

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

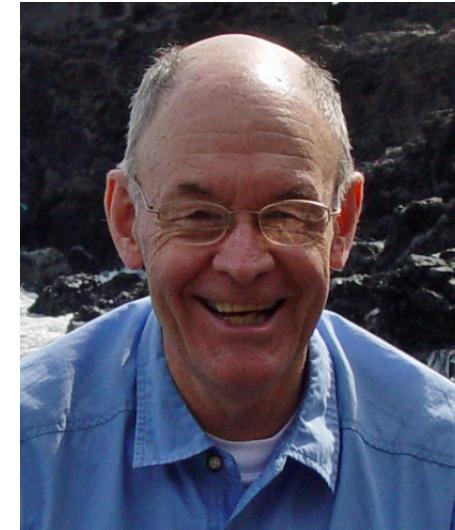
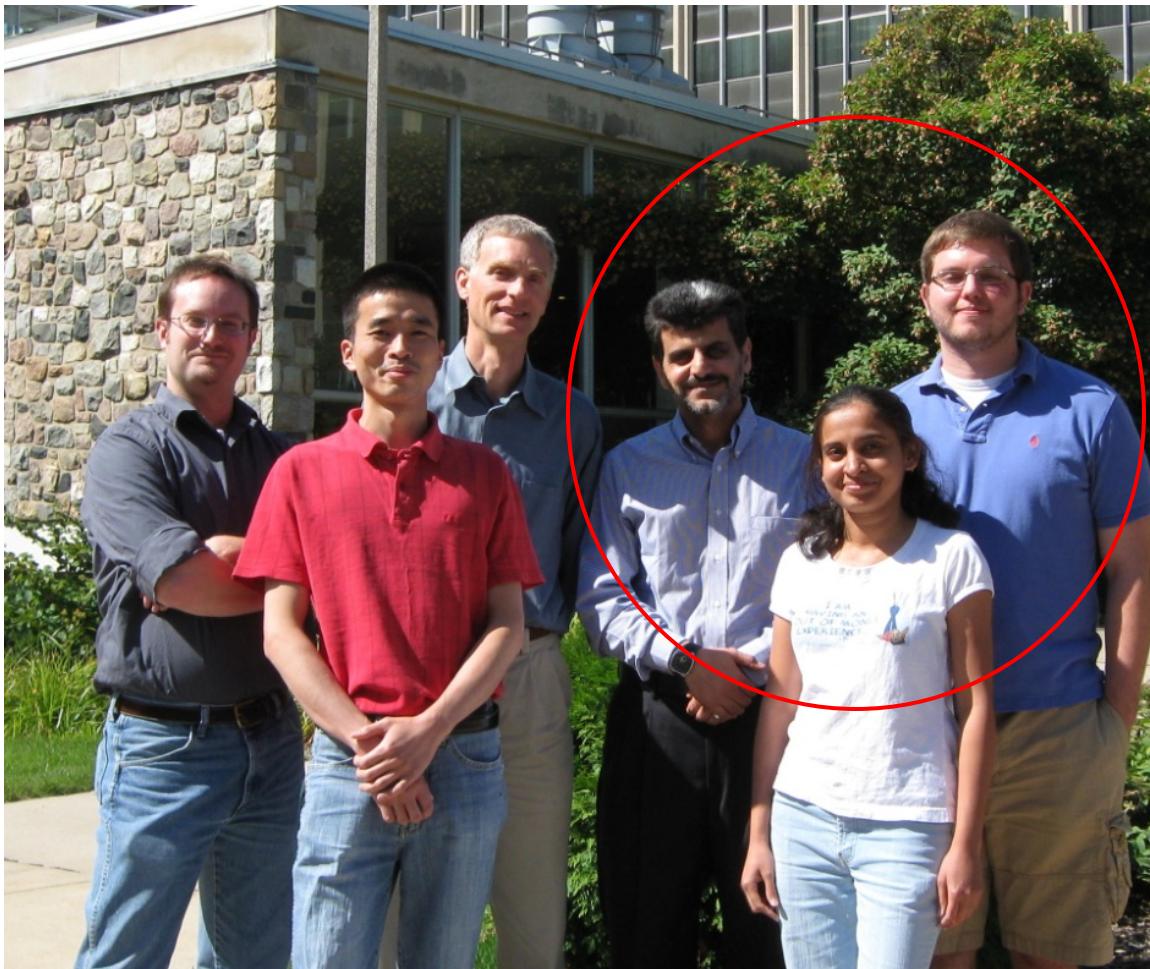
Norman Birge*, Michigan State University

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 ${}^3\text{He}$ Sr_2RuO_4
$\ell=2$ (d-wave)	high- T_c	X

Proposal by Berezinskii (1974):

New model of the anisotropic phase of superfluid He³

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 He³, 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 ^3He Sr_2RuO_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 ${}^3\text{He}$ Sr_2RuO_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	S/F
$\ell=1$	S/F	superfluid ^3He Sr_2RuO_4
$\ell=2$	high- T_c	S/F

especially
interesting

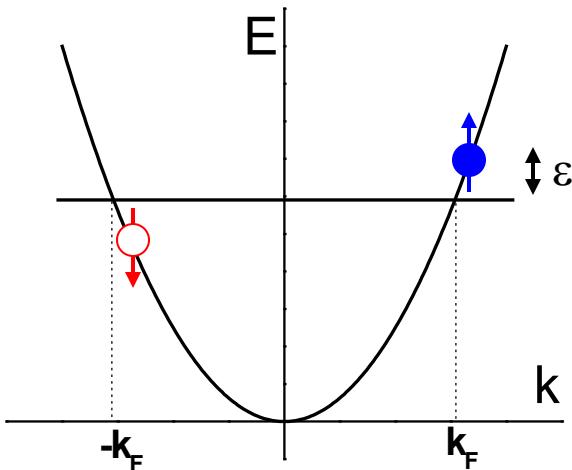
Bergeret, Volkov & Efetov, PRL 90, 117006 (2003); RMP 77, 1321 (2005)

