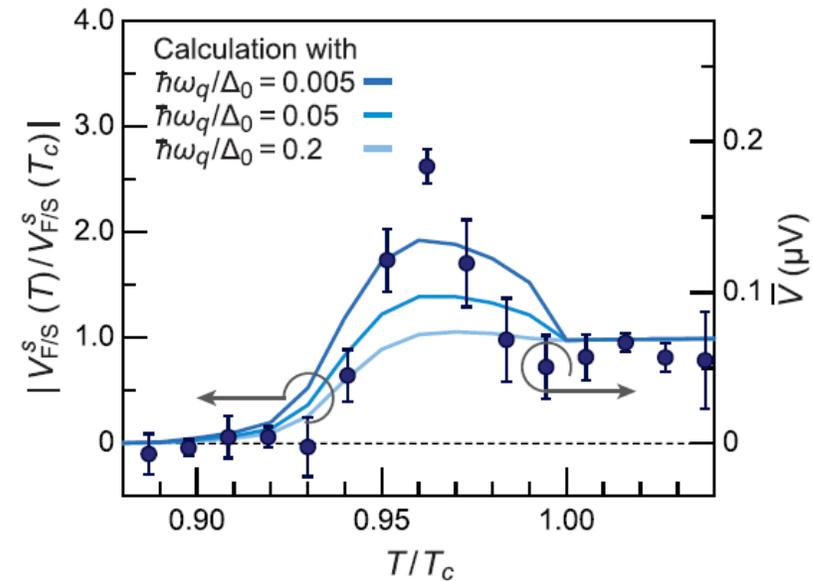


K.-R. Jeon *et al.* [arXiv:2004.09467](https://arxiv.org/abs/2004.09467) (2020)

# Different designs to study spin injection in S/F hybrids



Spin injection using a thermal gradient  
(Spin Seebeck effect)



M. Umeda *et al.* *Appl. Phys. Lett.* **112**, 232601 (2018)