

Autumn College on Non-Equilibrium Quantum Systems  
May 2-13, 2011  
Buenos Aires, Argentina

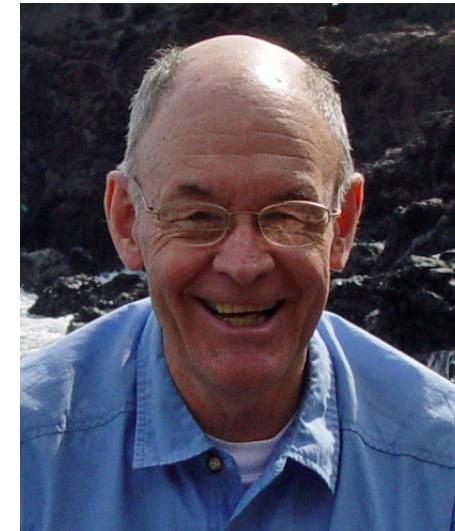
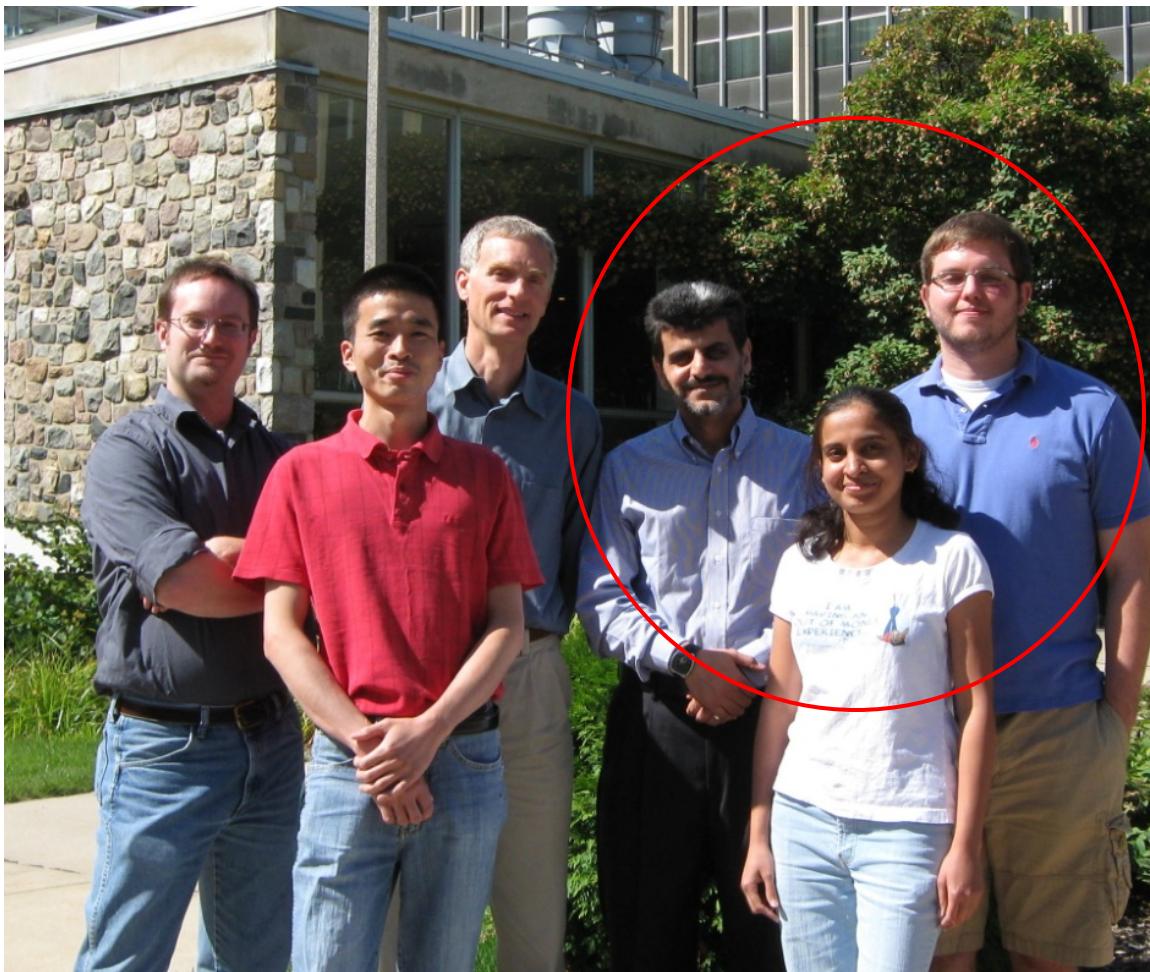
Norman Birge\*, Michigan State University

Lecture III: Spin-triplet supercurrent in  
ferromagnetic Josephson junctions

\* Work supported by DOE BES



Collaborators: Trupti Khaire, Mazin Khasawneh, Caroline Klose  
Hamood Arham, Kurt Boden, William P. Pratt, Jr.



# Prologue: Fermion pairing

- Condensed Matter Physics
  - superconductivity & superfluidity ( ${}^3\text{He}$ )
- Nuclear Physics
  - nucleon pairing (even-odd energy differences)
- Astrophysics
  - superfluidity in neutron stars
- Atomics Physics
  - BEC to BCS crossover in cold atomic gases  
*(see lectures by Christophe Salomon)*

# Prologue:

## What we remember from quantum mechanics

Wavefunction for two identical fermions (Spin-Statistics Thm or Pauli Exclusion)

$$\Psi(\vec{r}_1, s_1; \vec{r}_2, s_2) = -\Psi(\vec{r}_2, s_2; \vec{r}_1, s_1)$$

A convenient basis:

$$\Psi(\vec{r}_1, s_1; \vec{r}_2, s_2) = \Phi(\vec{r}_1, \vec{r}_2) \chi(s_1, s_2)$$

Two possibilities:

1.  $\Phi_S(\vec{r}_2, \vec{r}_1) = \Phi_S(\vec{r}_1, \vec{r}_2)$  and  $\chi_A(s_2, s_1) = -\chi_A(s_1, s_2)$
2.  $\Phi_A(\vec{r}_2, \vec{r}_1) = -\Phi_A(\vec{r}_1, \vec{r}_2)$  and  $\chi_S(s_2, s_1) = \chi_S(s_1, s_2)$

# Prologue:

## What we remember from quantum mechanics

Example: two electrons in free space:  $\Phi_{n,l}(\vec{r}_1, \vec{r}_2) = R_{n,l}(r)Y_l^m(\theta, \phi)$   $\vec{r} = \vec{r}_1 - \vec{r}_2$

Spatial state is eigenstate of orbital  $L^2$ :

$$\text{even } \ell \Rightarrow \Phi_S(\vec{r}_2, \vec{r}_1) = \Phi_S(\vec{r}_1, \vec{r}_2)$$

$$\text{odd } \ell \Rightarrow \Phi_A(r_2, r_1) = -\Phi_A(r_1, r_2)$$

Spin state is eigenstate of total spin  $S^2$ :  $\vec{S} = \vec{S}_1 + \vec{S}_2$

$$s=1 \text{ (triplet)} \Rightarrow \chi_s(s_2, s_1) = \chi_s(s_1, s_2)$$

$$s=0 \text{ (singlet)} \Rightarrow \chi_A(s_2, s_1) = -\chi_A(s_1, s_2)$$

$$\chi(s=1) = \begin{cases} |\uparrow\uparrow\rangle \\ \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle) \\ |\downarrow\downarrow\rangle \end{cases}$$

$$\chi(s=0) = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$$

# Allowed Cooper pair symmetries

$$\Psi(\vec{r}_1, s_1; \vec{r}_2, s_2) = -\Psi(\vec{r}_2, s_2; \vec{r}_1, s_1)$$

	$s=0$	$s=1$
$\ell=0$ (s-wave)	BCS	X
$\ell=1$ (p-wave)	X	superfluid ${}^3\text{He}$ $\text{Sr}_2\text{RuO}_4$
$\ell=2$ (d-wave)	high- $T_c$	X

# Proposal by Berezinskii (1974):

## New model of the anisotropic phase of superfluid He<sup>3</sup>

V. L. Berezinskii

(Submitted September 12, 1974)

ZhETF Pis. Red. 20, No. 9, 628–631 (November 5, 1974)

It is shown at a sufficiently large value of the exchange gain in He<sup>3</sup>, there exists a possibility of pairing in states with spin  $s = 1$  with even orbital angular momenta. This does not contradict the Pauli principle, since the average many-time wave functions of the pairs (the anomalous Green's function) depends in odd manner on the difference between the temporal arguments.

$F(r_1, r_2, s_1, s_2, t_1, t_2)$  ≡ “anomalous Green's function” = pair correlation function

What does this mean?

Fermions require:  $F(\vec{r}_1 - \vec{r}_2; s_1, s_2; t_1 - t_2) = -F(\vec{r}_2 - \vec{r}_1; s_2, s_1; t_2 - t_1)$

$F$  is odd in time:  $F(\vec{r}_1 - \vec{r}_2; s_1, s_2; t_1 - t_2) = -F(\vec{r}_1 - \vec{r}_2; s_1, s_2; t_2 - t_1)$

$F$  is even under  
exchange of space  $F(\vec{r}_1 - \vec{r}_2; s_1, s_2; t_1 - t_2) = +F(\vec{r}_2 - \vec{r}_1; s_2, s_1; t_1 - t_2)$   
and spin:

# Allowed Cooper pair symmetries

	$s=0$	$s=1$
$\ell=0$	BCS	X
$\ell=1$	X	superfluid $^3\text{He}$ $\text{Sr}_2\text{RuO}_4$
$\ell=2$	high- $T_c$	X



= allowed if correlation function is odd  
under time-reversal (or odd in frequency)

# Allowed Cooper pair symmetries

	$s=0$	$s=1$
$\ell=0$	BCS	Berezinski model for ${}^3\text{He}$ (1974)
$\ell=1$	Balatsky & Abrahams ('92)	superfluid ${}^3\text{He}$ $\text{Sr}_2\text{RuO}_4$
$\ell=2$	high- $T_c$	<b>X</b>



= allowed if correlation function is odd  
under time-reversal (or odd in frequency)

# Allowed Cooper pair symmetries

	$s=0$	$s=1$
$\ell=0$	BCS	S/F
$\ell=1$	S/F	superfluid $^3\text{He}$ $\text{Sr}_2\text{RuO}_4$
$\ell=2$	high- $T_c$	S/F

especially  
interesting

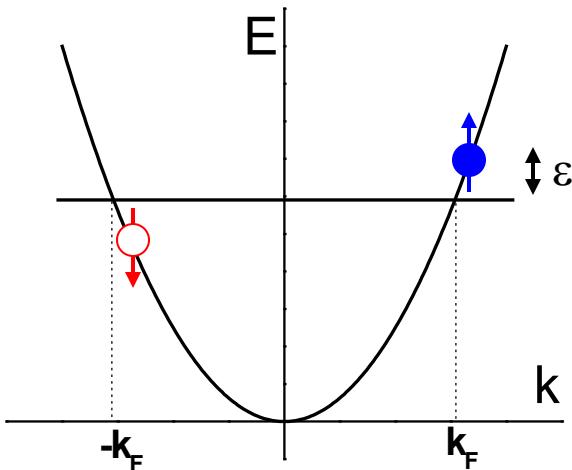
Bergeret, Volkov & Efetov, PRL 90, 117006 (2003); RMP 77, 1321 (2005)

# Outline

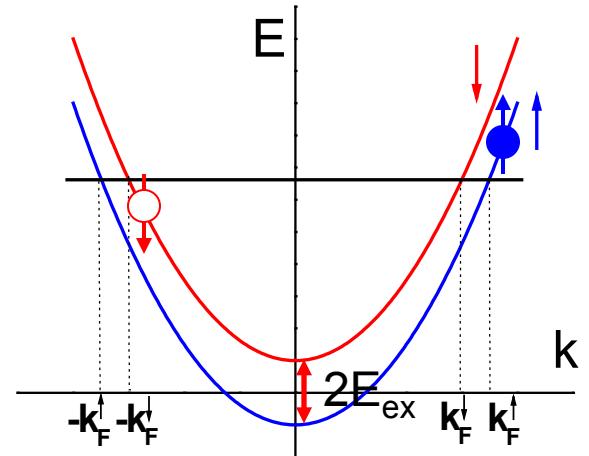
- Proximity effect in S/N and S/F systems
- Prediction: Spin-triplet pair correlations
- Our search: S/F/S Josephson junctions
  - PdNi – a weak ferromagnet
  - Co/Ru/Co – a synthetic antiferromagnet
  - Combining the best of both materials
- Conclusions and Future Prospects

# Andreev Reflection: N/S vs. F/S

S/N



S/F



Coherence time between electron and hole:  $\tau = \frac{\hbar}{2\varepsilon}$

$$k_F^\uparrow - k_F^\downarrow \equiv Q = 2 E_{ex} / \hbar v_F$$

$$\xi_N = v_F \tau = \frac{\hbar v_F}{2\pi k_B T} \quad \text{ballistic}$$

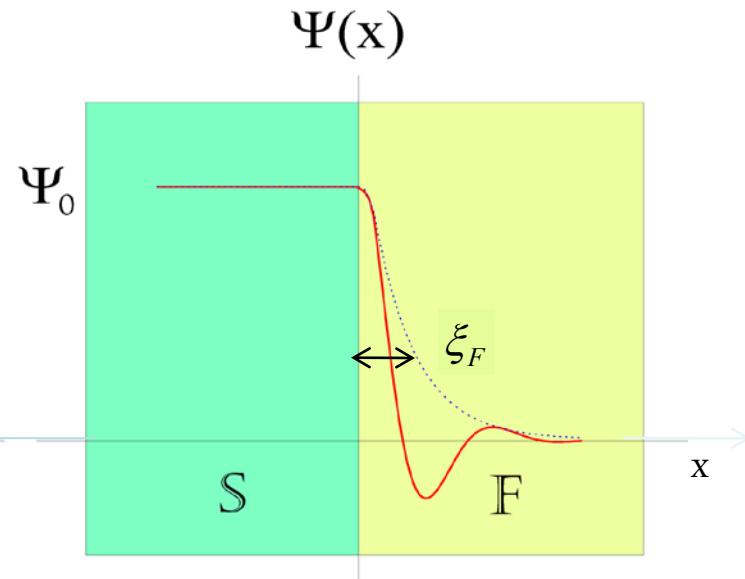
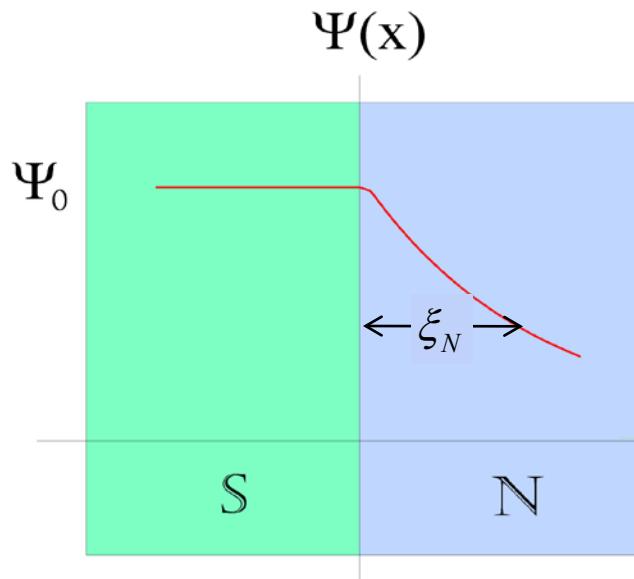
$$\xi_N = \sqrt{D_N \tau} = \sqrt{\frac{\hbar D_N}{2\pi k_B T}} \quad \text{diffusive}$$

$D$  = diffusion constant

$$\xi_F = Q^{-1} = \frac{\hbar v_F}{2 E_{ex}} \quad \text{ballistic}$$

$$\xi_F = \sqrt{\frac{\hbar D_F}{E_{ex}}} \quad \text{diffusive}$$

# Proximity effect: S/N vs. S/F



$$\xi_N = \sqrt{\frac{\hbar D_N}{2\pi k_B T}} \approx \text{few } \mu m$$

$$\xi_F \sim \sqrt{\frac{\hbar D_F}{E_{ex}}} \approx \text{few } nm$$

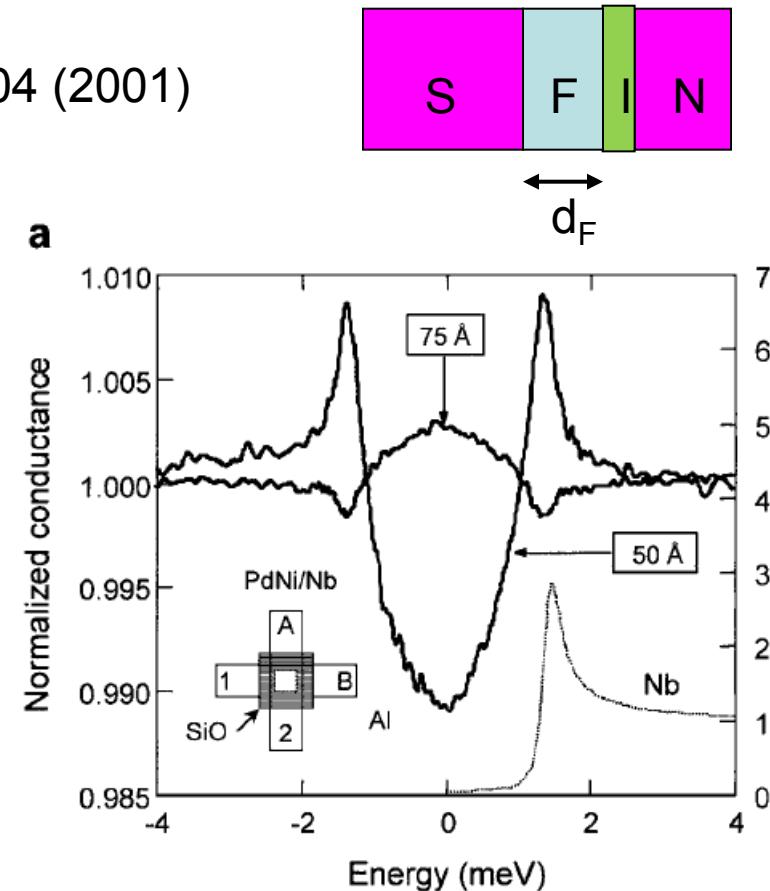
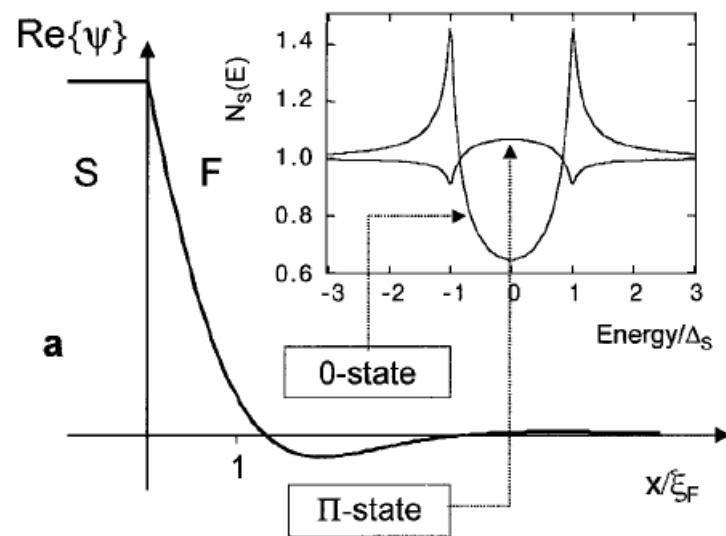
$$\Psi(x) = \Psi_0 \exp(-x/\xi_N)$$

$$\Psi(x) = \Psi_0 \cos(x/\xi_F) \exp(-x/\xi_F)$$

# How to detect the oscillating pair correlation function?

1. Measure tunneling density of states in S/F/I/N structure, as function of  $d_F$

Kontos, Aprili, Lesueur, Grison, PRL 86, 304 (2001)

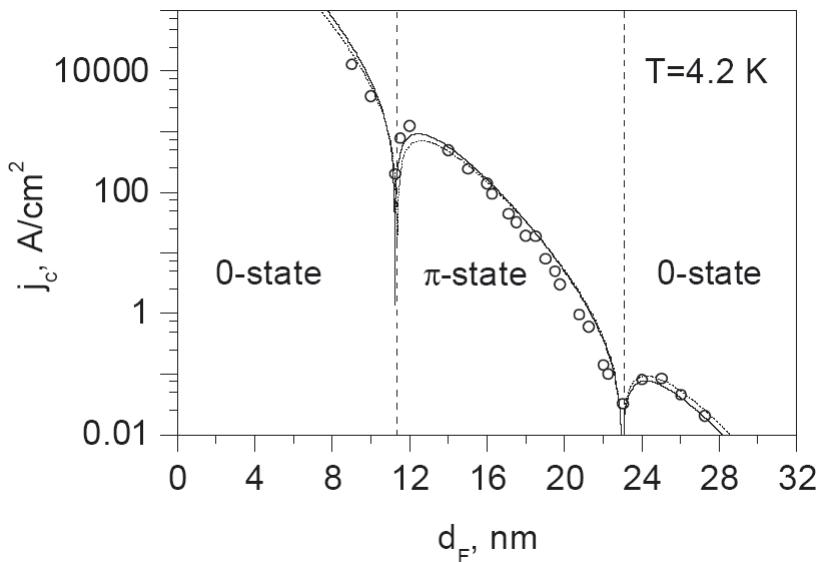


# How to detect the oscillating pair correlation function?

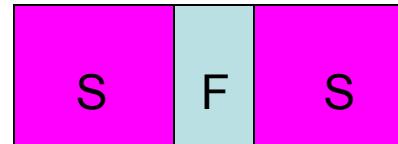
2. Measure critical current of S/F/S Josephson junction, as function of  $d_F$

$$\text{0-state: } I_s = I_c \sin(\phi_2 - \phi_1)$$
$$\pi\text{-state: } I_s = I_c \sin(\phi_2 - \phi_1 + \pi)$$

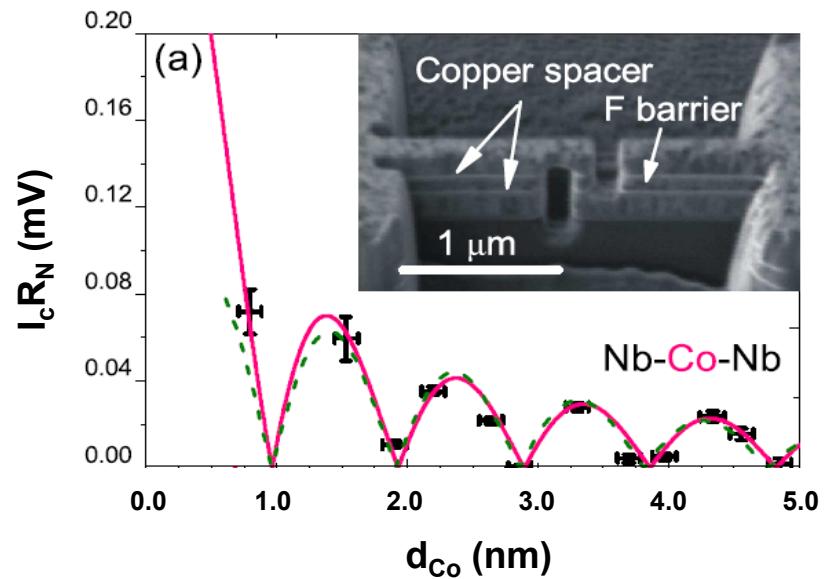
Weak F: Cu<sub>48</sub>Ni<sub>52</sub> alloy



Ryazanov *et al.*, PRL **86**, 2427 (2001);  
**96**, 197003 (2006).