Outline

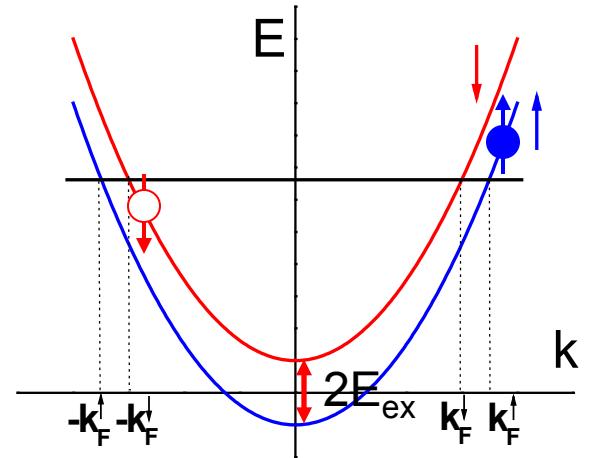
- Proximity effect in S/N and S/F systems
- Prediction: Spin-triplet pair correlations
- Our search: S/F/S Josephson junctions
 - PdNi – a weak ferromagnet
 - Co/Ru/Co – a synthetic antiferromagnet
 - Combining the best of both materials
- Conclusions and Future Prospects

Andreev Reflection: N/S vs. F/S

S/N



S/F



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

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

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

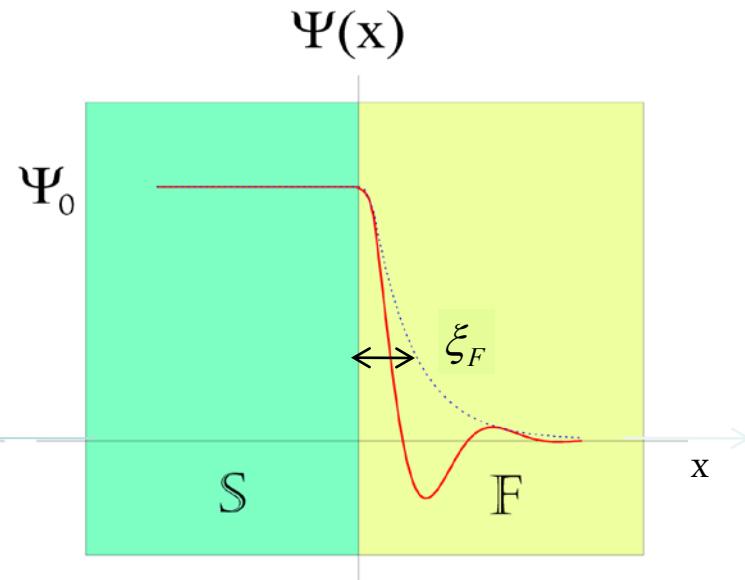
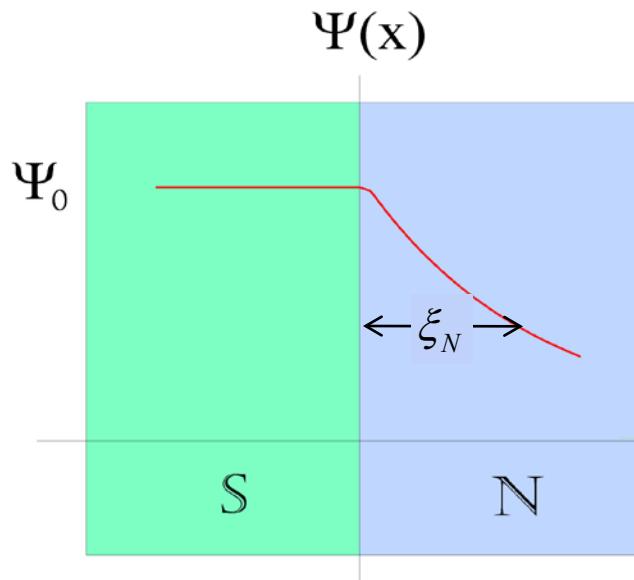
$$\xi_N = \sqrt{D_N \tau} = \sqrt{\frac{\hbar D_N}{2\pi k_B T}} \quad \text{diffusive}$$