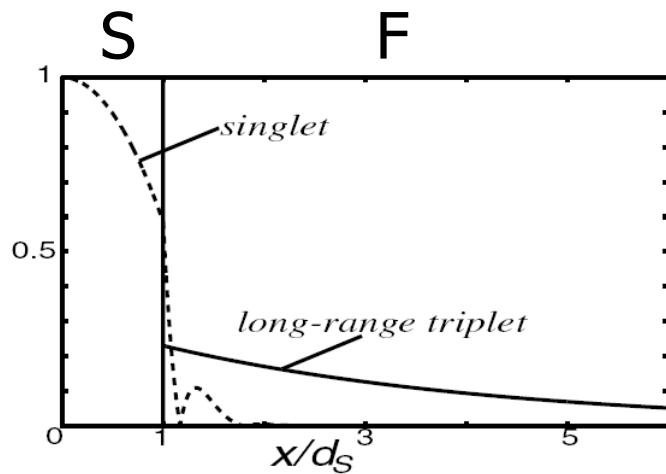
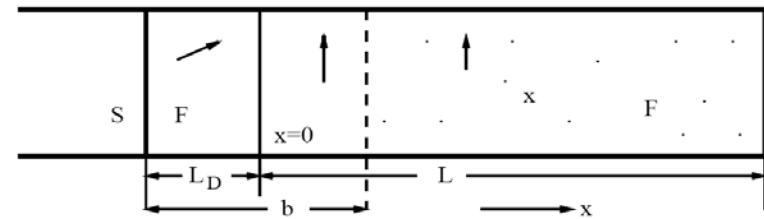
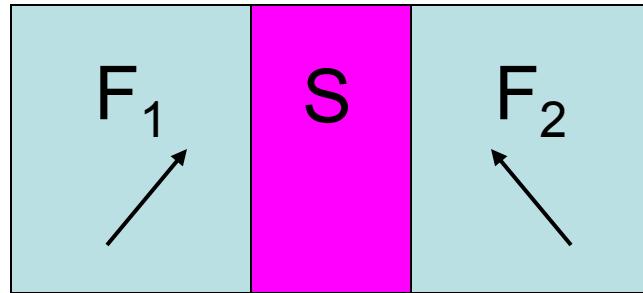


Strong F: Co



Robinson, Piano, Burnell, Bell, Blamire,  
PRL **97**, 177003 (2005)

# Prediction: long-range spin-triplet pair correlations induced by noncollinear magnetization



Singlet

$$\xi_F^S = \sqrt{\frac{\hbar D_F}{E_{ex}}}$$

Triplet

$$\xi_F^T = \sqrt{\frac{\hbar D_F}{2\pi k_B T}}$$

$$\boxed{\xi_F^S \ll \xi_F^T}$$

Bergeret, Volkov & Efetov, Phys. Rev. B 64, 134506 (2001); PRL **90**, 117006 (2003)

Kadrigrobov, Shekhter & Jonson, Europhys. Lett. **54**, 394 (2001)

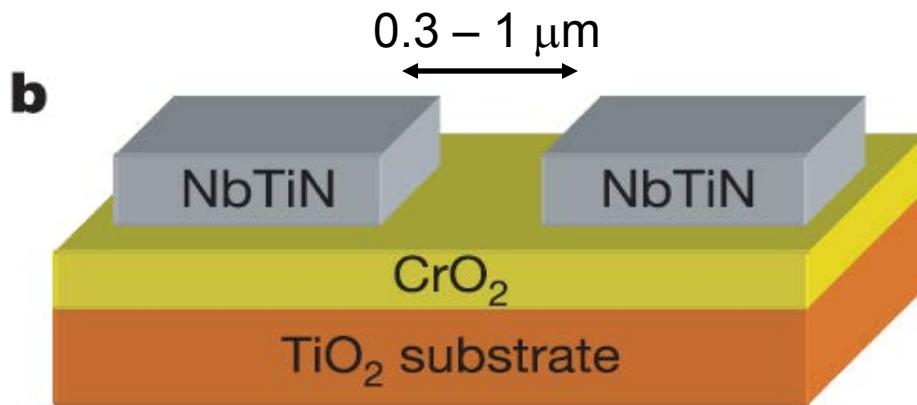
# Features of odd-frequency, spin-triplet pair correlations

	$S=0$	$S=1$
$\ell=0$	BCS	S/F
$\ell=1$	S/F	$\text{Sr}_2\text{RuO}_4$
$\ell=2$	high- $T_c$	S/F

- $s=1$ : pairs not subject to  $E_{\text{ex}}$   
 $\Rightarrow$  long-range penetration in F
- $\ell=0$ : insensitive to disorder

# Possible observation of triplet:

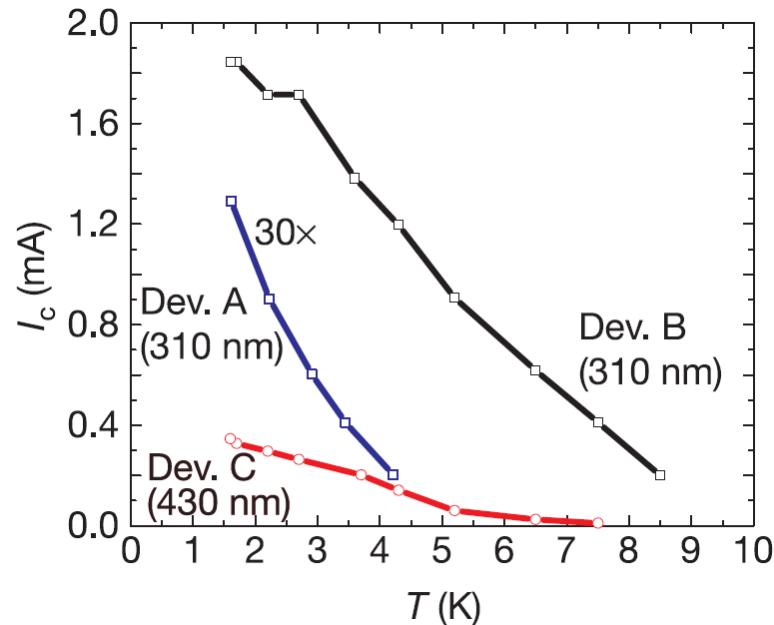
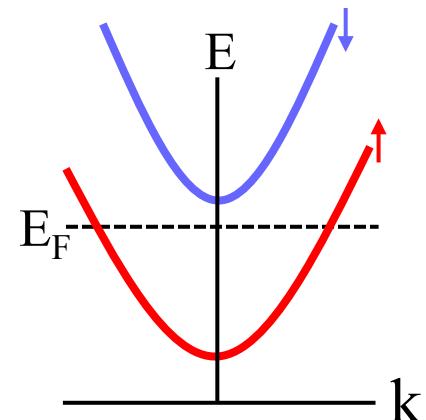
Keizer, Goennenwein, Klapwijk, Miao, Xiao, Gupta,  
Nature **439**, 825 (2006)



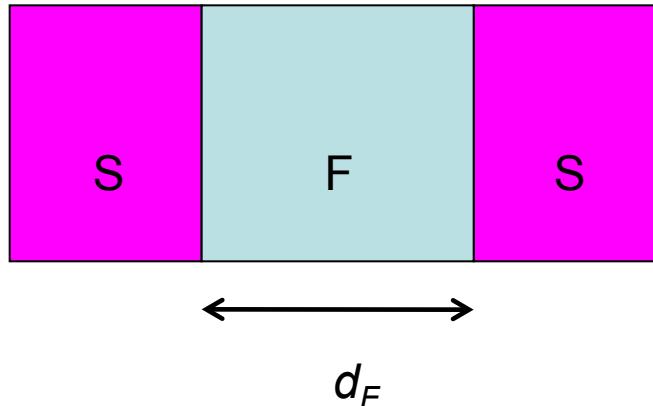
Long-range propagation of supercurrent,  
but large sample-to-sample variations in  $I_c$ .

*Reproduced last year by J. Aarts*

CrO<sub>2</sub> is a “half metal”



# Our approach: Systematic study of S/F/S junctions:



$$\xi_F^S \ll d_F \ll \xi_F^T \leq l_{sf}$$

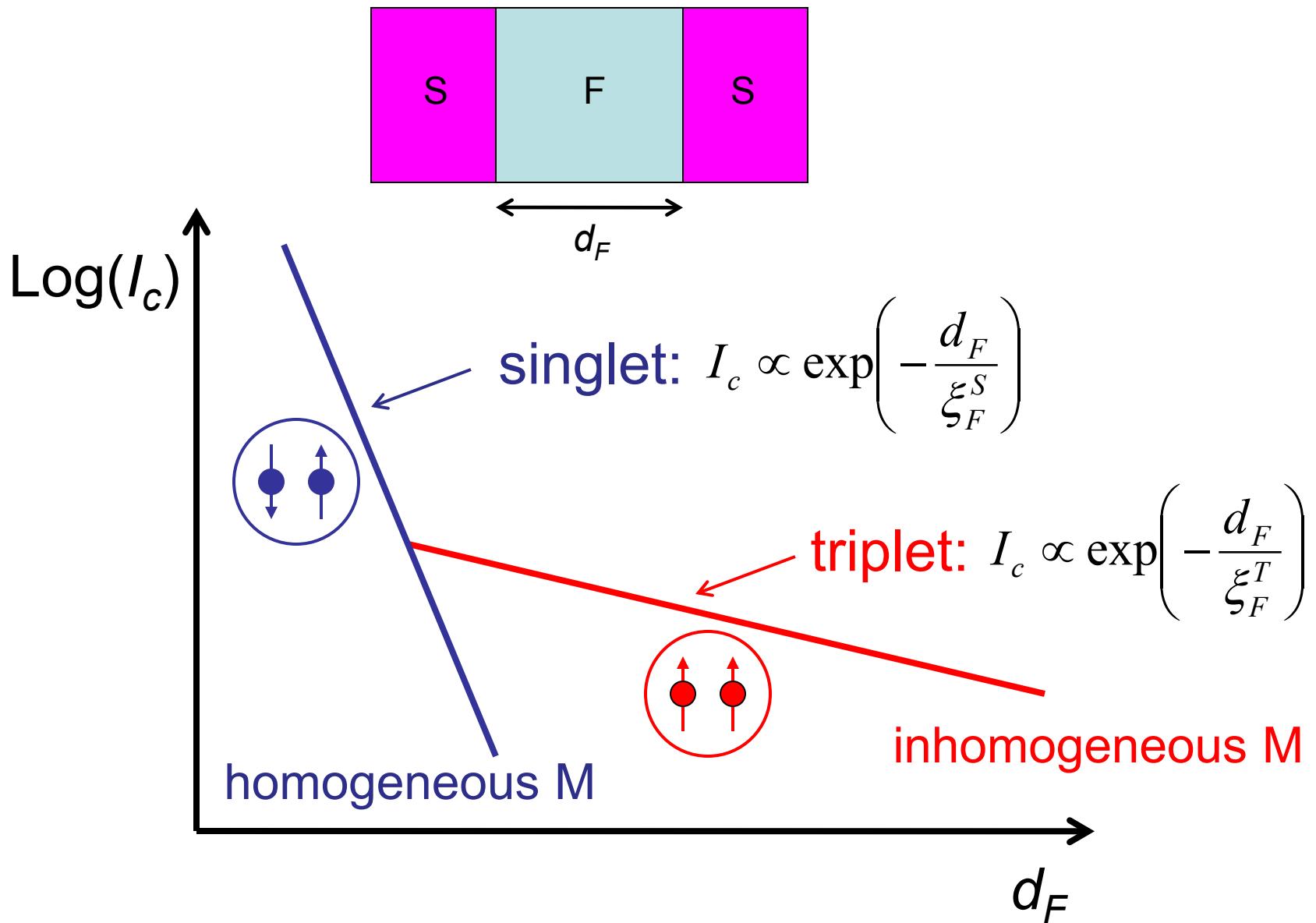
suppress singlet

triplet limited by spin-flip scattering

$$\xi_F^S = \sqrt{\frac{\hbar D_F}{E_{ex}}}$$

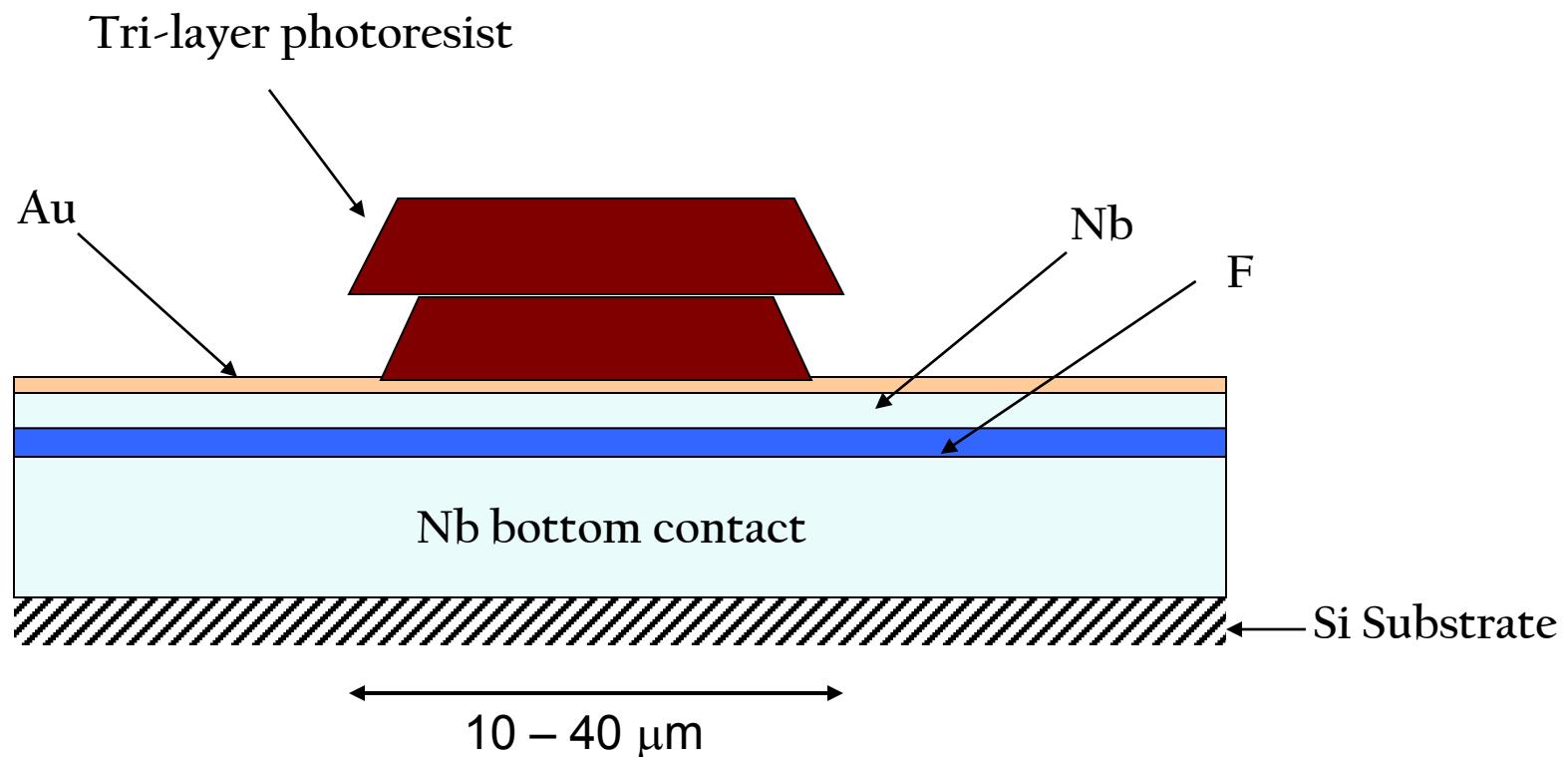
$$\xi_F^T = \sqrt{\frac{\hbar D_F}{2\pi k_B T}}$$

# Signature of spin-triplet:



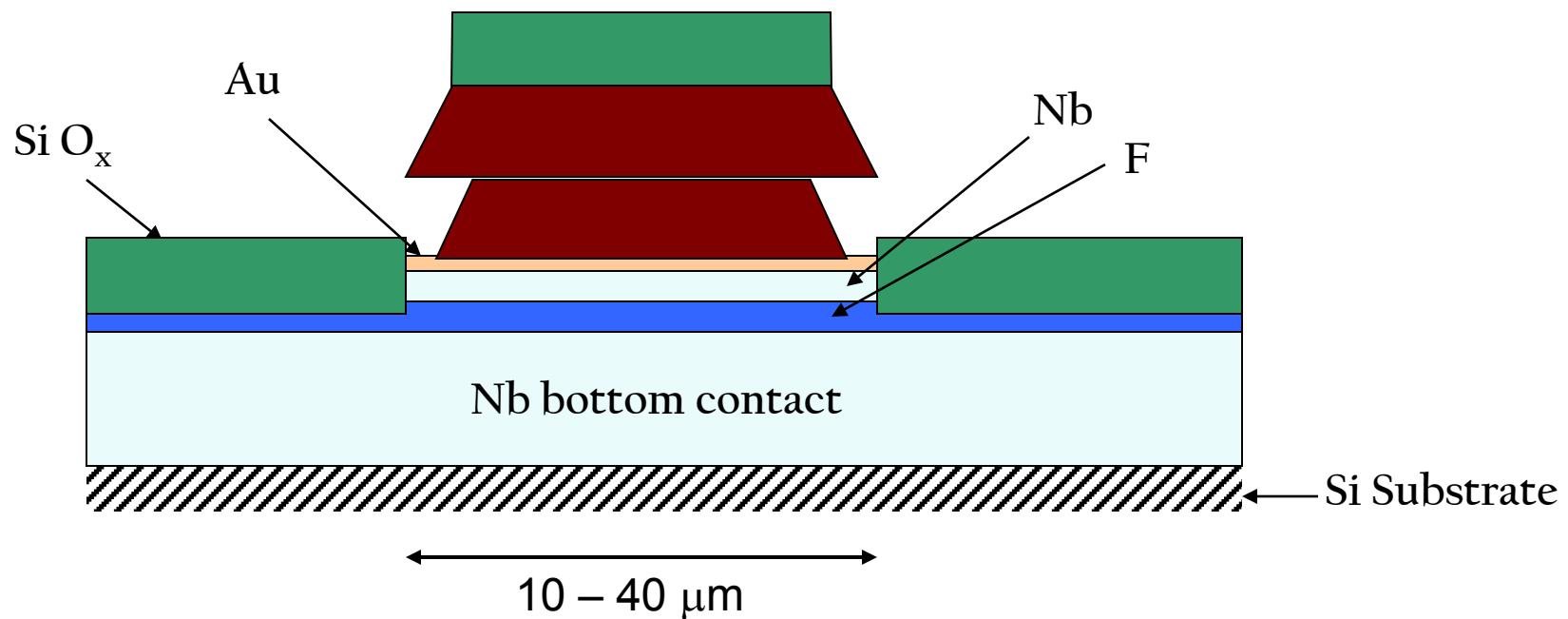
# Sample Fabrication

1. Sputter S/F/S
2. Pattern pillars with photolithography



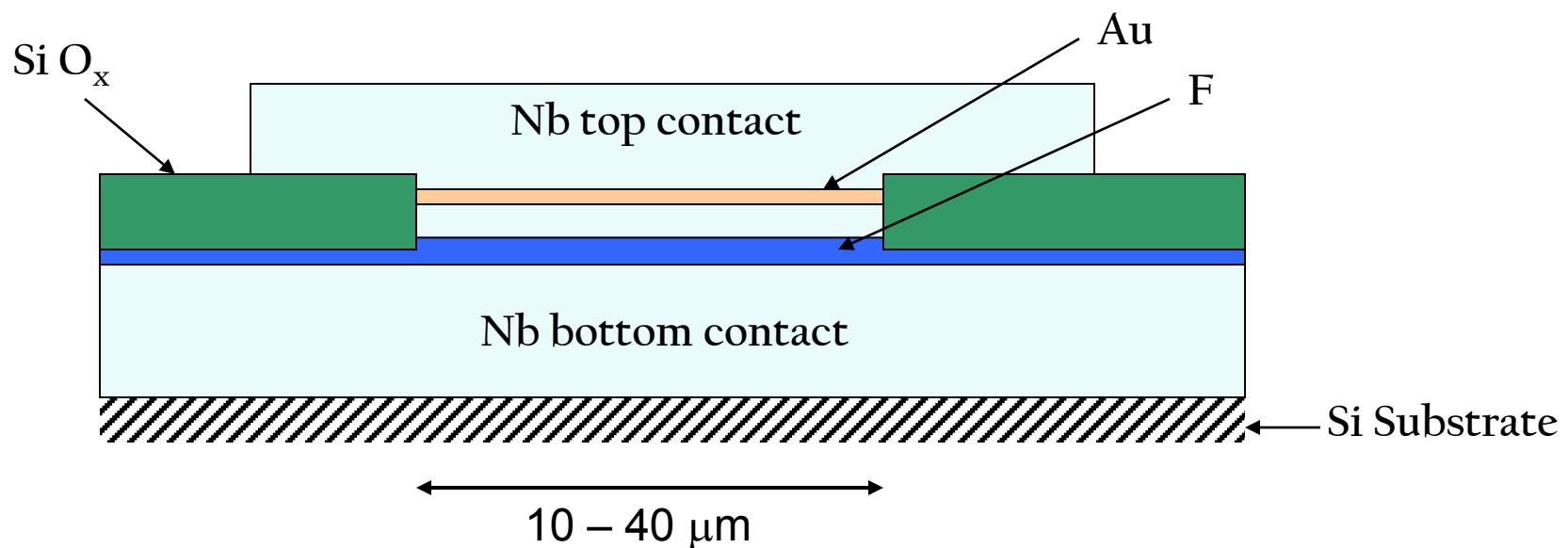
# Sample Fabrication

1. Sputter S/F/S
2. Pattern pillars with photolithography
3. Ion mill
4. Deposit SiO<sub>x</sub>



# Sample Fabrication

1. Sputter S/F/S
2. Pattern pillars with photolithography
3. Ion mill
4. Deposit SiO<sub>x</sub>
5. Lift off
6. Deposit top Nb contact



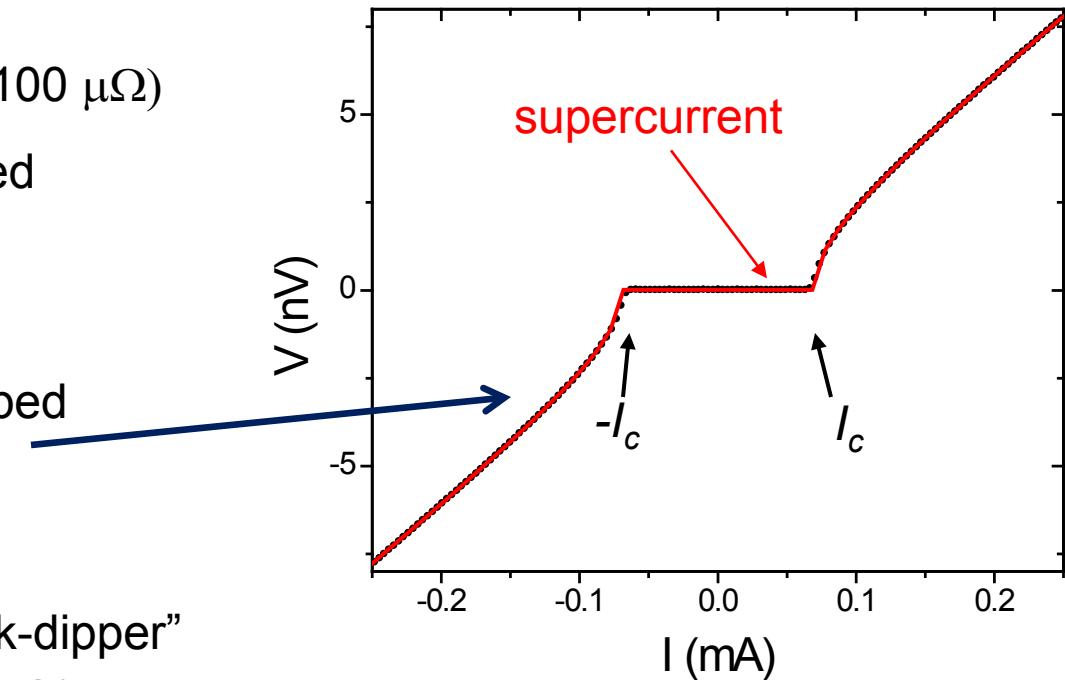
# Measurement

Low sample resistance ( $7 - 100 \mu\Omega$ )

→ Measure with SQUID-based  
current comparator circuit

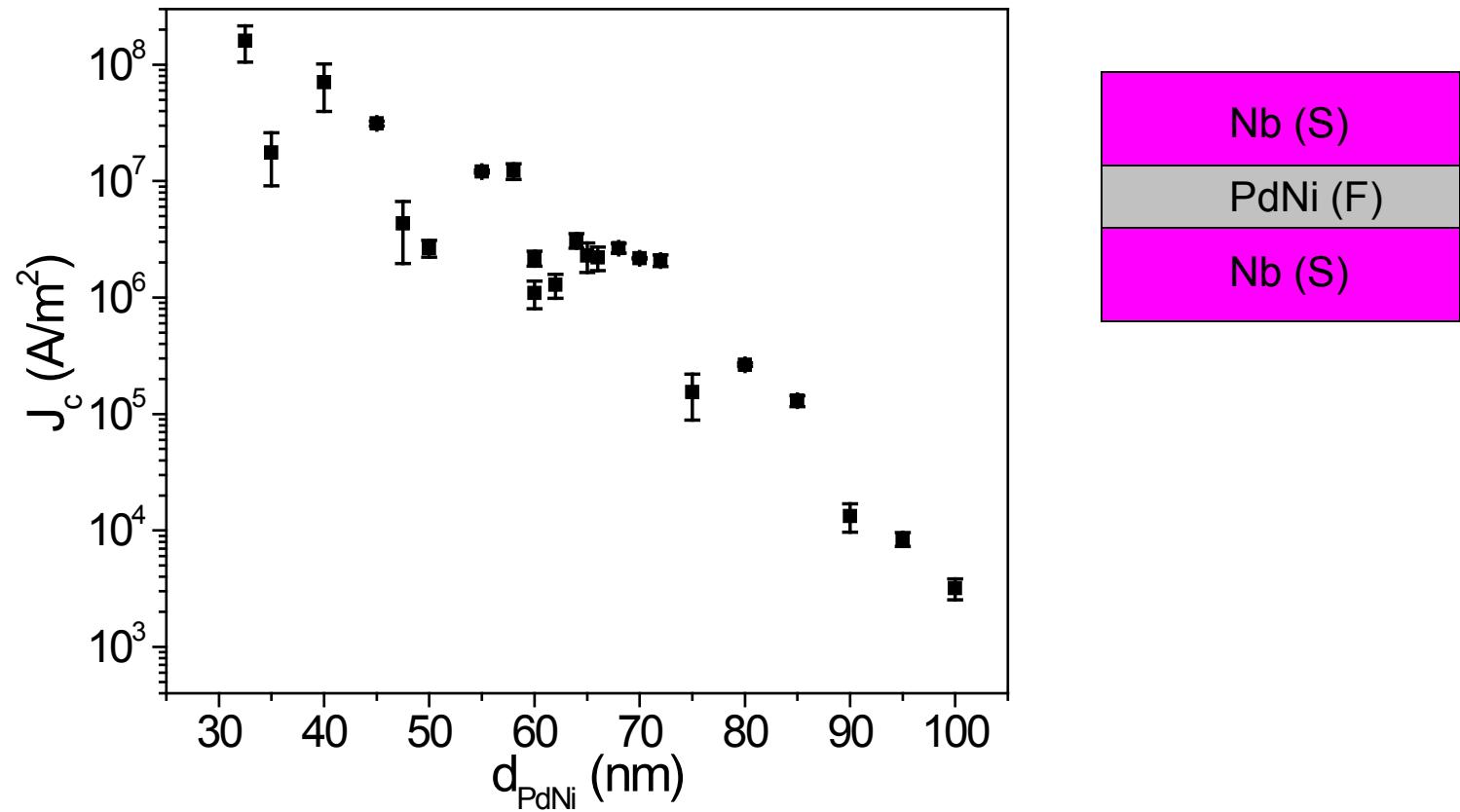
I-V characteristic of overdamped  
Josephson junctions

Measure at  $T = 4.2 \text{ K}$  in “quick-dipper”  
cryostat in helium storage dewar



$I_c \equiv$  critical current

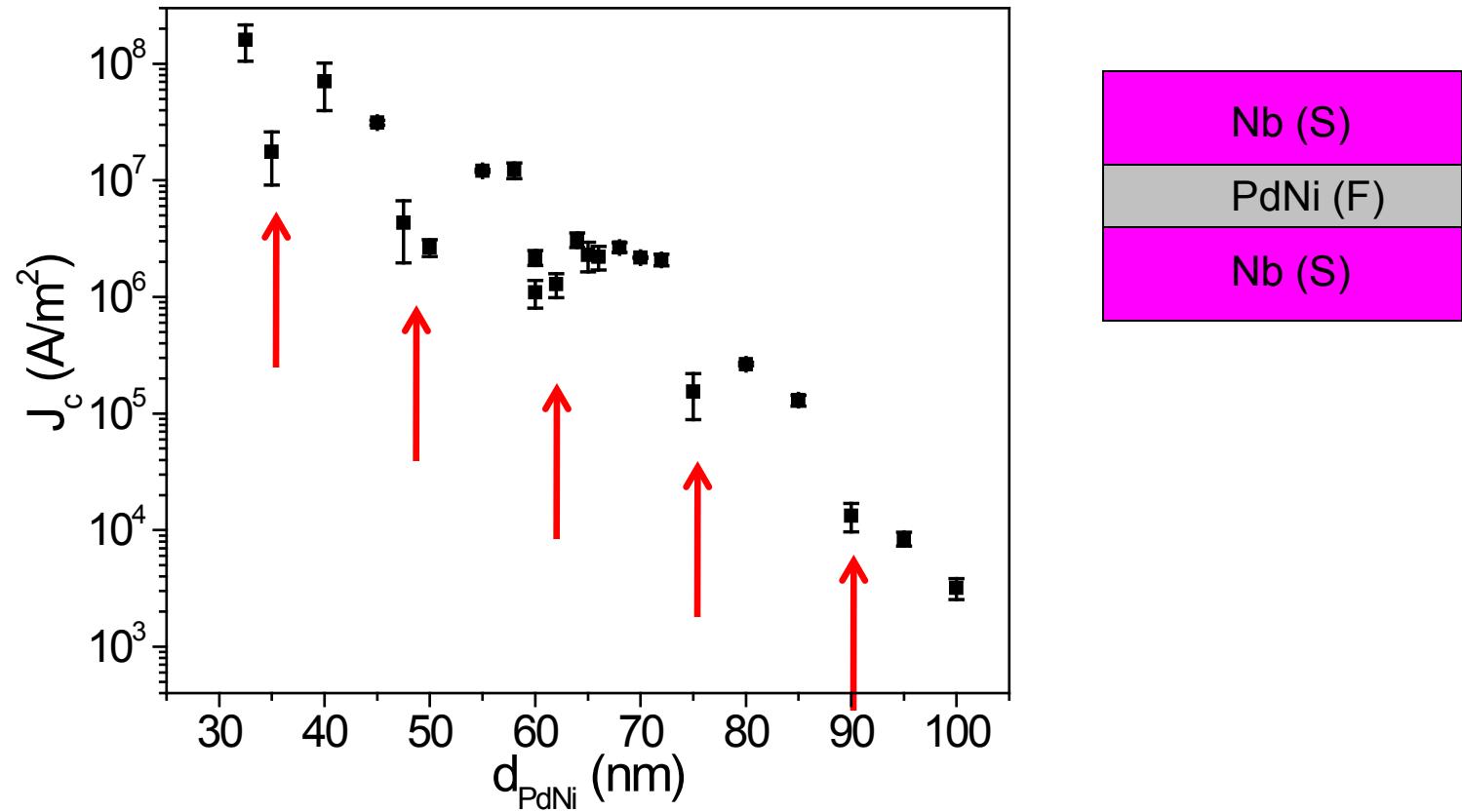
# S/F/S junctions with a weak F: Pd<sub>88</sub>Ni<sub>12</sub> alloy



(previous work on PdNi Josephson junctions had  $d_{\text{PdNi}} < 15$  nm)

Each point represents average over 2 - 4 junctions on same substrate

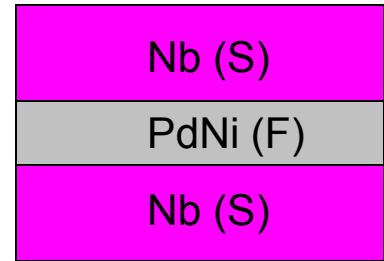
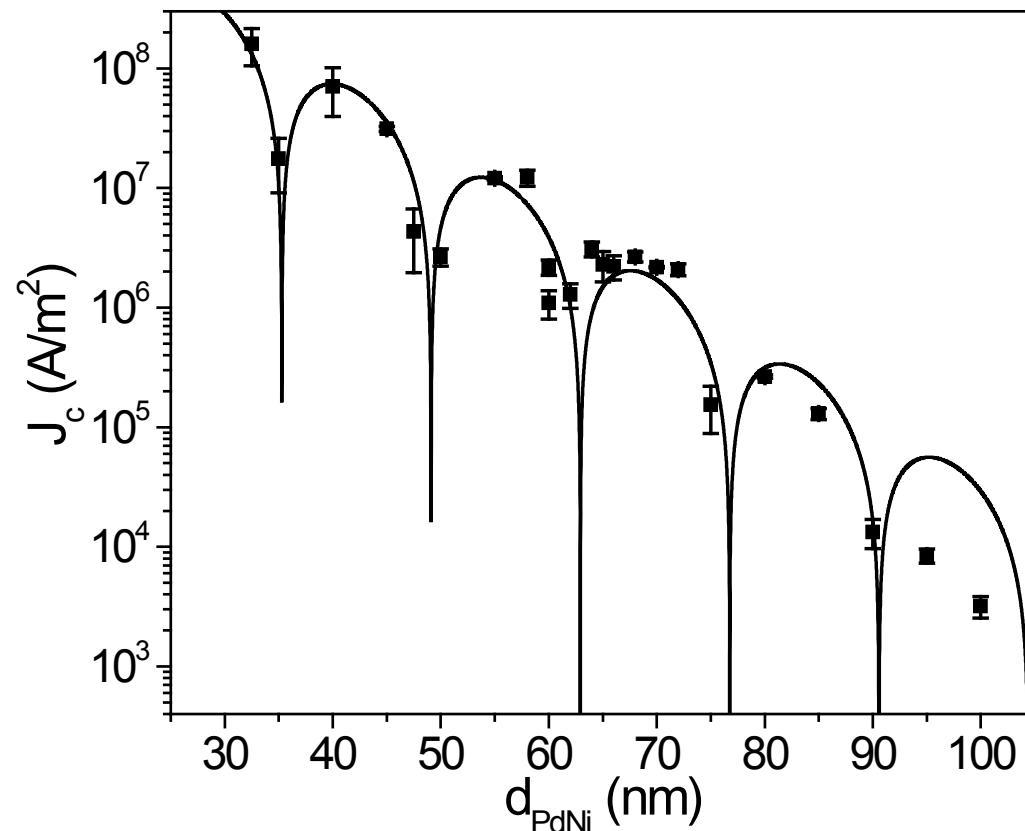
# S/F/S junctions with a weak F: Pd<sub>88</sub>Ni<sub>12</sub> alloy



**Periodic minima in  $J_c$**

Each point represents average over 2 - 4 junctions on same substrate

# S/F/S junctions with a weak F: Pd<sub>88</sub>Ni<sub>12</sub> alloy

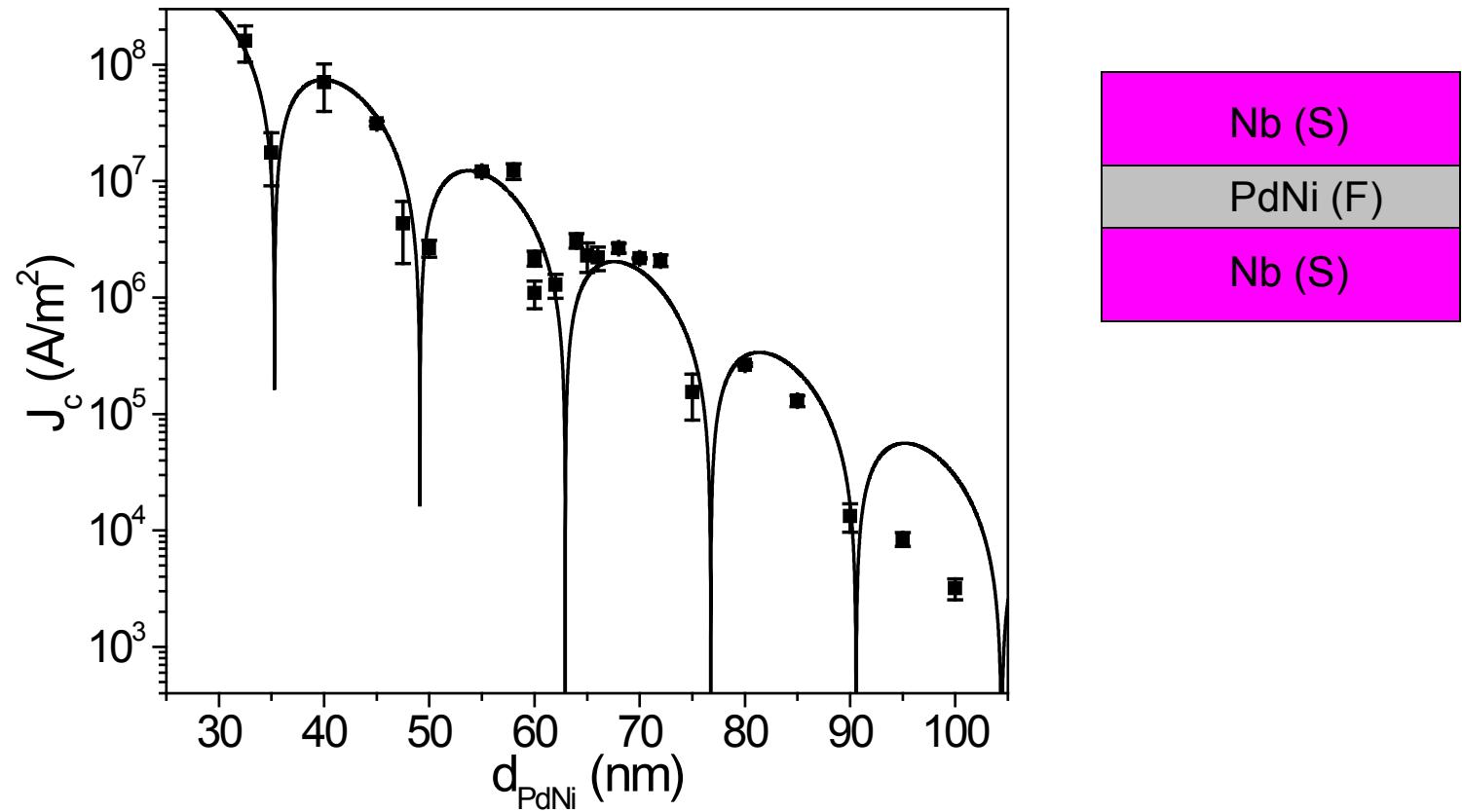


$$I_c \propto \exp(-x/\xi_{F1}) \cos(x/\xi_{F2})$$

$$\xi_1 = 7.7 \pm 0.5 \text{ nm} \quad \xi_2 = 4.4 \pm 0.1 \text{ nm}$$

T.S. Khaire, W.P. Pratt, and N.O. Birge, Phys. Rev. B 79, 094523 (2009)

# S/F/S junctions with a weak F: Pd<sub>88</sub>Ni<sub>12</sub> alloy



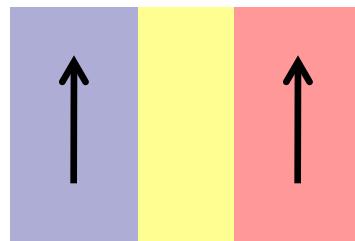
No sign of spin-triplet supercurrent!