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



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

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

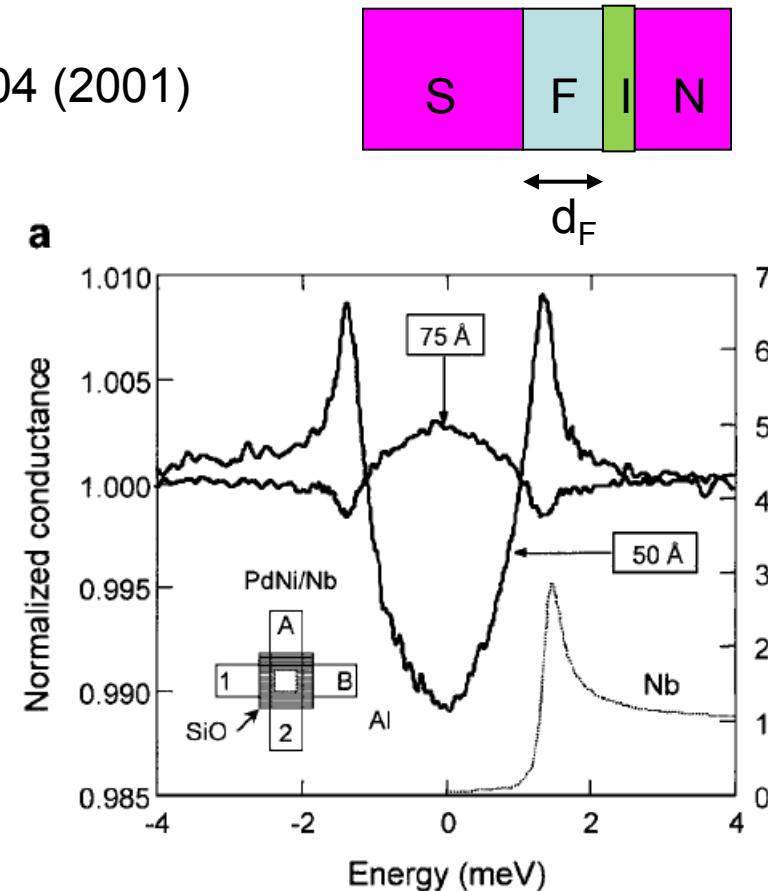
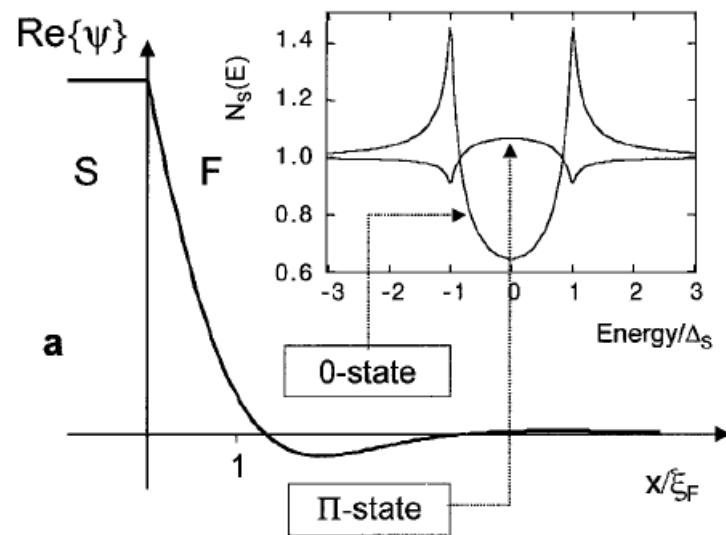
$$\Psi(x) = \Psi_0 \exp(-x/\xi_N)$$

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

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)

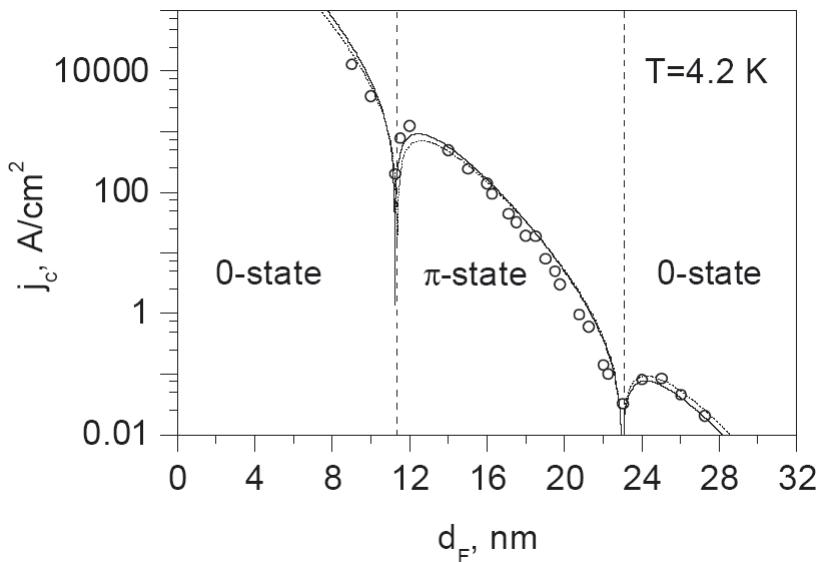


How to detect the oscillating pair correlation function?

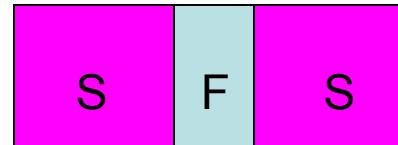
2. Measure critical current of S/F/S Josephson junction, as function of d_F