# Why don't we see spin-triplet supercurrent in S/F/S Josephson junctions with PdNi?

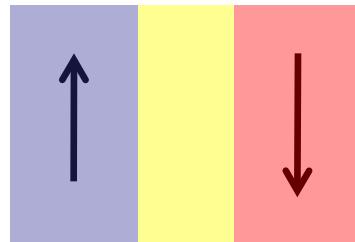
- Not enough non-collinear magnetization
- Magnetic inhomogeneity on wrong length scale
- Too much spin-flip and/or spin-orbit scattering

# How to measure the spin memory length using Giant Magnetoresistance (GMR)

P state



AP state



$$R_P < R_{AP}$$

Albert Fert &  
Peter Grunberg  
2007 Nobel Prize

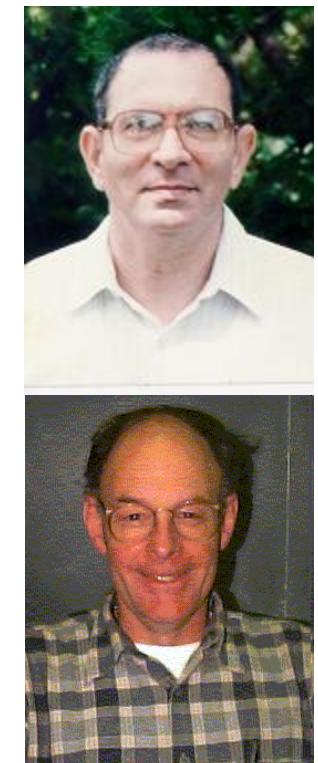


$Pd_{0.88}Ni_{0.12}$

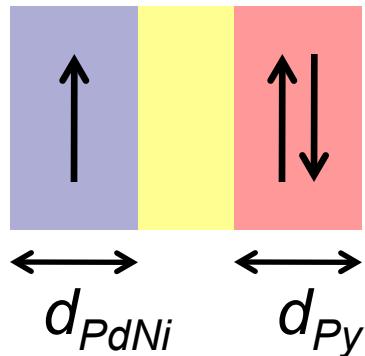
Cu

Permalloy =  
 $Ni_{0.84}Fe_{0.16}$

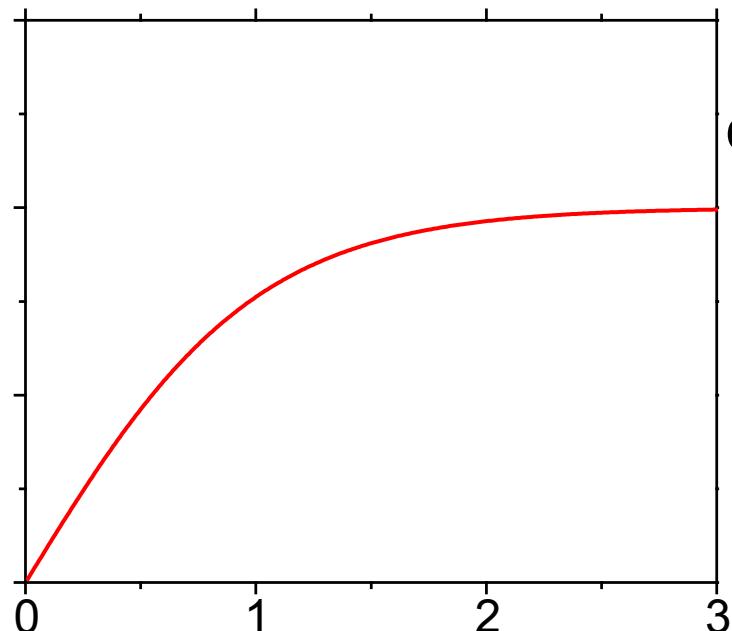
Jack Bass  
& Bill Pratt



# Dependence of GMR signal on $d_{PdNi}$



Fix  $d_{Py}$ , vary  $d_{PdNi}$

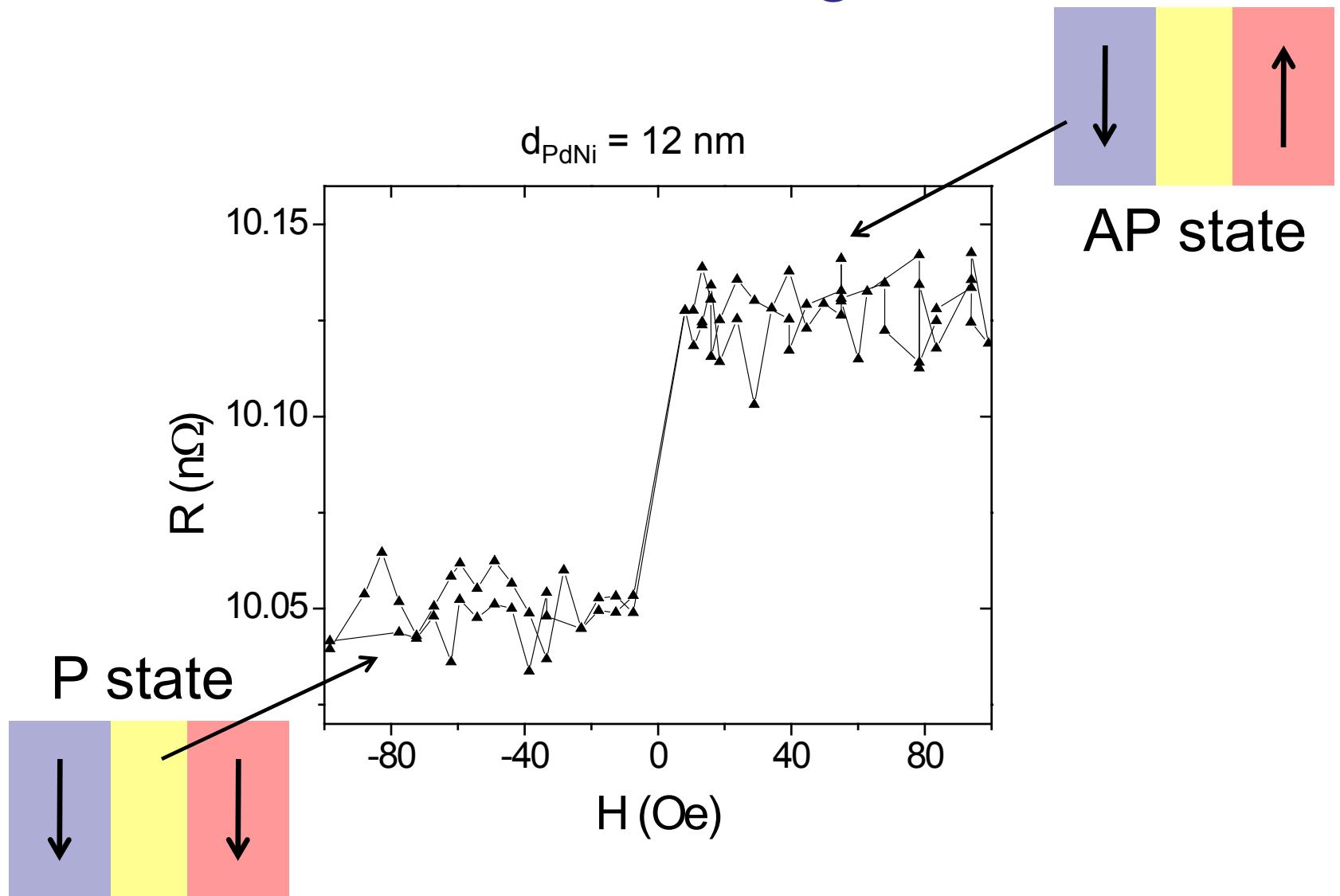


If  $d_{PdNi} < l_{PdNi}^{sf}$      $A\Delta R \propto \beta_{PdNi} \rho_{PdNi}^* d_{PdNi}$

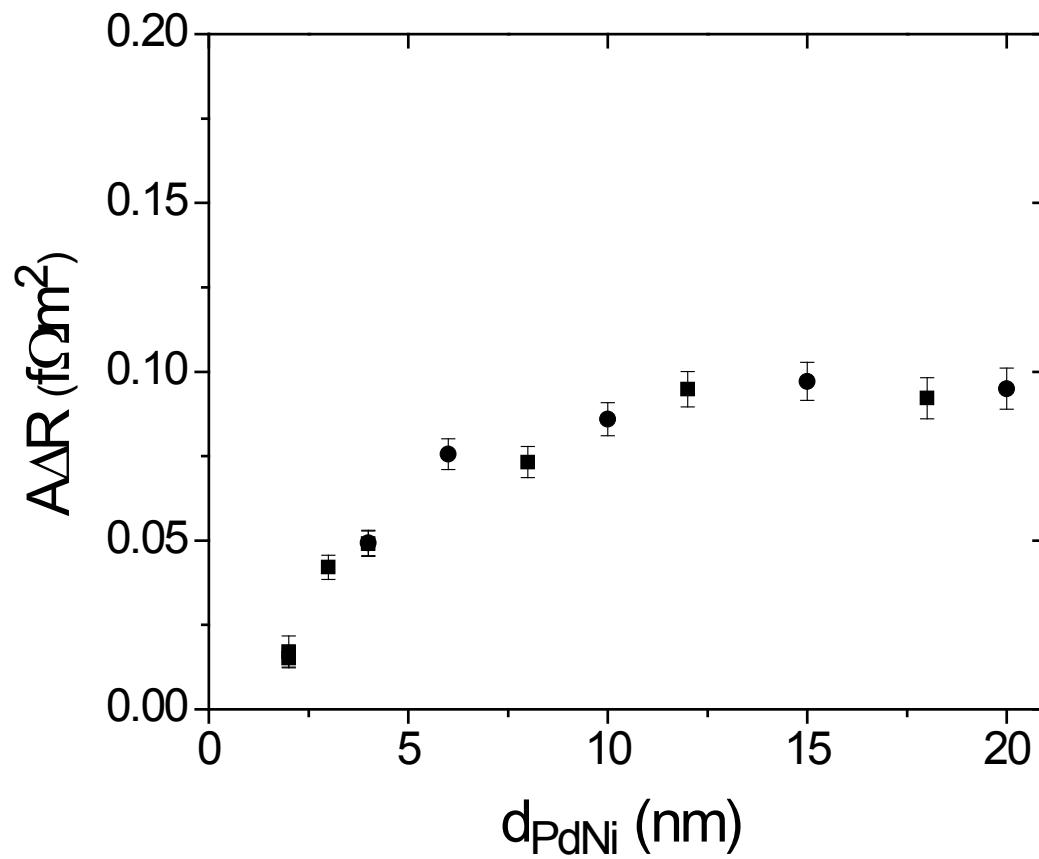
If  $d_{PdNi} > l_{PdNi}^{sf}$      $A\Delta R \propto \beta_{PdNi} \rho_{PdNi}^* l_{PdNi}^{sf}$

$$\beta \equiv \frac{\rho^{\uparrow} - \rho^{\downarrow}}{\rho^{\uparrow} + \rho^{\downarrow}}$$

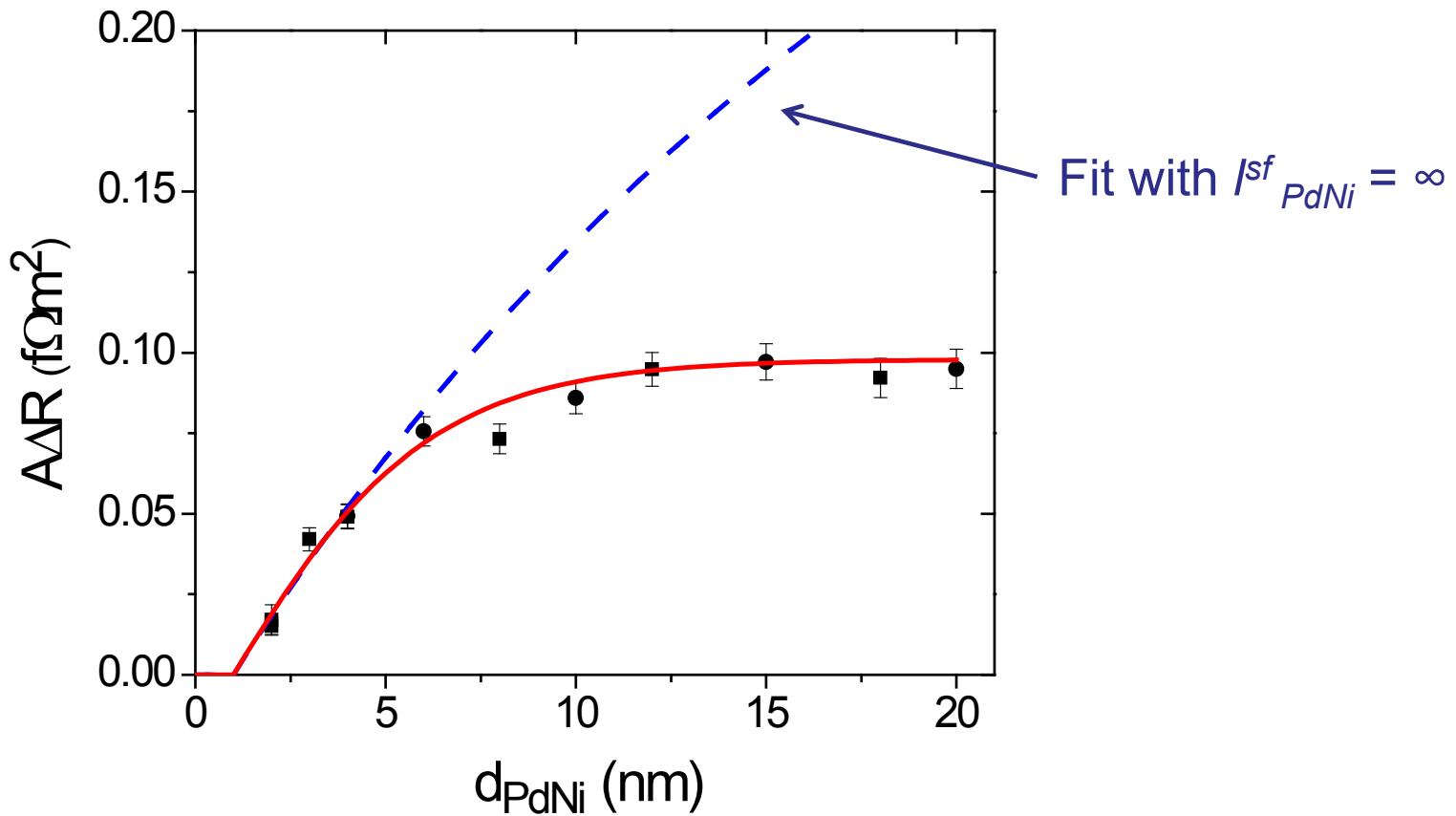
# Raw GMR signal



# GMR signal vs. $d_{\text{PdNi}}$



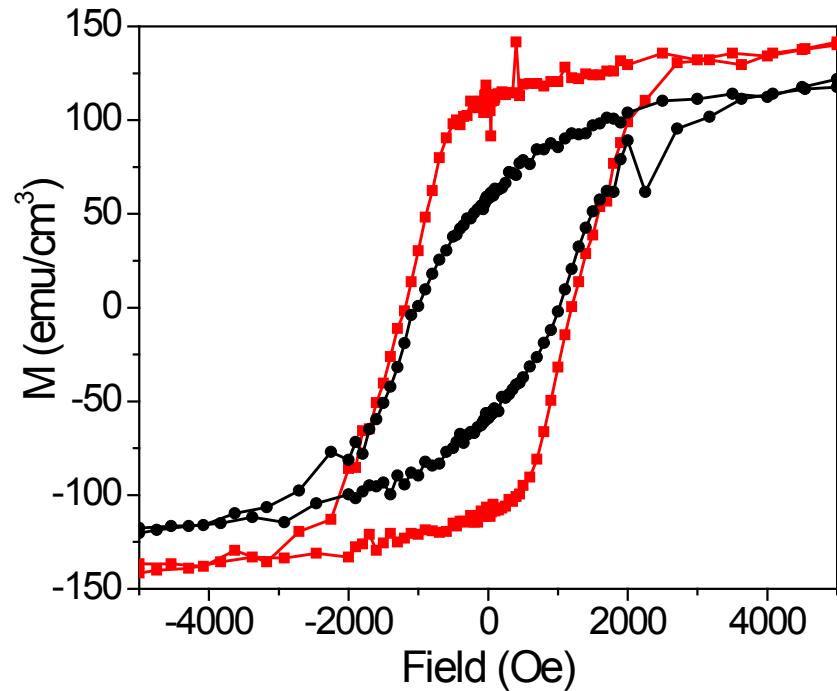
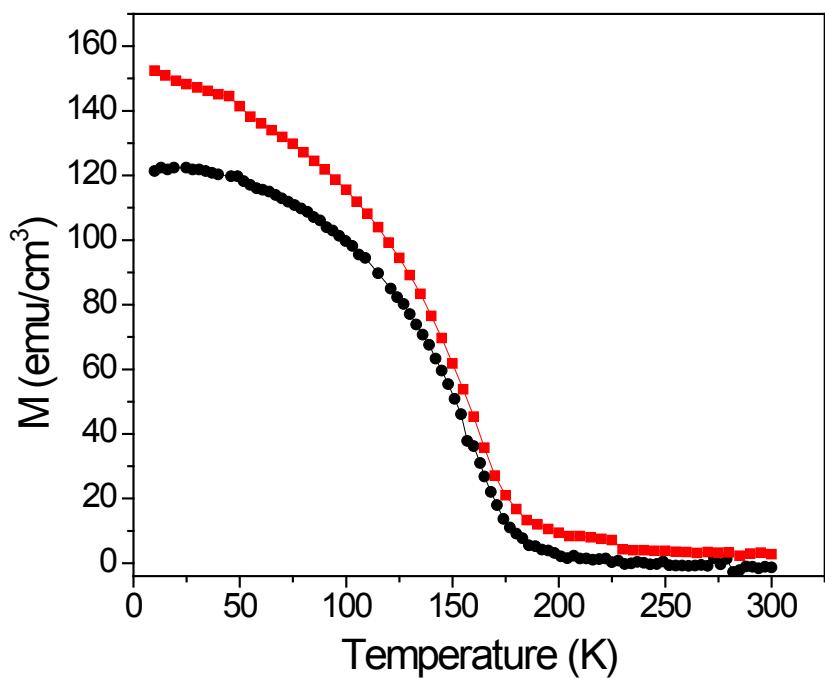
# Fit to Valet-Fert theory



Result:  $I_{sf}^{PdNi} = 2.8 \pm 0.5$  nm

# $Pd_{1-x}Ni_x$ magnetic characterization

- H out-of-plane
- H in-plane



PdNi alloy has out-of-plane magnetic anisotropy!!