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

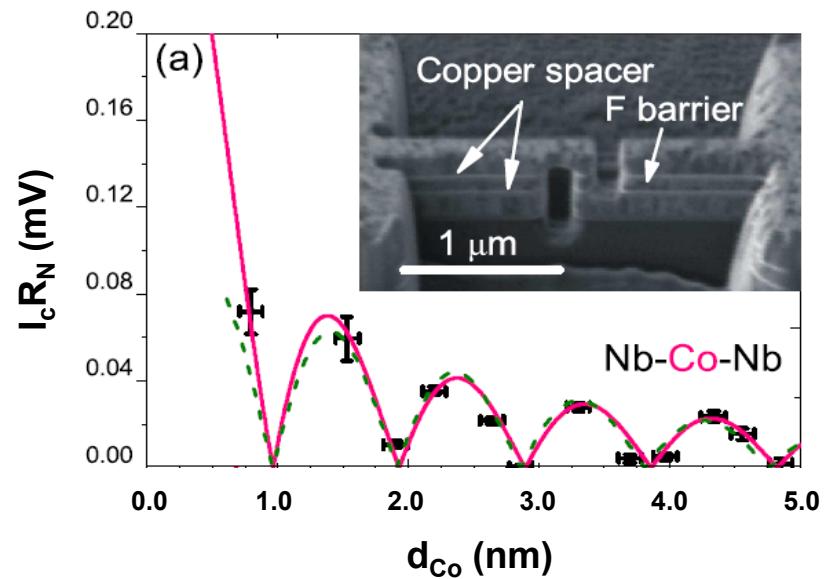
Weak F: Cu₄₈Ni₅₂ alloy



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

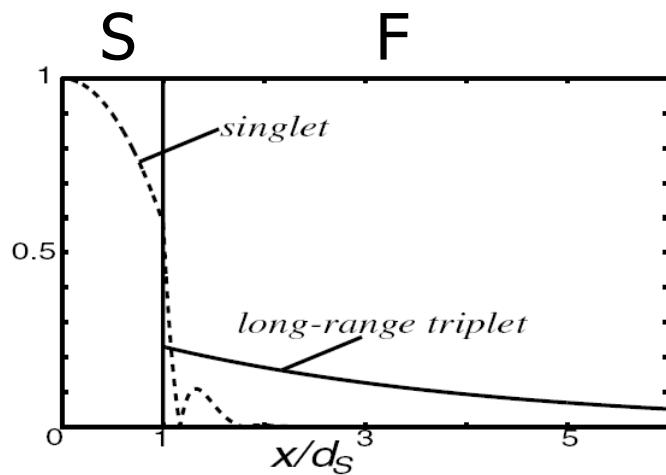
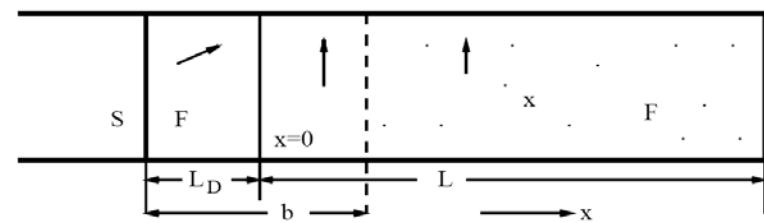
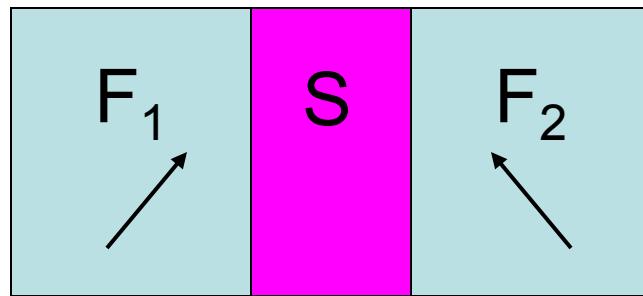


Strong F: Co



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)

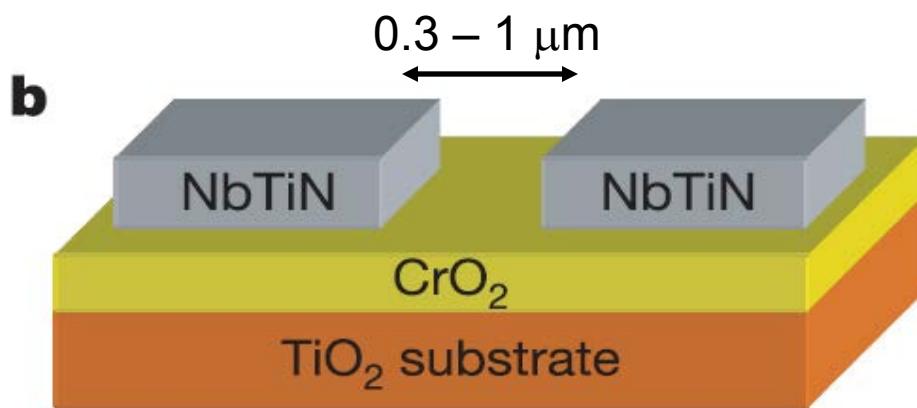
Features of odd-frequency, spin-triplet pair correlations

	$S=0$	$S=1$
$\ell=0$	BCS	S/F
$\ell=1$	S/F	Sr_2RuO_4
$\ell=2$	high- T_c	S/F

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

Possible observation of triplet:

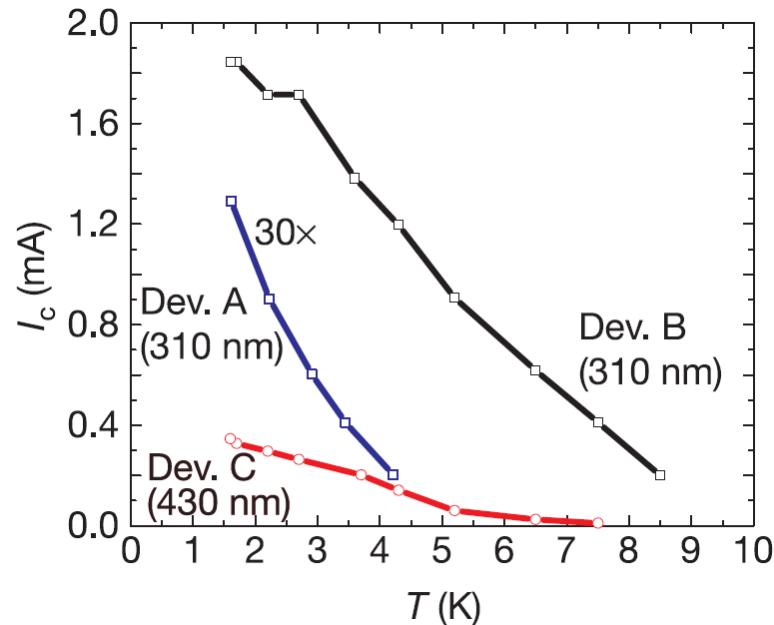
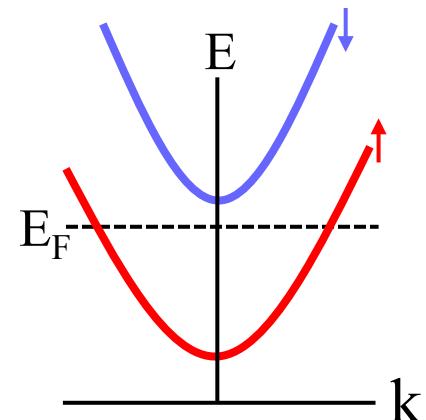
Keizer, Goennenwein, Klapwijk, Miao, Xiao, Gupta,
Nature **439**, 825 (2006)



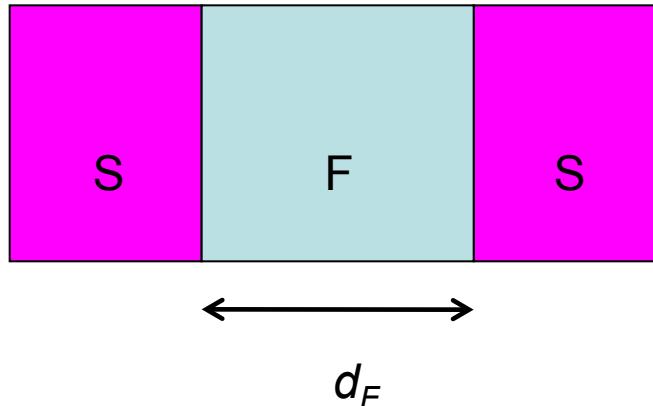
Long-range propagation of supercurrent,
but large sample-to-sample variations in I_c .

Reproduced last year by J. Aarts

CrO₂ is a “half metal”



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



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

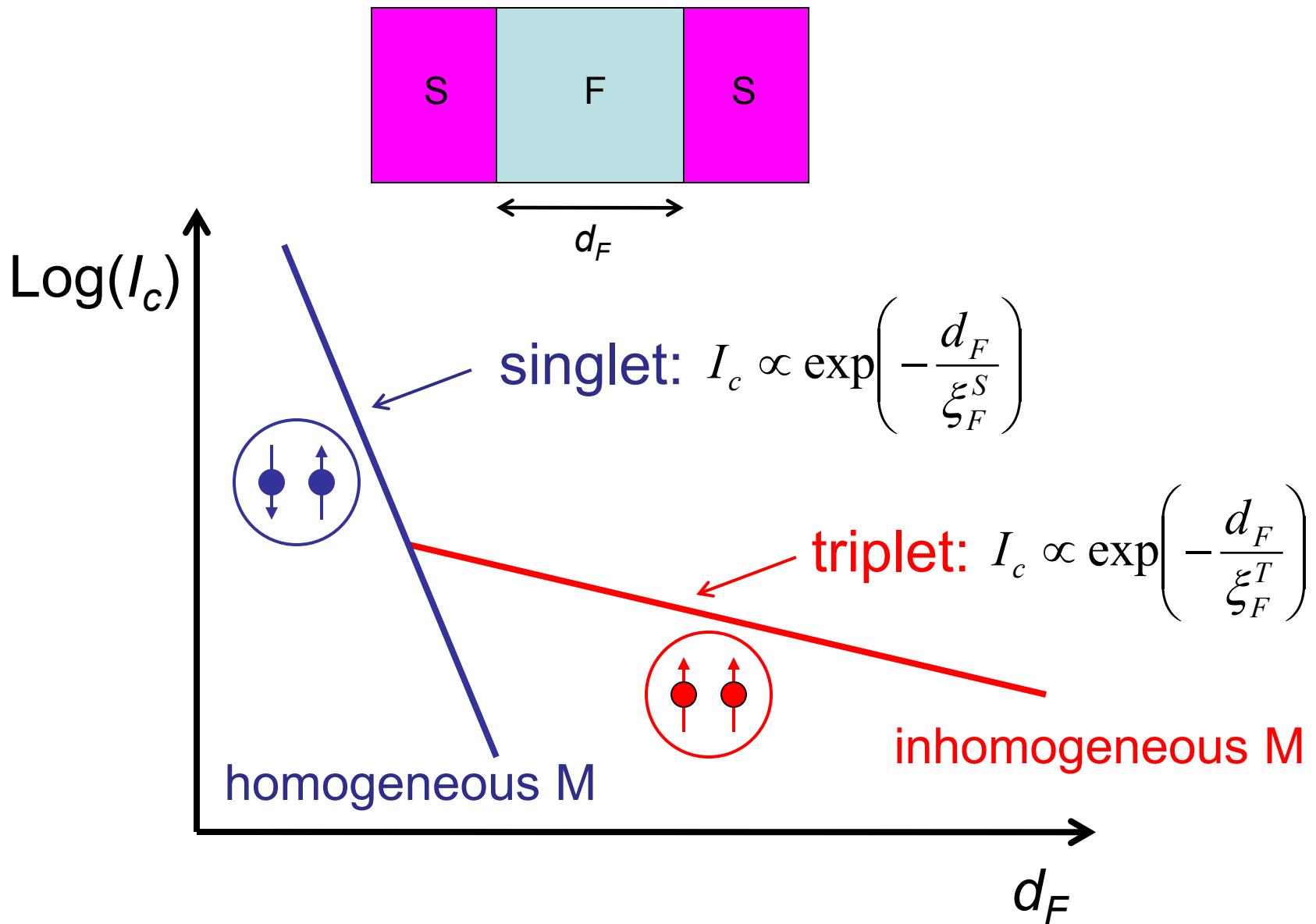
suppress
singlet

triplet limited by
spin-flip scattering

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

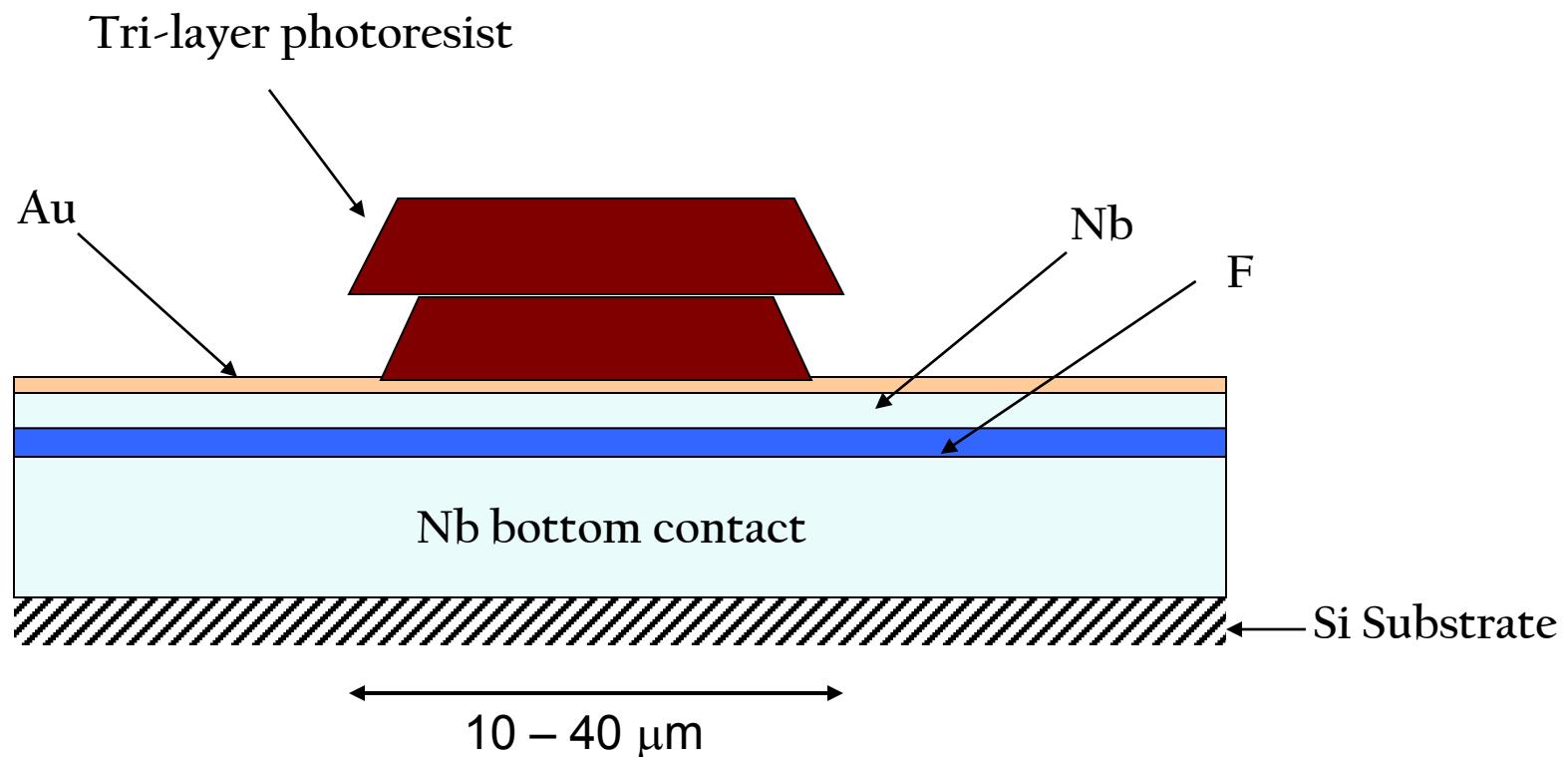
$$\xi_F^T = \sqrt{\frac{\hbar D_F}{2\pi k_B T}}$$

Signature of spin-triplet:



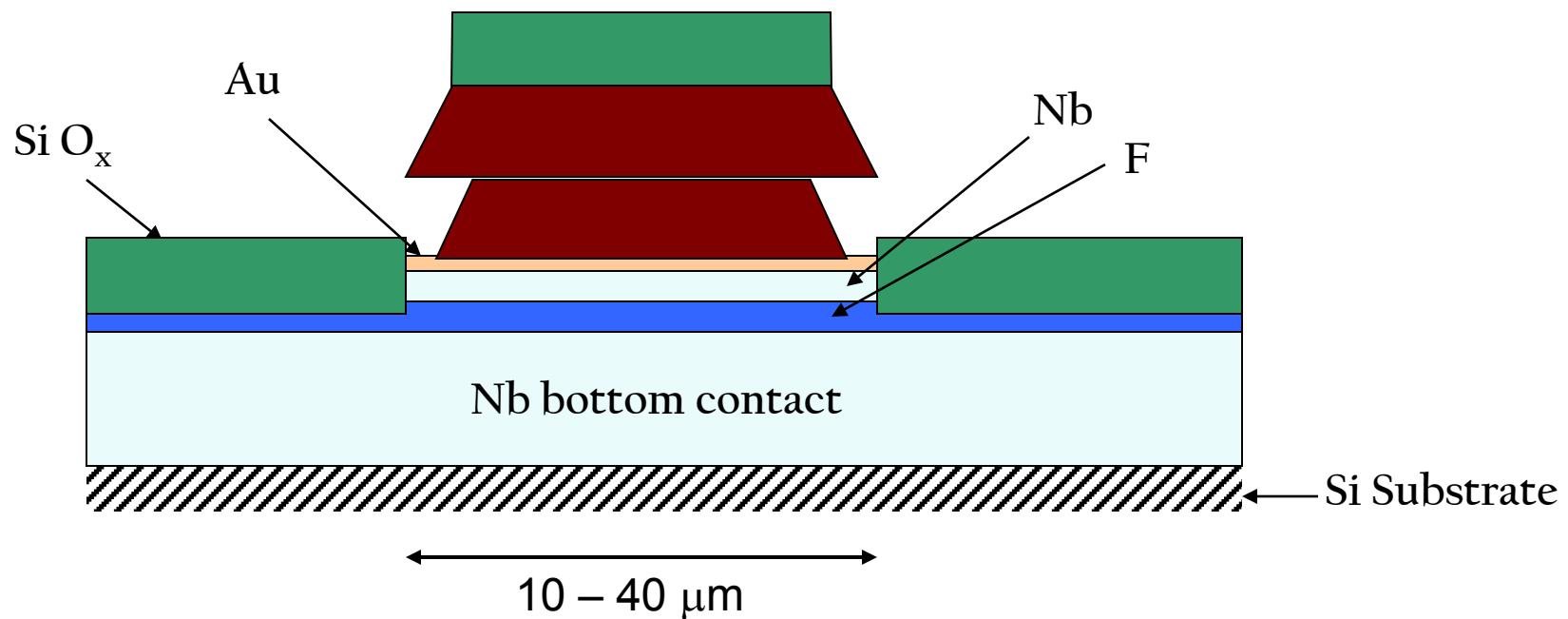
Sample Fabrication

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



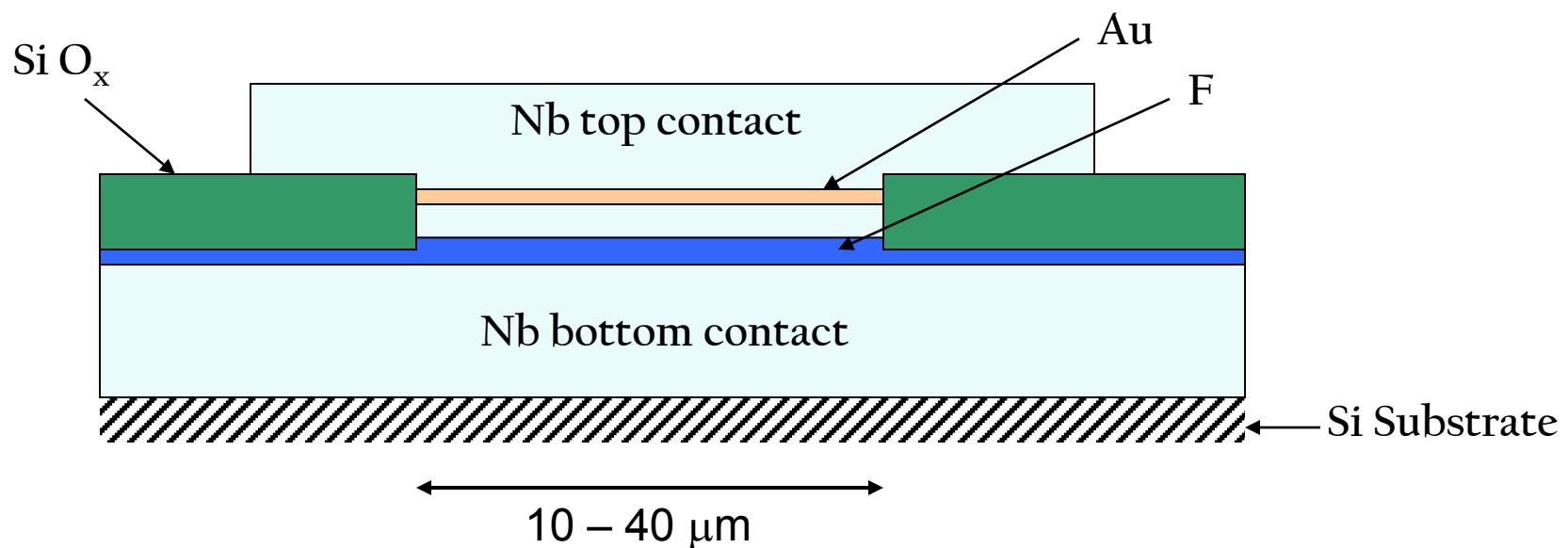
Sample Fabrication

1. Sputter S/F/S
2. Pattern pillars with photolithography
3. Ion mill
4. Deposit SiO_x



Sample Fabrication

1. Sputter S/F/S
2. Pattern pillars with photolithography
3. Ion mill
4. Deposit SiO_x
5. Lift off
6. Deposit top Nb contact