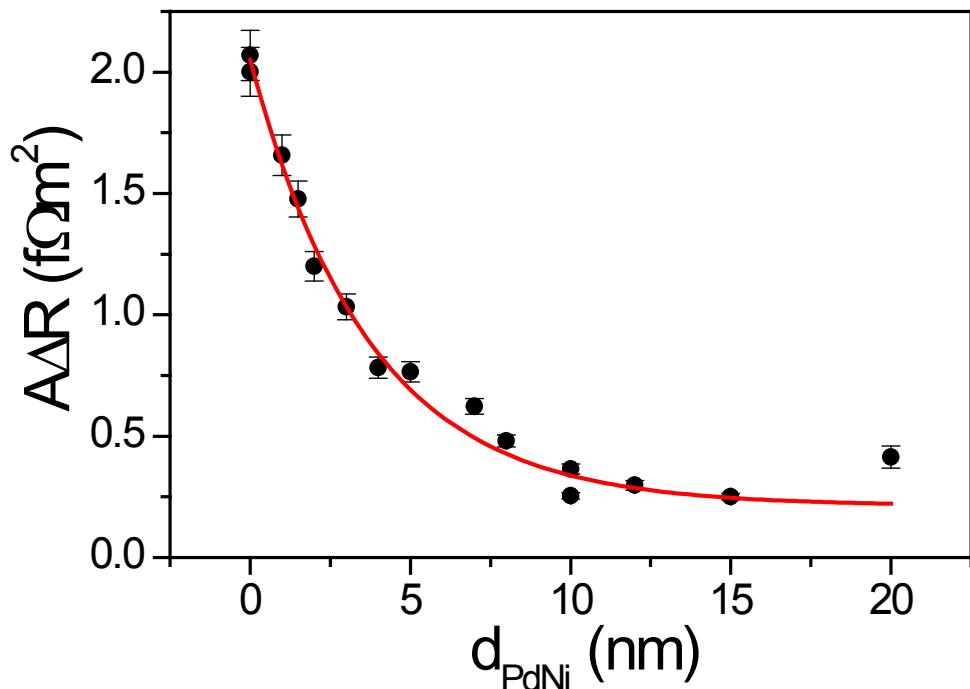
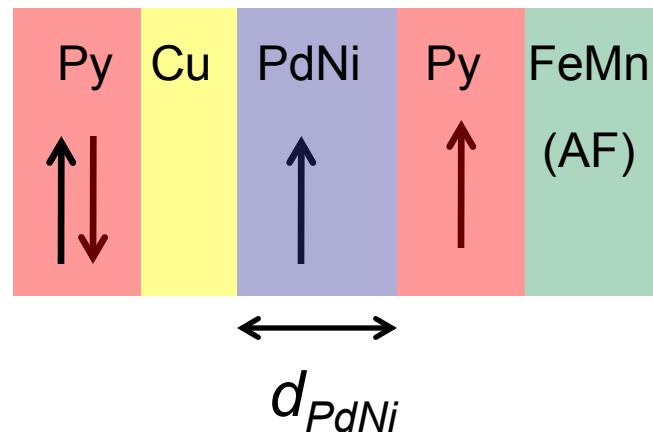
(violates assumptions about P and AP states in GMR experiment)

# Alternate “spoiler” spin valve geometry

PdNi coupled to Py; Py coupled to FeMn

$$\beta_{PdNi} \ll \beta_{Py}$$

$$\beta \equiv \frac{\rho^{\uparrow} - \rho^{\downarrow}}{\rho^{\uparrow} + \rho^{\downarrow}}$$



If  $d_{PdNi} \ll l_{PdNi}^{sf}$

Sample acts like Py/Cu/Py spin valve

If  $d_{PdNi} > l_{PdNi}^{sf}$

Sample acts like Py/Cu/PdNi spin valve

Fit:  $l_{PdNi}^{sf} = 5.4 \pm 0.6 \text{ nm}$

*Still very short!!*

# Why don't we see spin-triplet supercurrent in S/F/S Josephson junctions with PdNi?

- Not enough non-collinear magnetization
- Magnetic inhomogeneity on wrong length scale
- Too much spin-flip and/or spin-orbit scattering

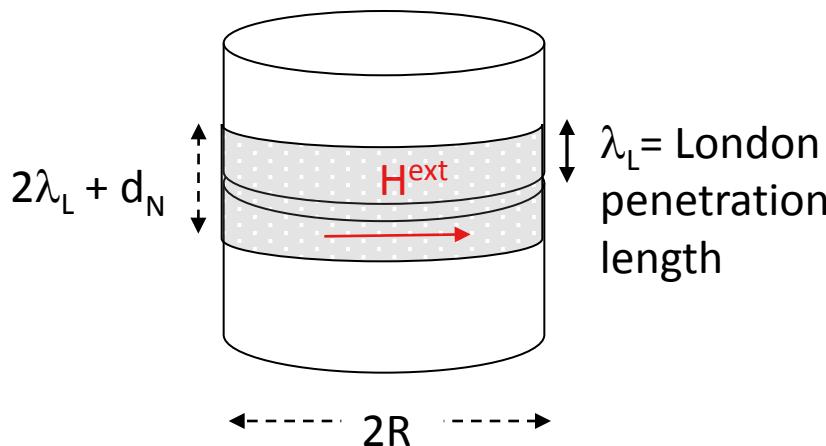


Try a different approach:

Use a strong F with long spin-memory length: Co

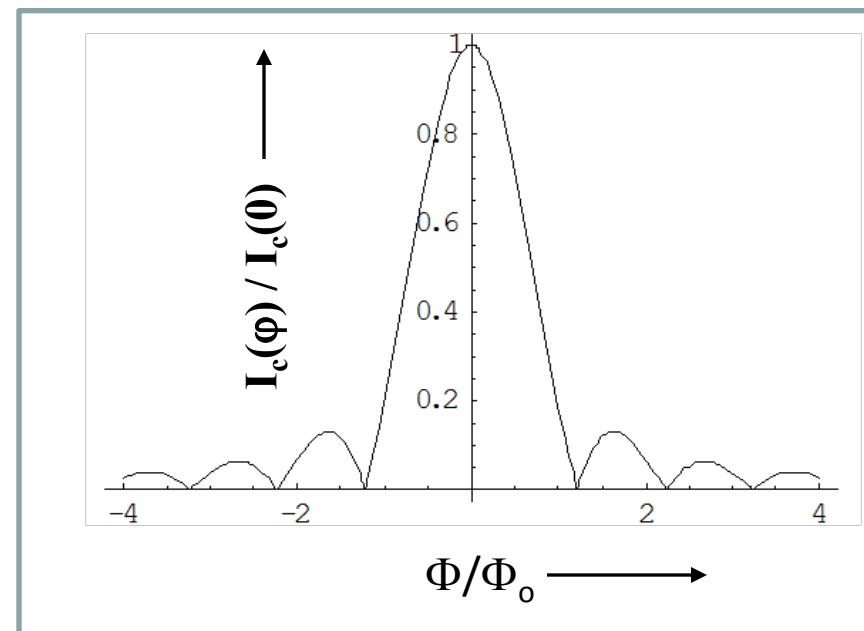
# Aside: Characterization of Josephson Junctions by the Fraunhofer pattern

“I never believe anybody’s Josephson junction data unless they show me a Fraunhofer pattern.” -- Dale van Harlingen



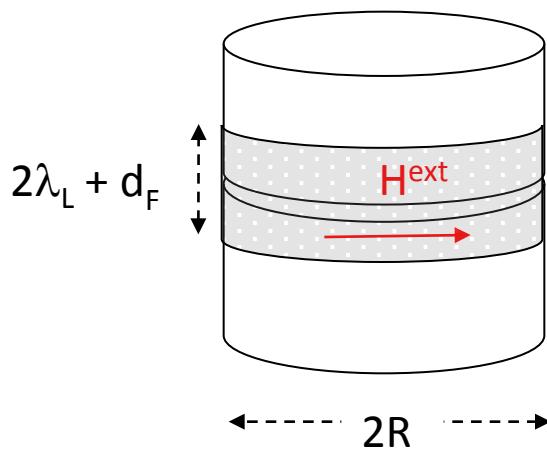
$$I_C(\Phi) = I_C(0) \frac{2 \times J_1\left(\frac{\pi\Phi}{\Phi_o}\right)}{\frac{\pi\Phi}{\Phi_o}}$$

where  $\Phi = H^{\text{ext}}(2\lambda_L + d_N)2R$

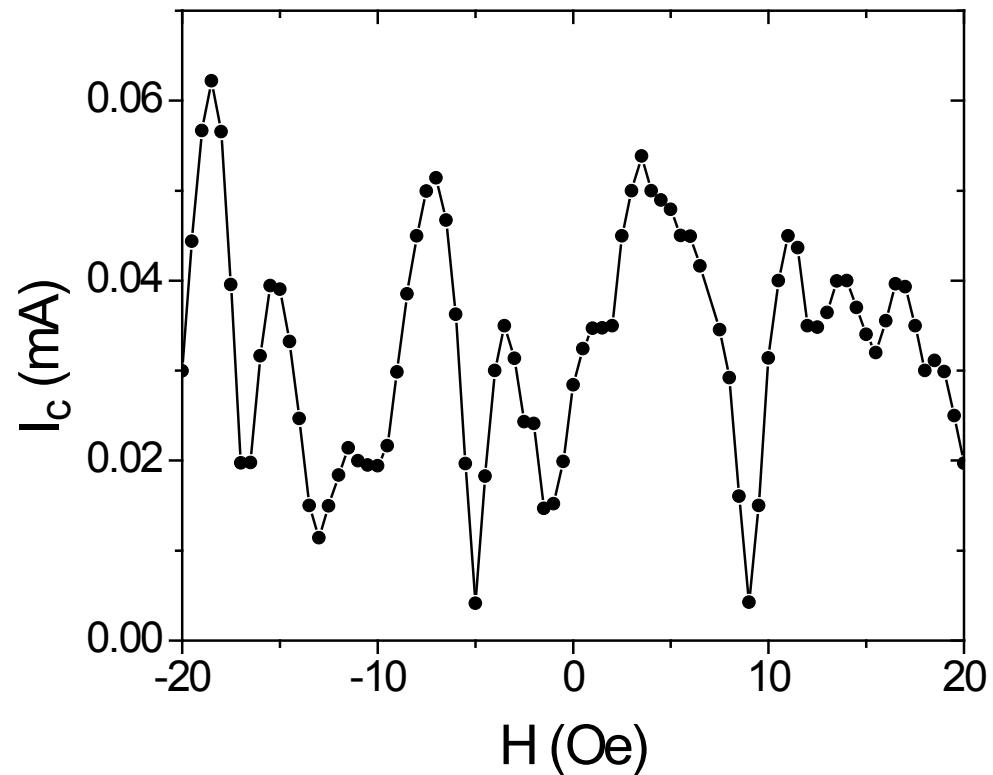


Airy diffraction pattern for a circular junction

# Large-area Nb/Co/Nb junctions

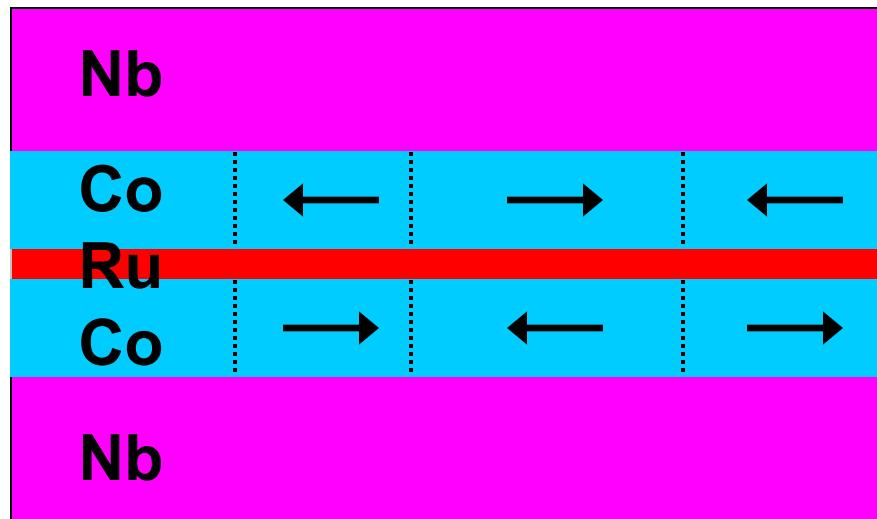


Nb/Co/Nb,  $d_{\text{Co}} = 5 \text{ nm}$ ,  $2R = 40 \mu\text{m}$



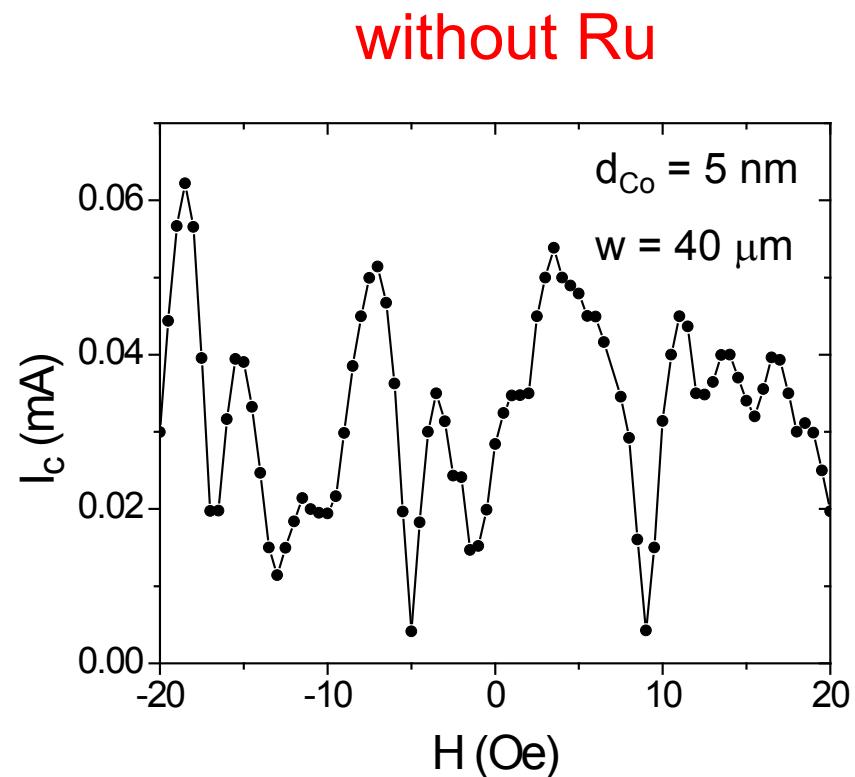
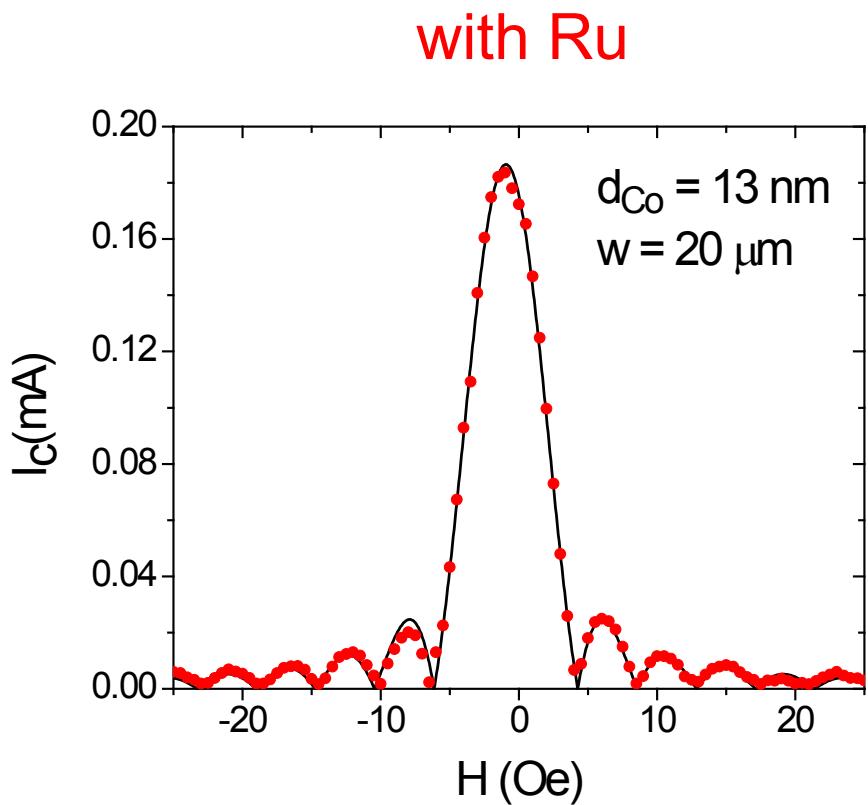
Random Fraunhofer pattern due  
to complex domain configuration

# Trick: achieve flux cancellation with Co/Ru/Co synthetic antiferromagnet

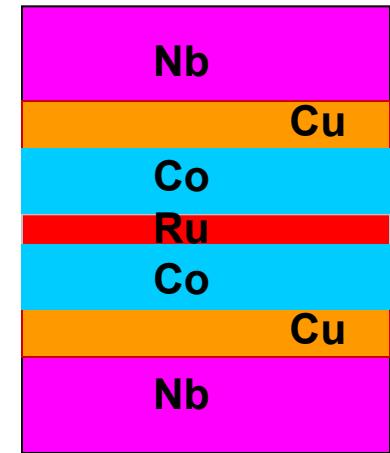
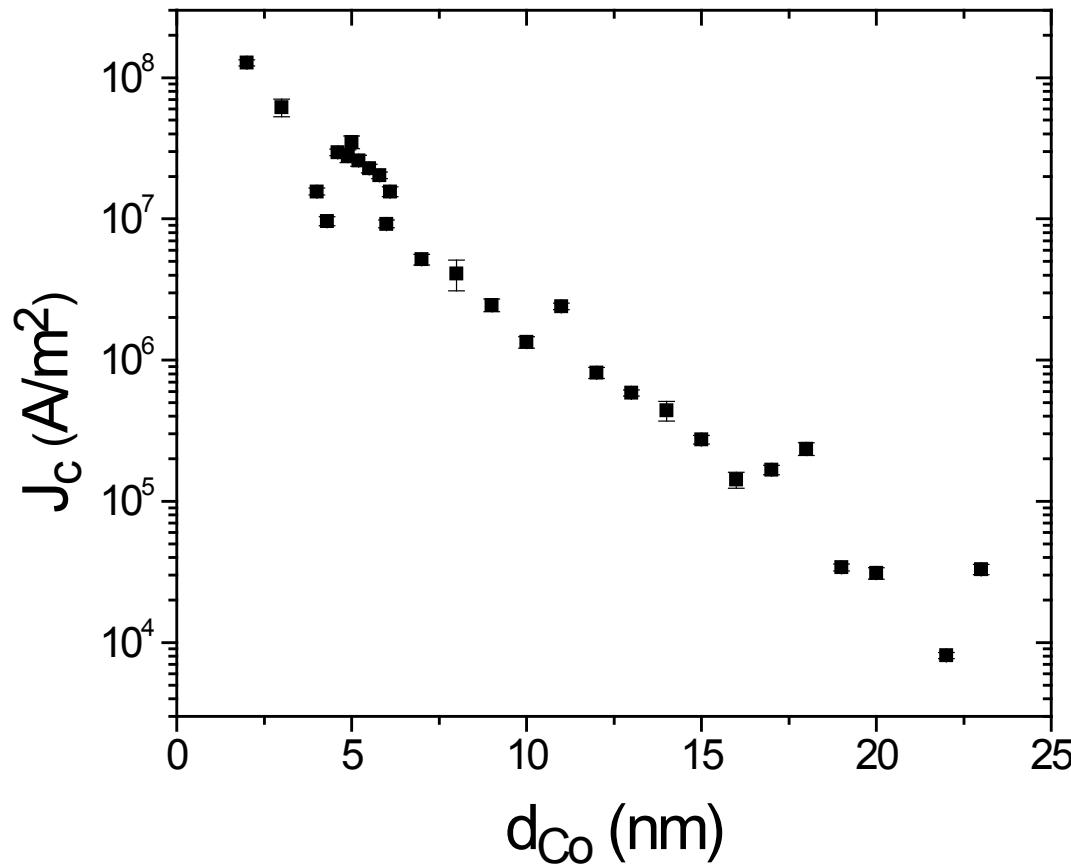


H.A.M. vandenBerg *et al.*, J. Mag. Magn. Mat. 165, 524 (1997).

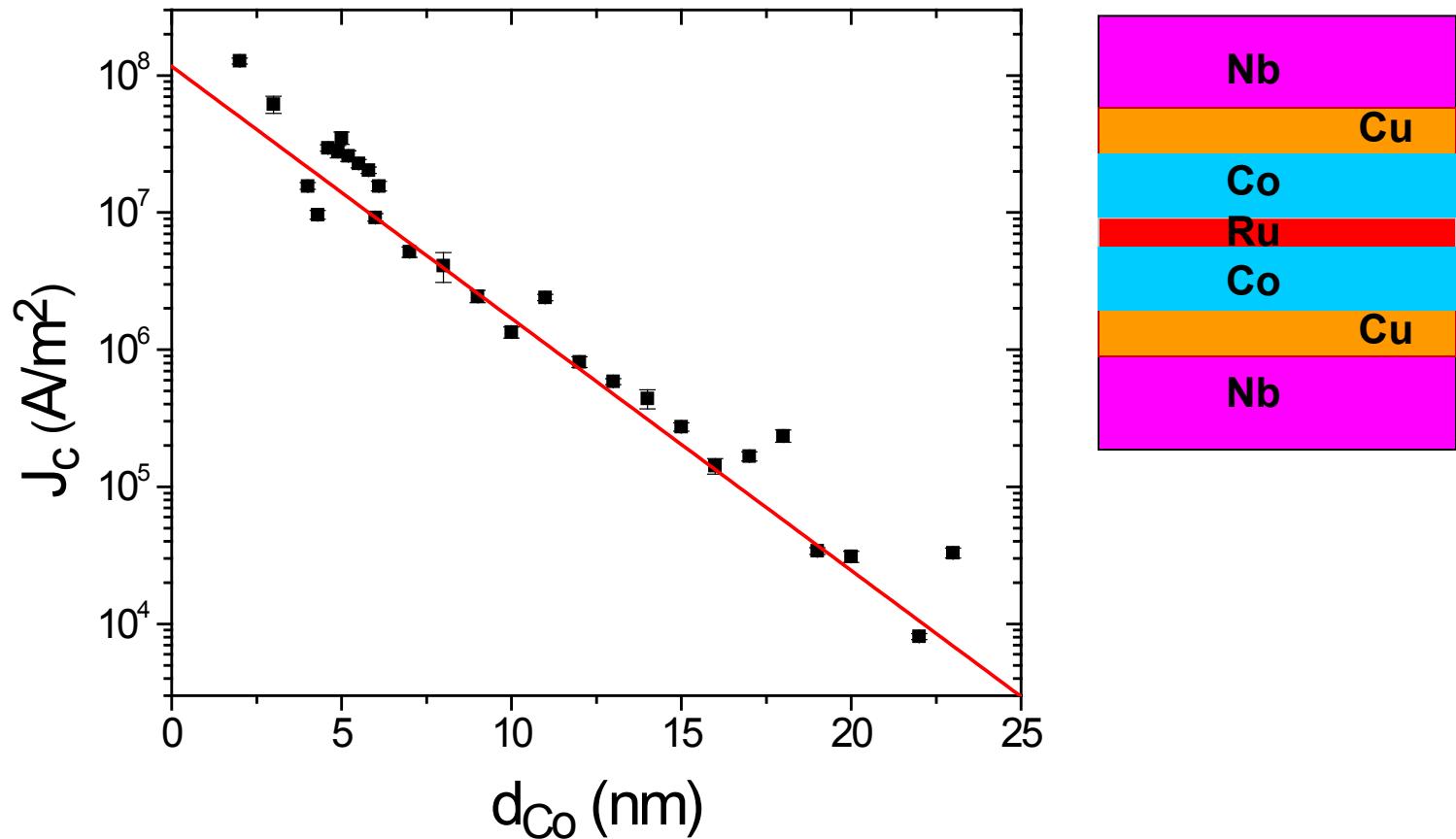
# Co/Ru/Co synthetic antiferromagnet restores Fraunhofer pattern!



# S/F/S junction with a strong F: Co



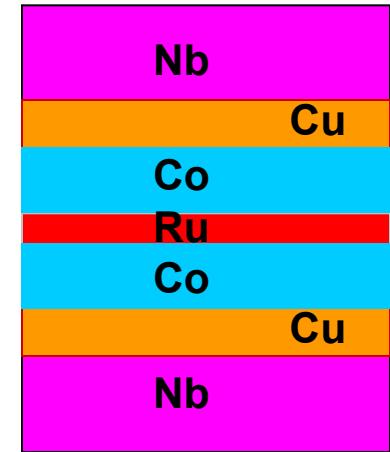
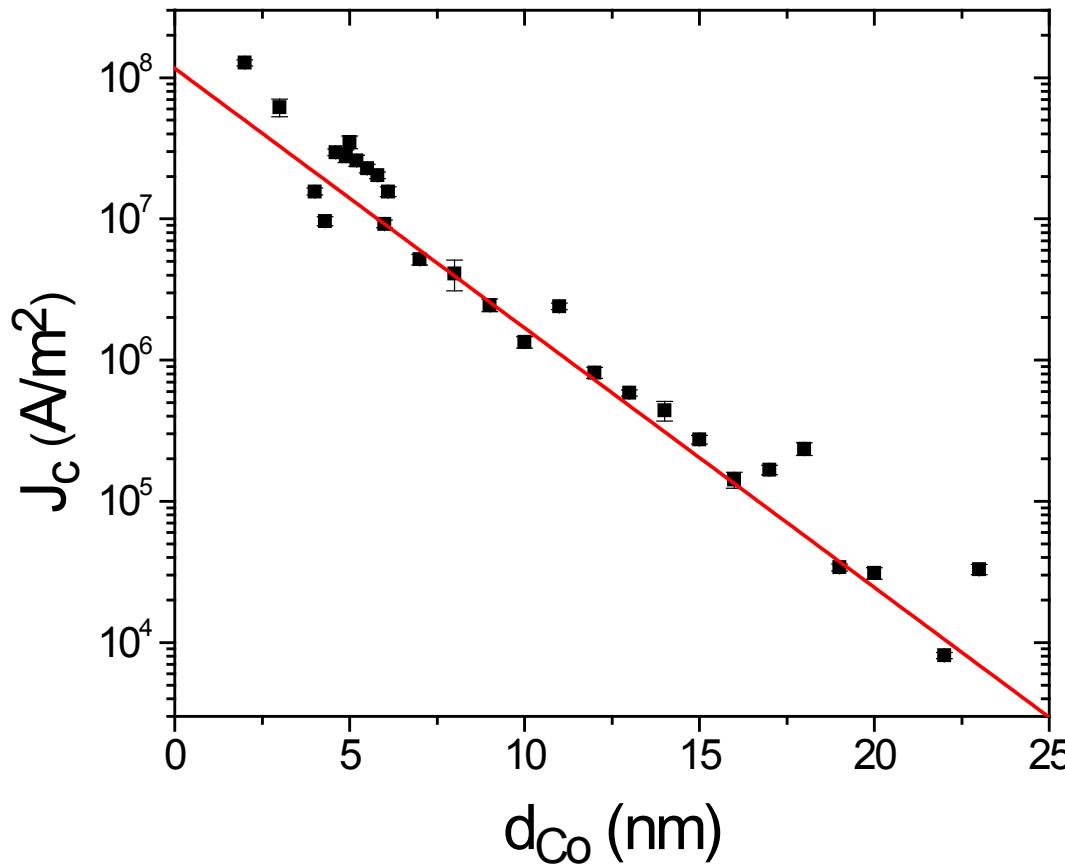
# S/F/S junction with a strong F: Co



No oscillations of  $I_c$ : phase shifts cancel in two F layers

Blanter & Hekking, PRB **69**, 024525 (2004); Crouzy et al., PRB **75**, 054503 (2007).

# S/F/S junction with a strong F: Co



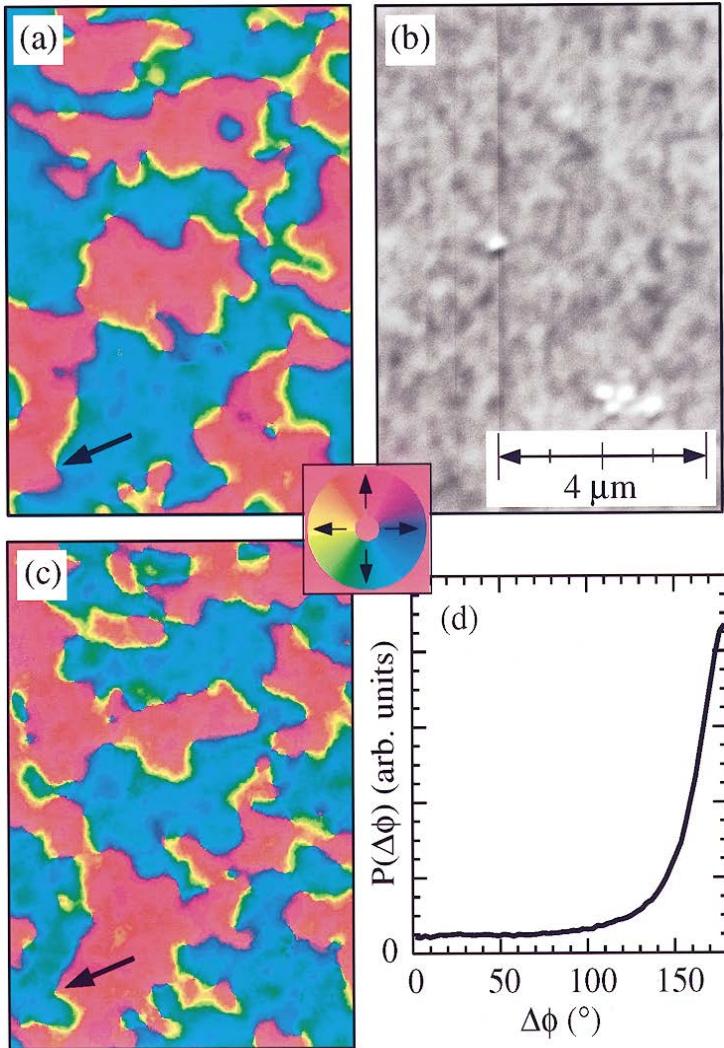
No sign of spin-triplet supercurrent!

# Why don't we see spin-triplet supercurrent in S/F/S Josephson junctions with Co/Ru/Co?

- Not enough non-collinear magnetization
- Magnetic inhomogeneity on wrong length scale
- ~~Too much spin-flip scattering at Co/Ru interface~~

Measure spin-flip scattering at Co/Ru interface using GMR

# Co domain structure



Neighboring domains have mostly anti-parallel magnetization

Not much non-collinear M

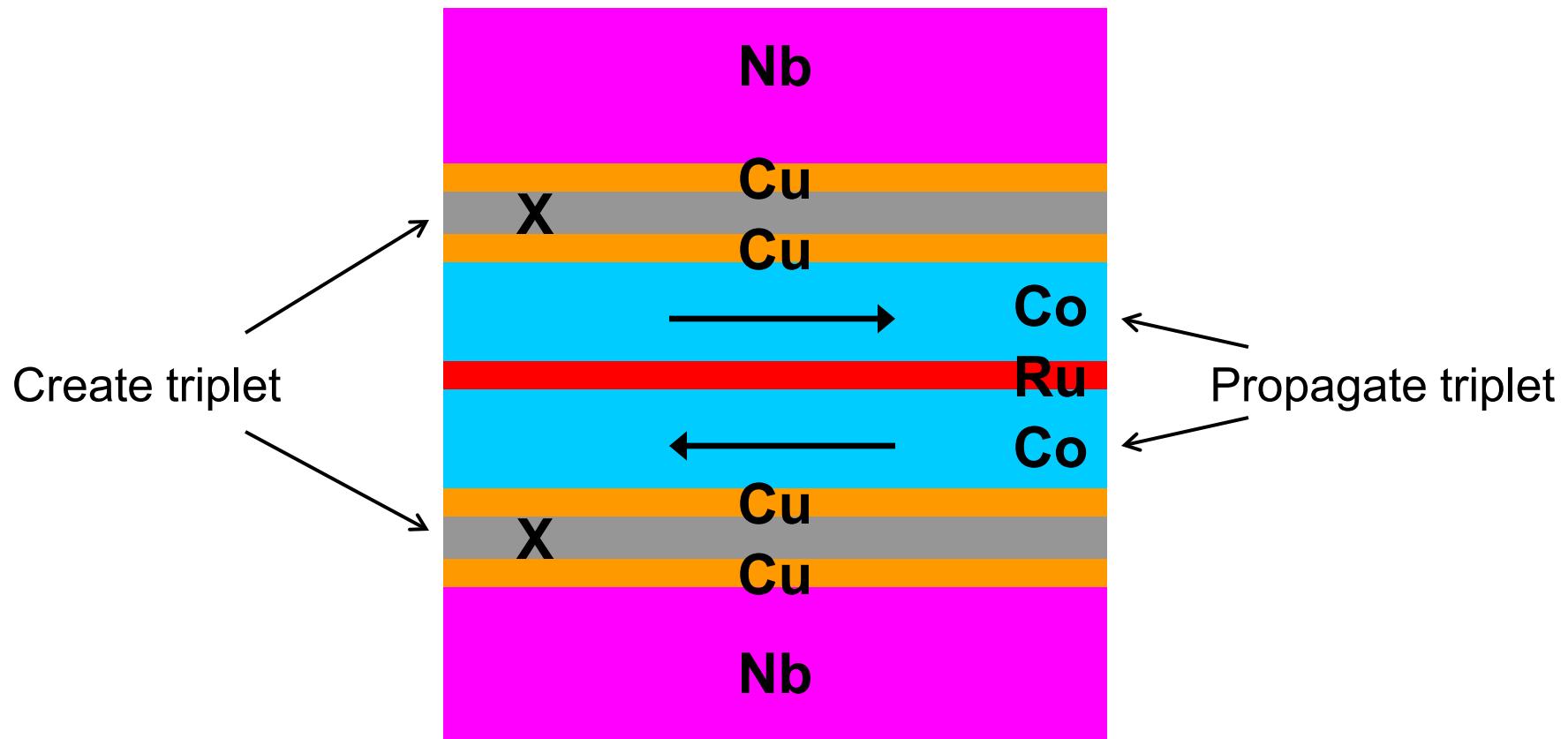
Domains are large ~ 3 μm

Borchers et al., PRL **82**, 2796 (1999).  
SEMPA = scanning electron microscopy  
with polarization analysis

# Why haven't we seen spin-triplet correlations?

- PdNi
  - Spin memory length too short
  - Bad for propagation of triplet
  - Good for generation of triplet
- Co/Ru/Co
  - Not enough magnetic inhomogeneity
  - Bad for generation of triplet
  - Good for propagation of triplet

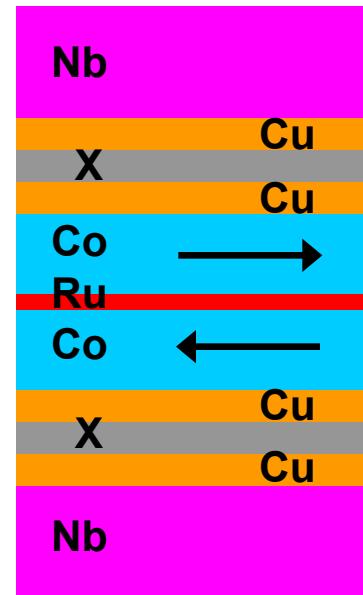
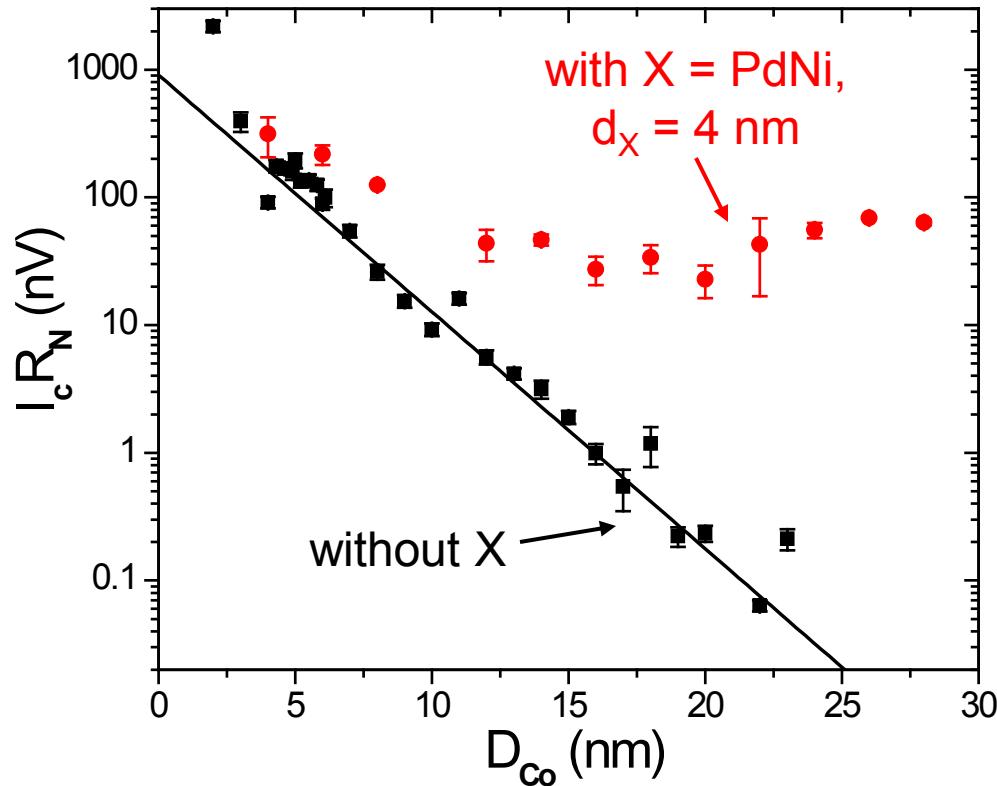
# New Idea: combine best of two materials



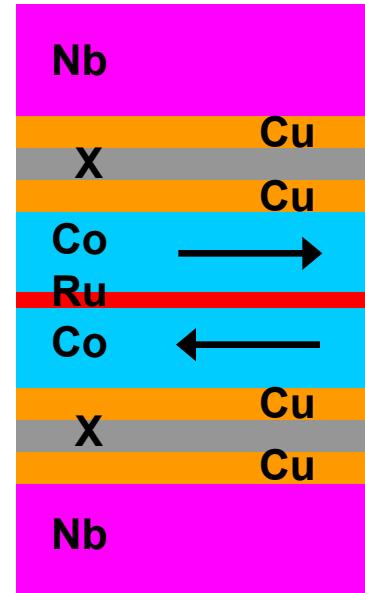
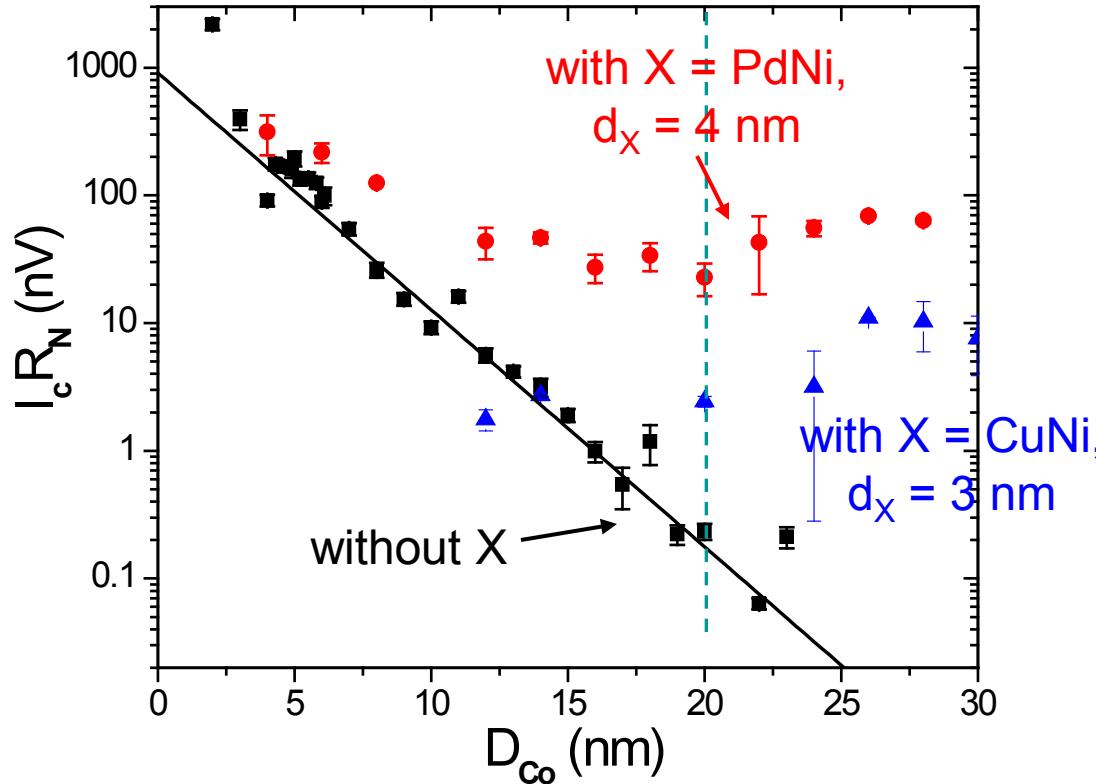
**X = PdNi or CuNi alloy**

(Cu buffer layers magnetically isolate X from Co)

# Finally, the triplet appears!

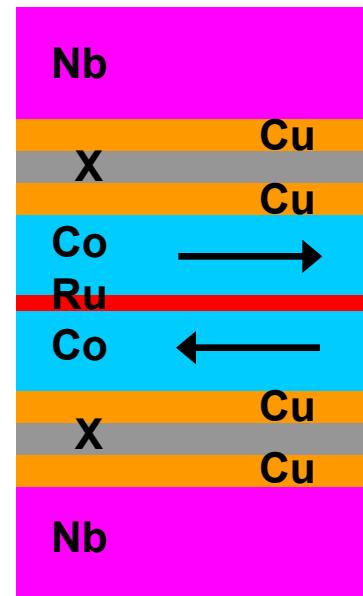
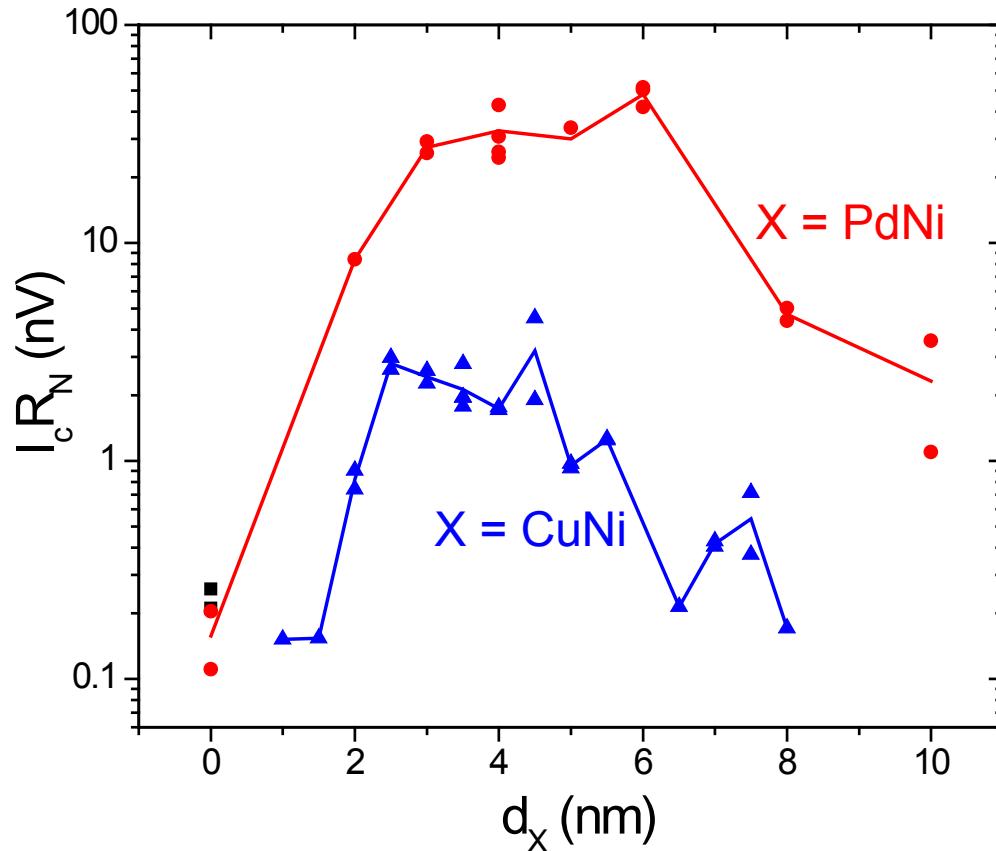


# Finally, the triplet appears!

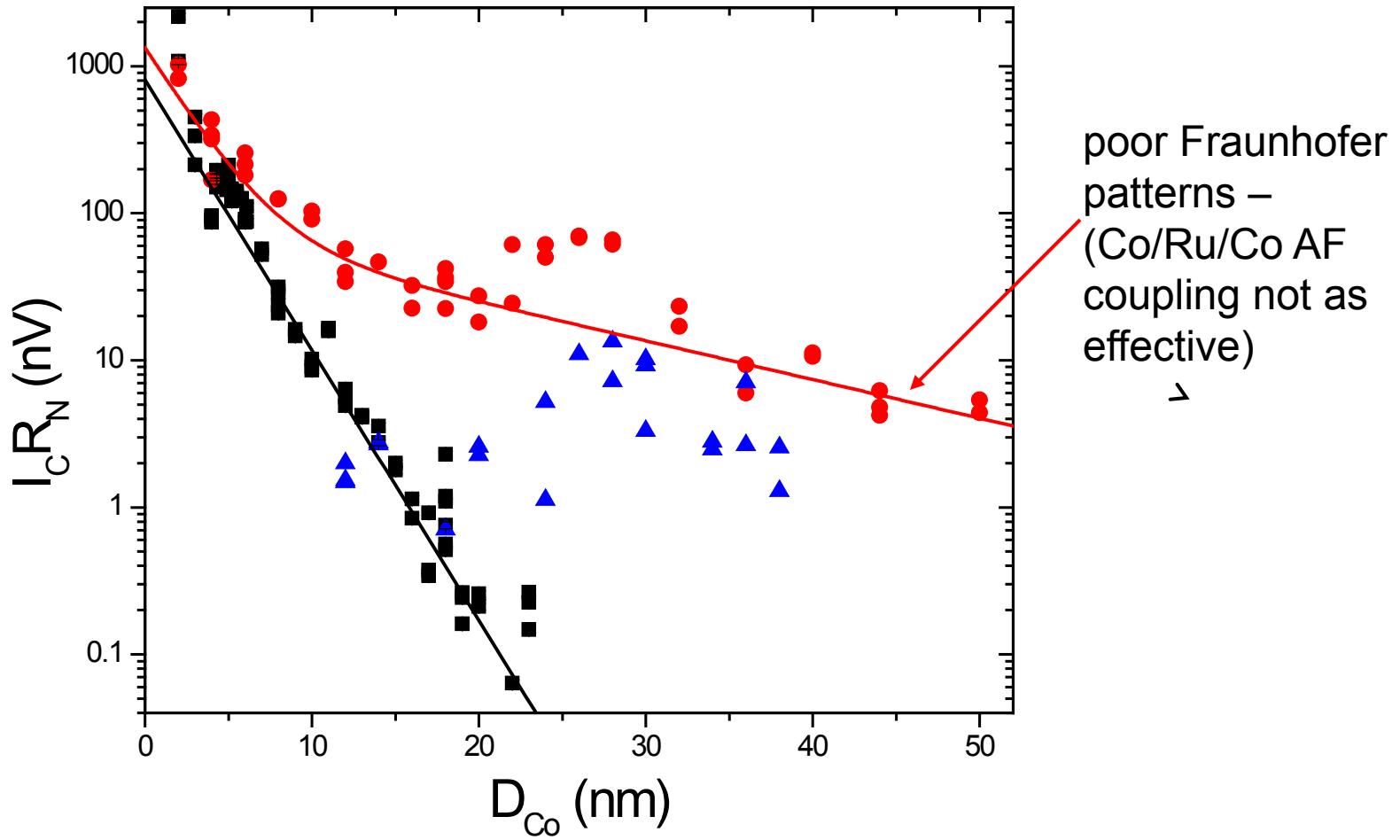


Fix  $D_{Co} = 20 \text{ nm}$  and vary  $d_X$

# Control amplitude of triplet with $d_X$

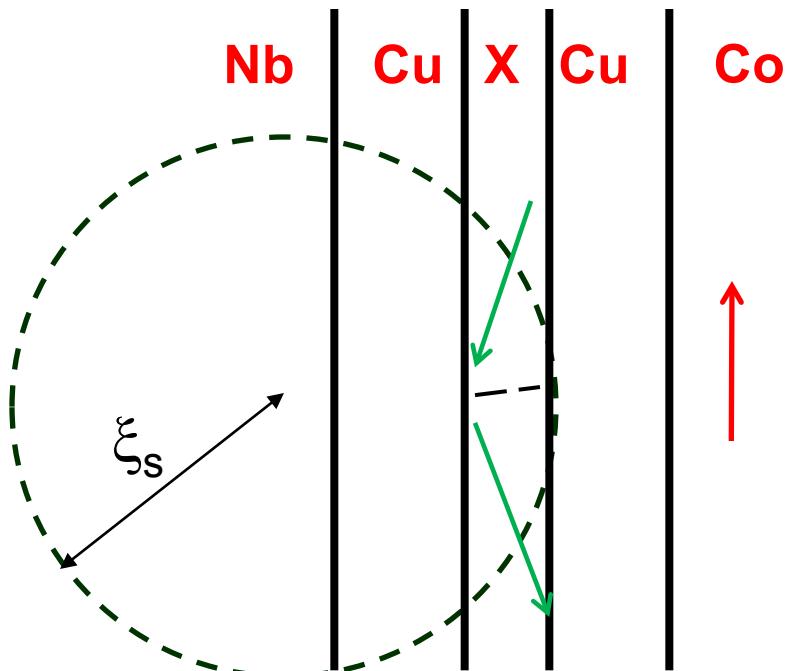


# What happens with thicker Co layers?

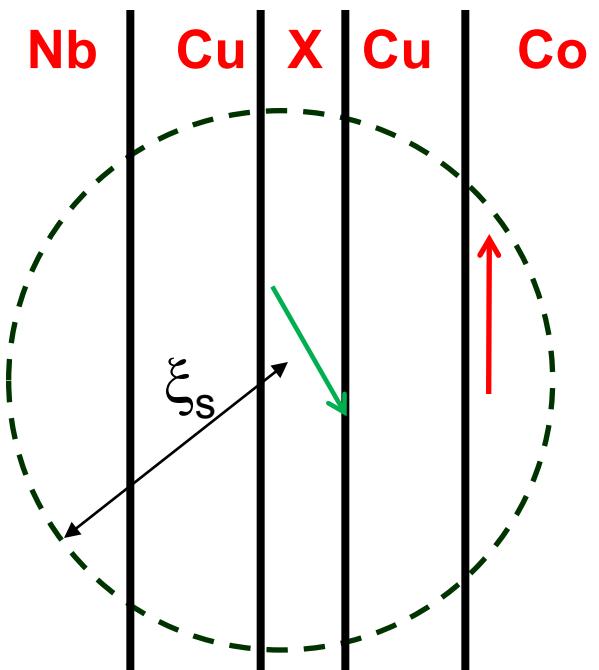


# Mechanism for generating triplet

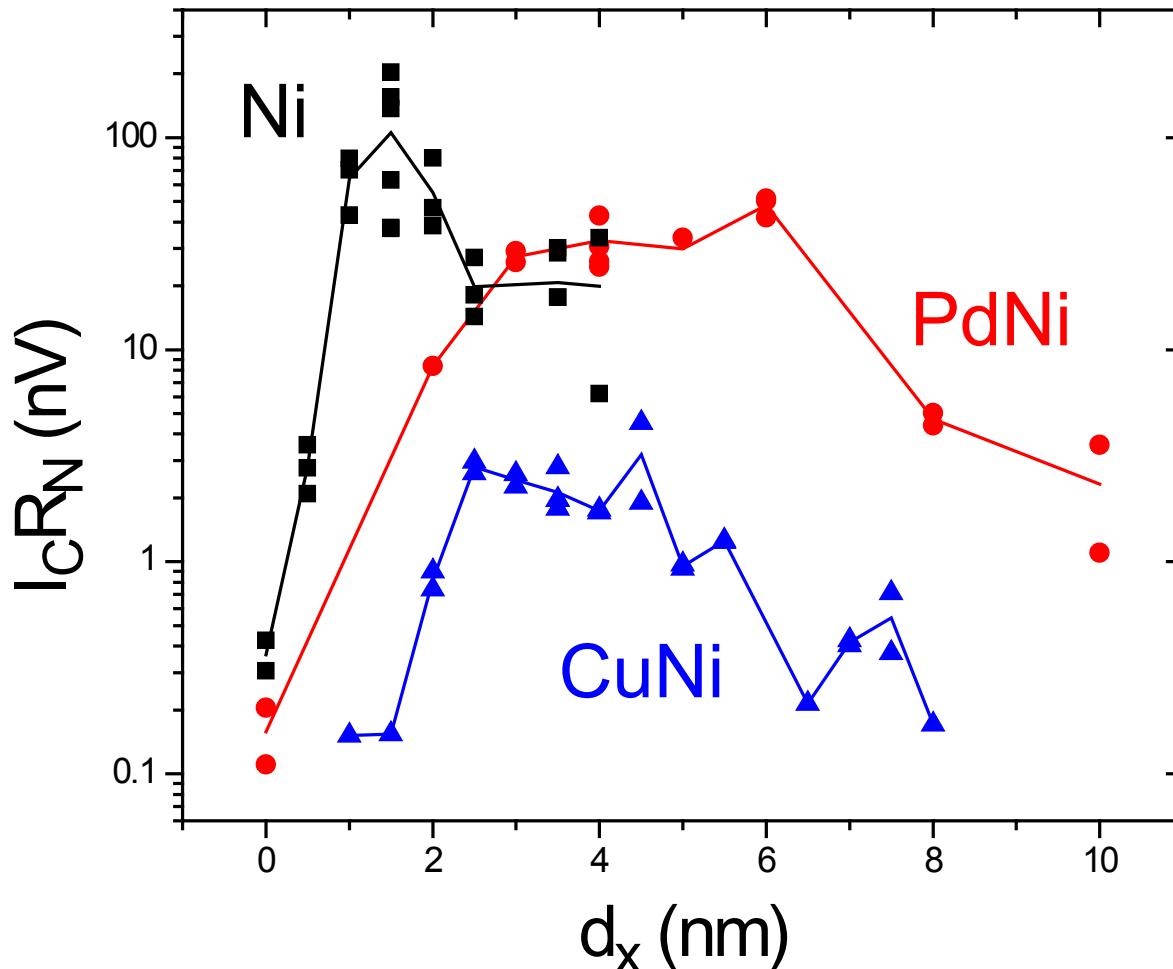
A. Cooper pairs feel non-collinear M between X-layer domains



B. Cooper pairs feel non-collinear M between X and Co layers.

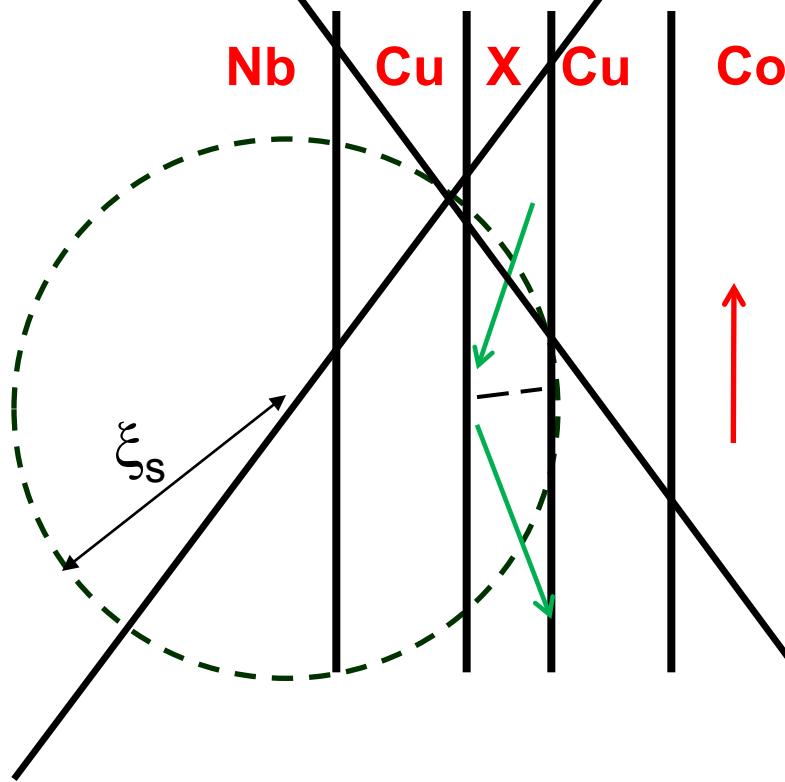


# Pure Ni works well for X layer (no need for inhomogeneous X layer)

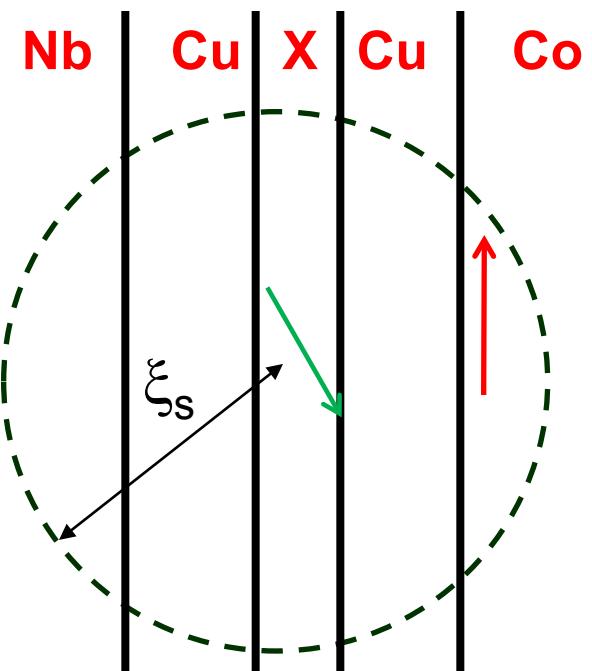


# Mechanism for generating triplet

A. Cooper pairs feel non-collinear M between X-layer domains

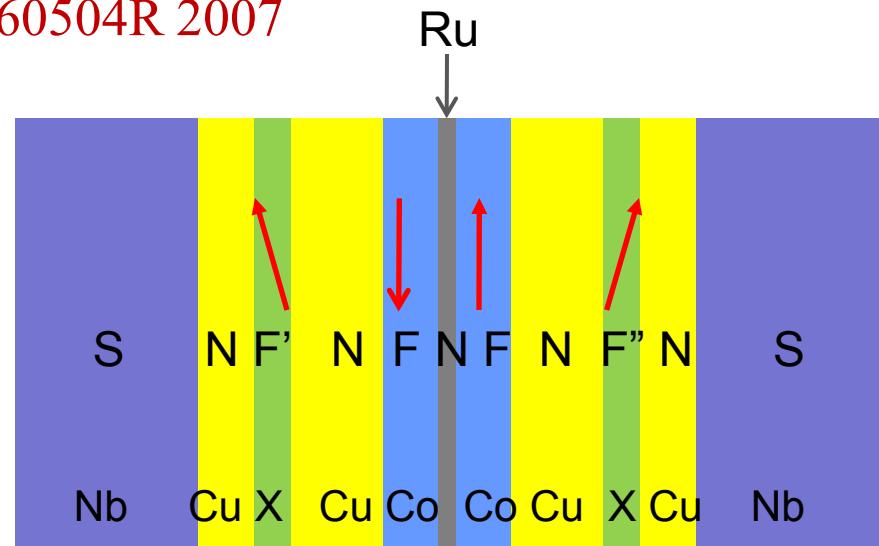
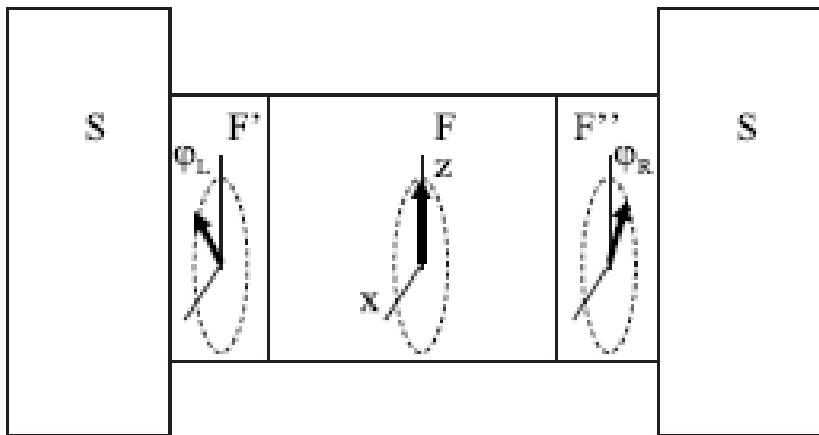


B. Cooper pairs feel non-collinear M between X and Co layers.



# Mechanism for generating triplet

M. Houzet and A. I. Buzdin, PRB **76**, 060504R 2007



$$X = \text{PdNi, CuNi, or Ni}$$

$F'$  and  $F''$  are not required to be inhomogeneous

# Microscopic mechanism for triplet generation

(from discussion with M. Eschrig)

S

$$|\psi\rangle = |0,0\rangle_z = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$$

S  $\uparrow$   
F<sub>1</sub>

$$\begin{aligned} |\psi\rangle &= \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle e^{iQx} - |\downarrow\uparrow\rangle e^{-iQx}) \\ &= \frac{1}{\sqrt{2}}[|0,0\rangle + |1,0\rangle_z \sin(Qx)] \end{aligned}$$

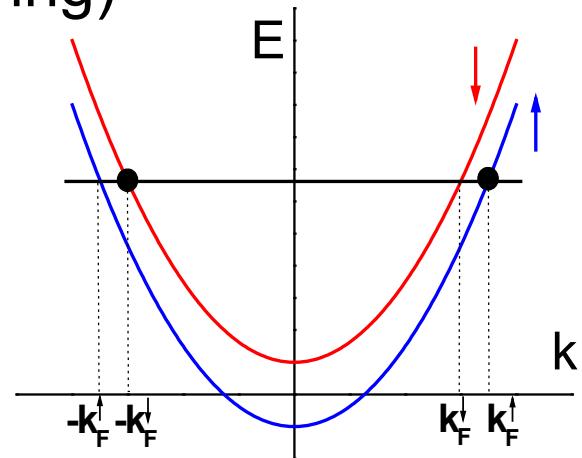
$$Q = k_F^\uparrow - k_F^\downarrow$$

S  $\uparrow$   
F<sub>1</sub>  $\nearrow$   
F<sub>2</sub>

$$|1,0\rangle_z \rightarrow \begin{cases} |1,1\rangle_\theta = |\uparrow\uparrow\rangle_\theta \\ |1,0\rangle_\theta = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle) \\ |1,-1\rangle_\theta = |\downarrow\downarrow\rangle_\theta \end{cases}$$

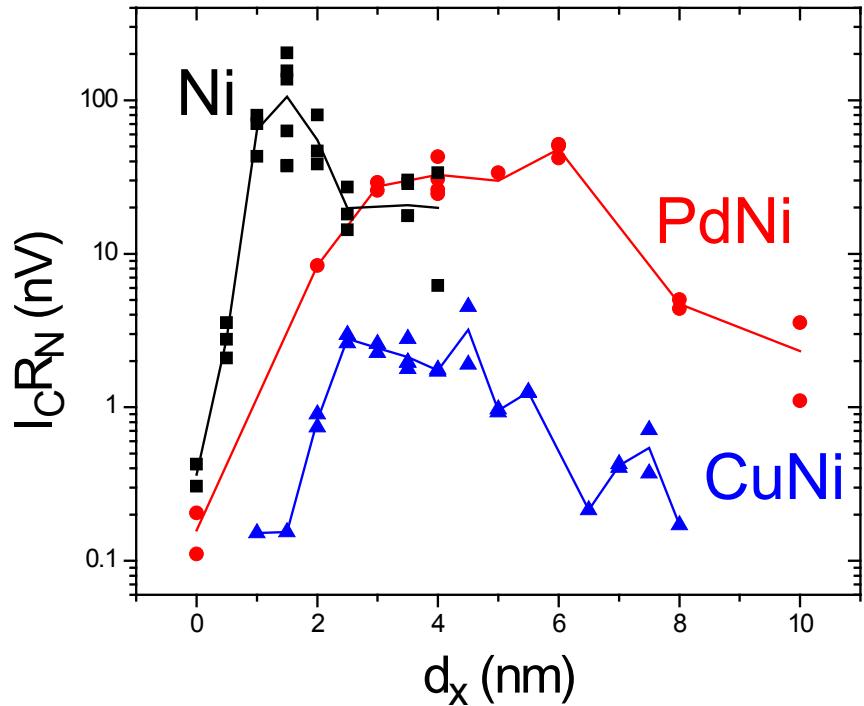
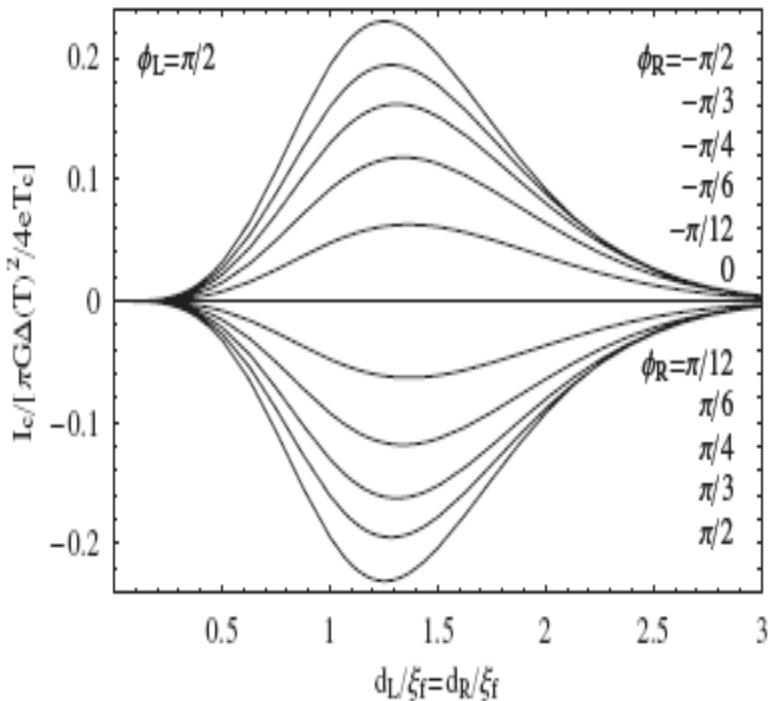
short-range triplet component

long-range triplet components



# Optimization of triplet generation

M. Houzet and A. I. Buzdin, PRB **76**, 060504R 2007

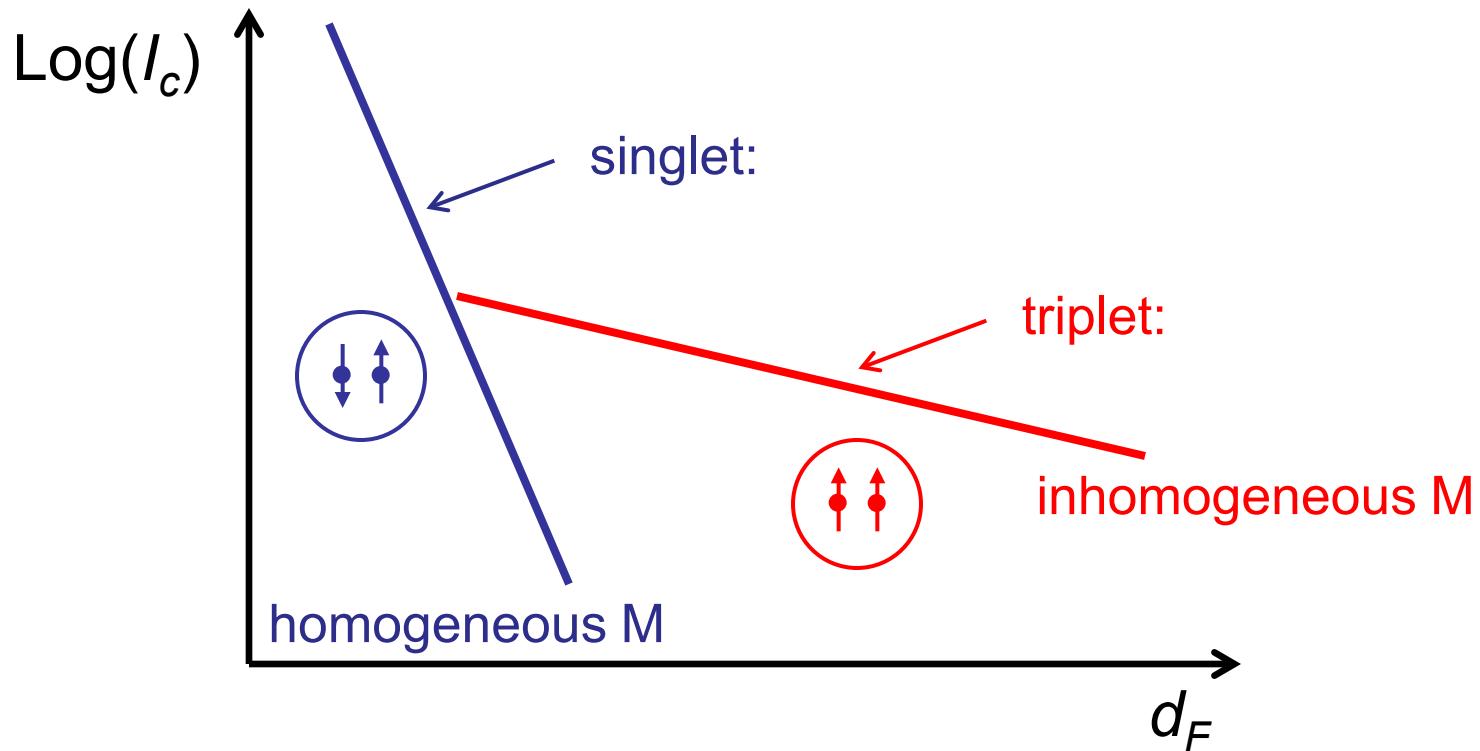


Triplet contribution to the critical current  
is observed only for  $d_x \approx (0.5\text{--}2.5)\xi_F$

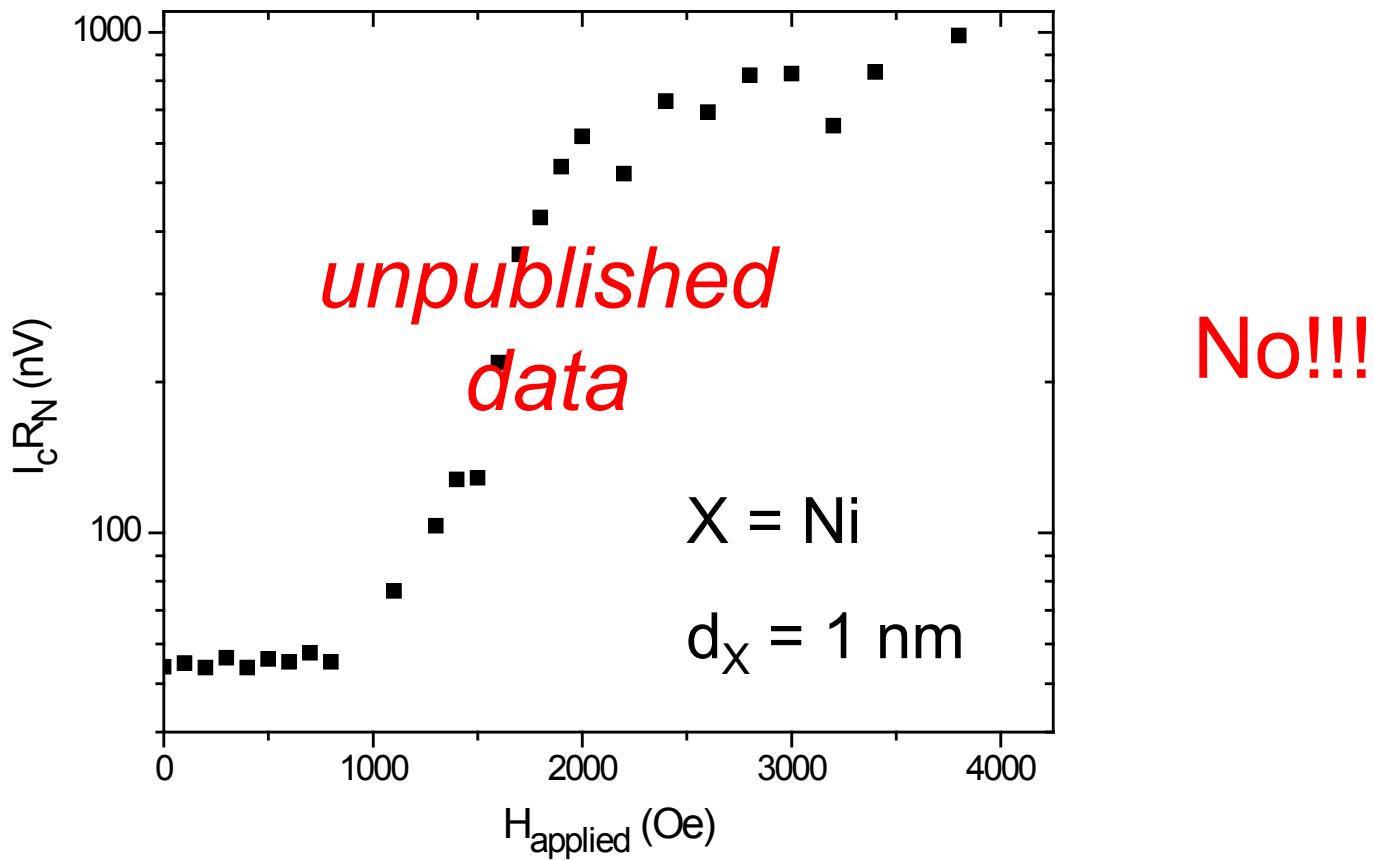
$$E_{ex}^{Ni} > E_{ex}^{PdNi} > E_{ex}^{CuNi}$$

$$\xi_F^{Ni} < \xi_F^{PdNi} < \xi_F^{CuNi}$$

# Does triplet disappear after we magnetize the samples? (makes M more homogeneous)

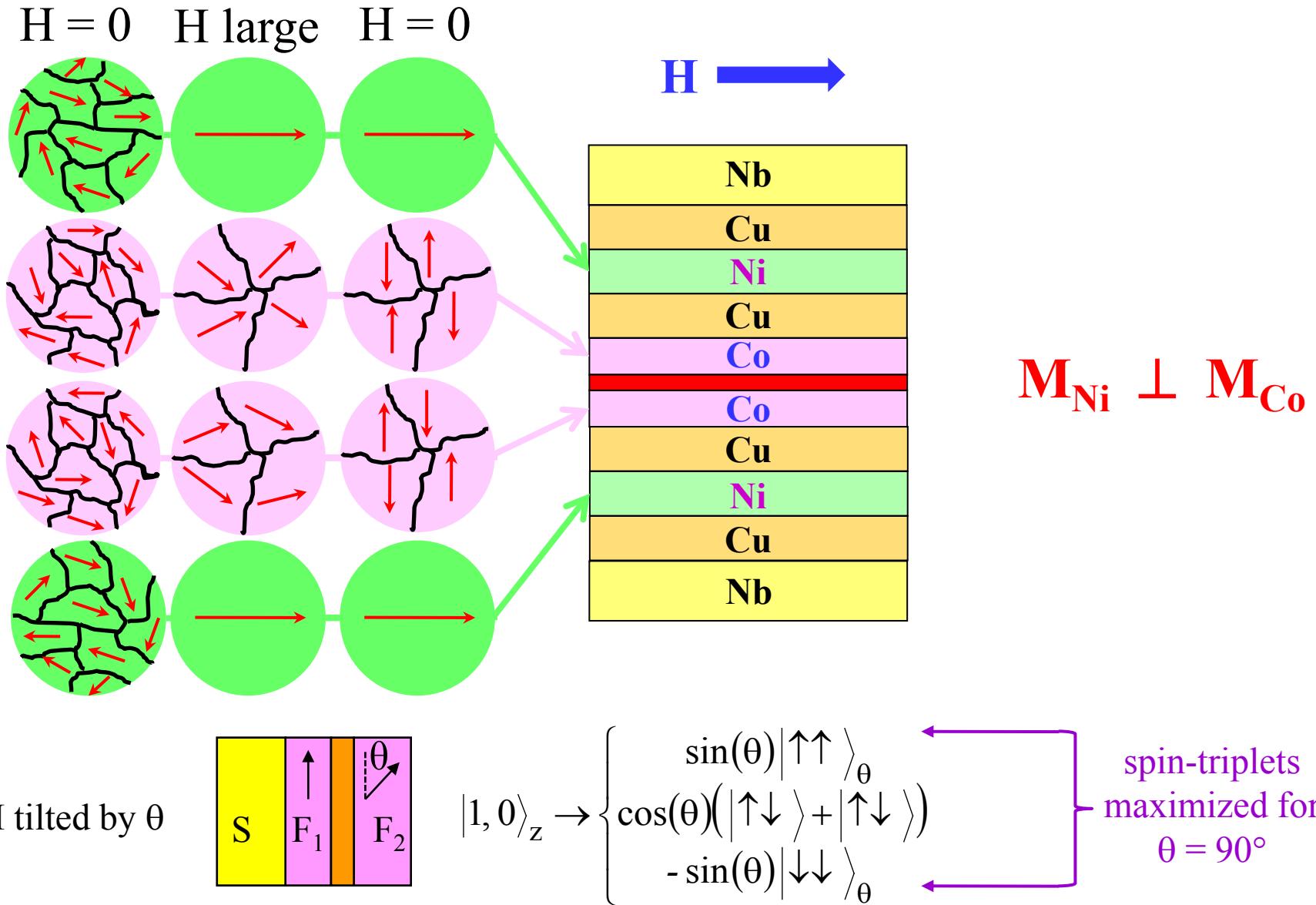


# Does triplet disappear after we magnetize the samples?



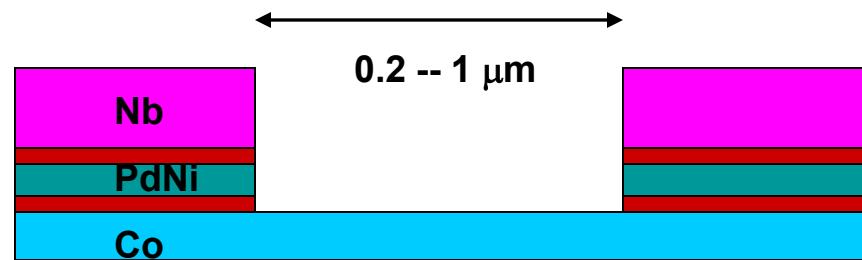
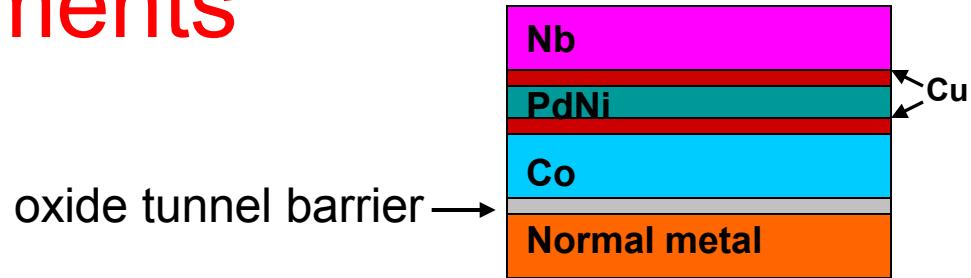
Why does  $I_c$  increase?

# Co/Ru/Co undergoes “spin-flop” transition



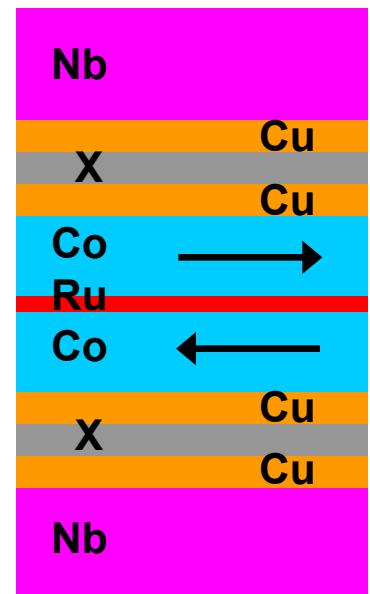
# Future Experiments

- Tunneling
  - Energy dependence of triplet pair correlations
  - Independent signature of odd-frequency triplet
  - No need for Ru layer
- Lateral geometry
  - Longer distances in F
  - Measure proximity effect & Josephson effect



# Summary

- New type of Fermion pairing occurs in S/F systems: odd-frequency, spin-triplet, s-wave.
  - Triplet  $\Rightarrow$  long-range penetration in F
  - S-wave  $\Rightarrow$  insensitive to disorder
- S/F/S Josephson junctions with PdNi and Co: clear signature of spin-triplet supercurrent
  - Separately optimize generation and propagation
- Stay tuned for future results!



# References

For a general introduction to this subject, read  
“Spin-Polarized Supercurrents for Spintronics” by  
Matthias Eschrig in Physics Today, January 2011:

See also the “News & Views” commentary by Teun  
Klapwijk, “Magnetic nanostructures: Supercurrents  
in ferromagnets” in Nature Physics **6**, 329 (2010).