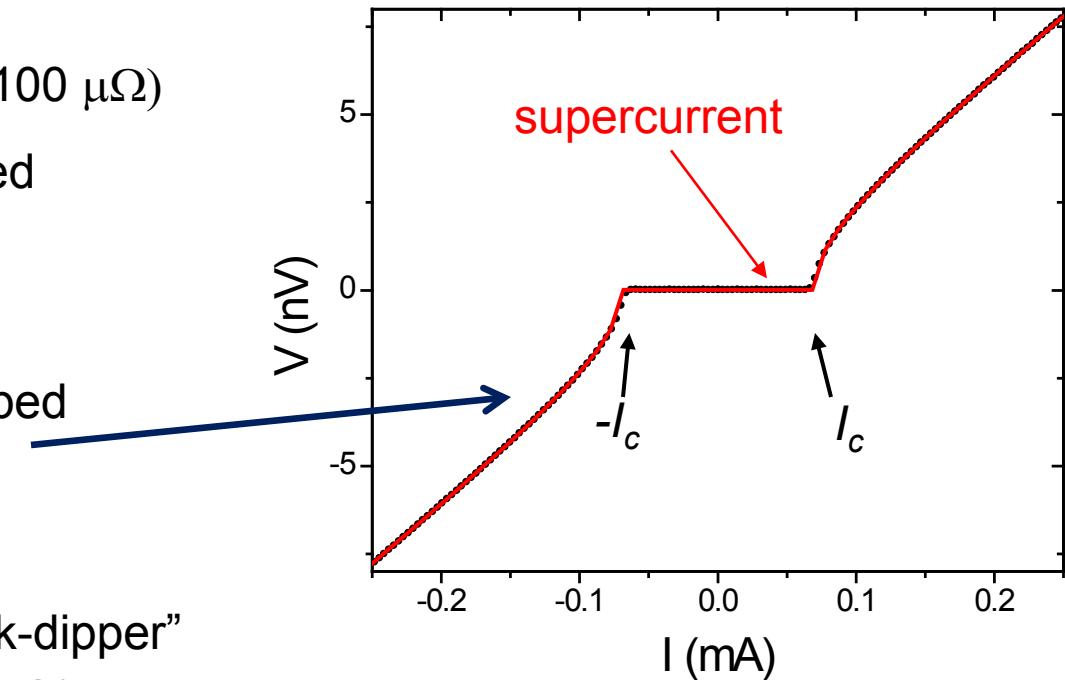
Measurement

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

→ Measure with SQUID-based
current comparator circuit

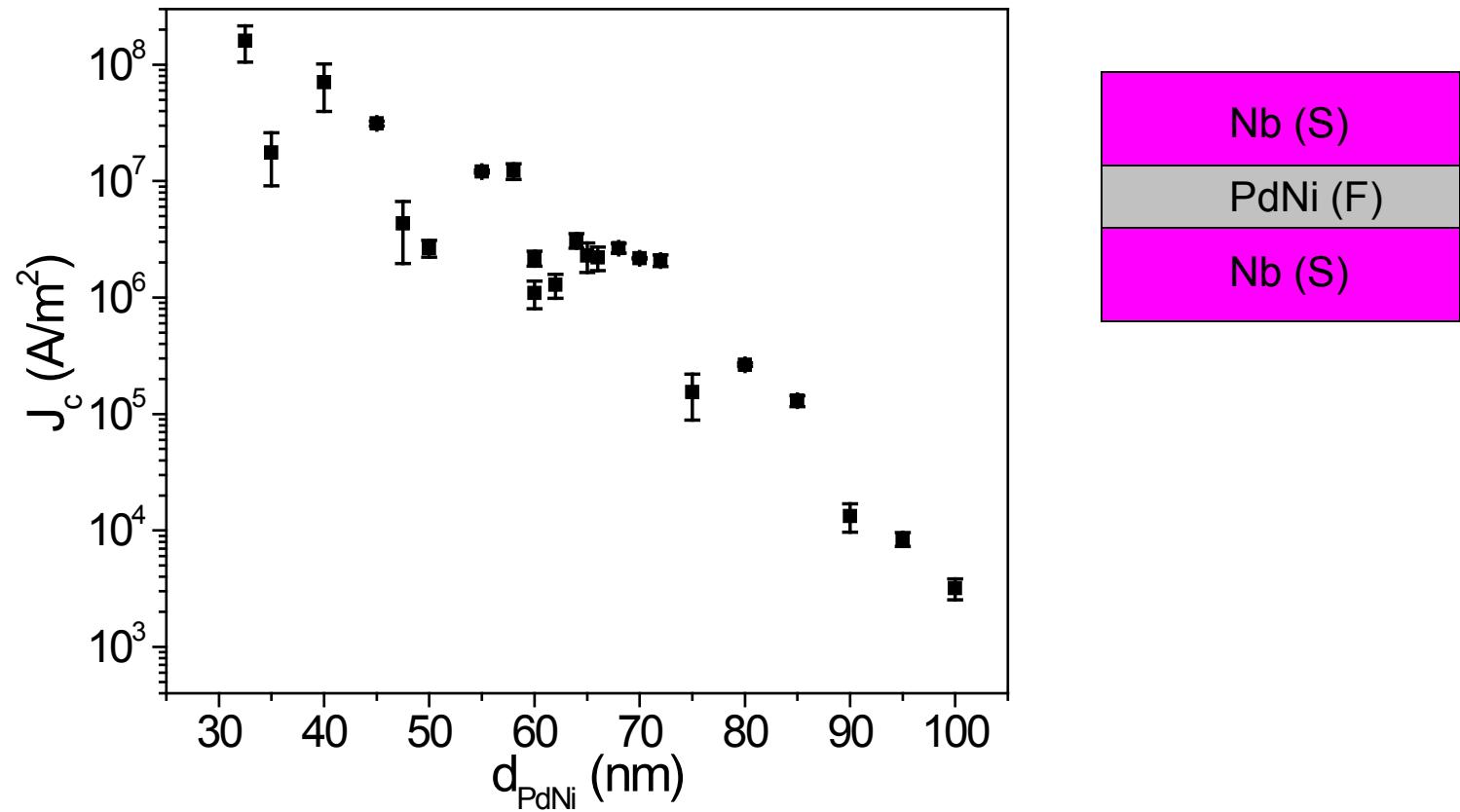
I-V characteristic of overdamped
Josephson junctions

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



$I_c \equiv$ critical current

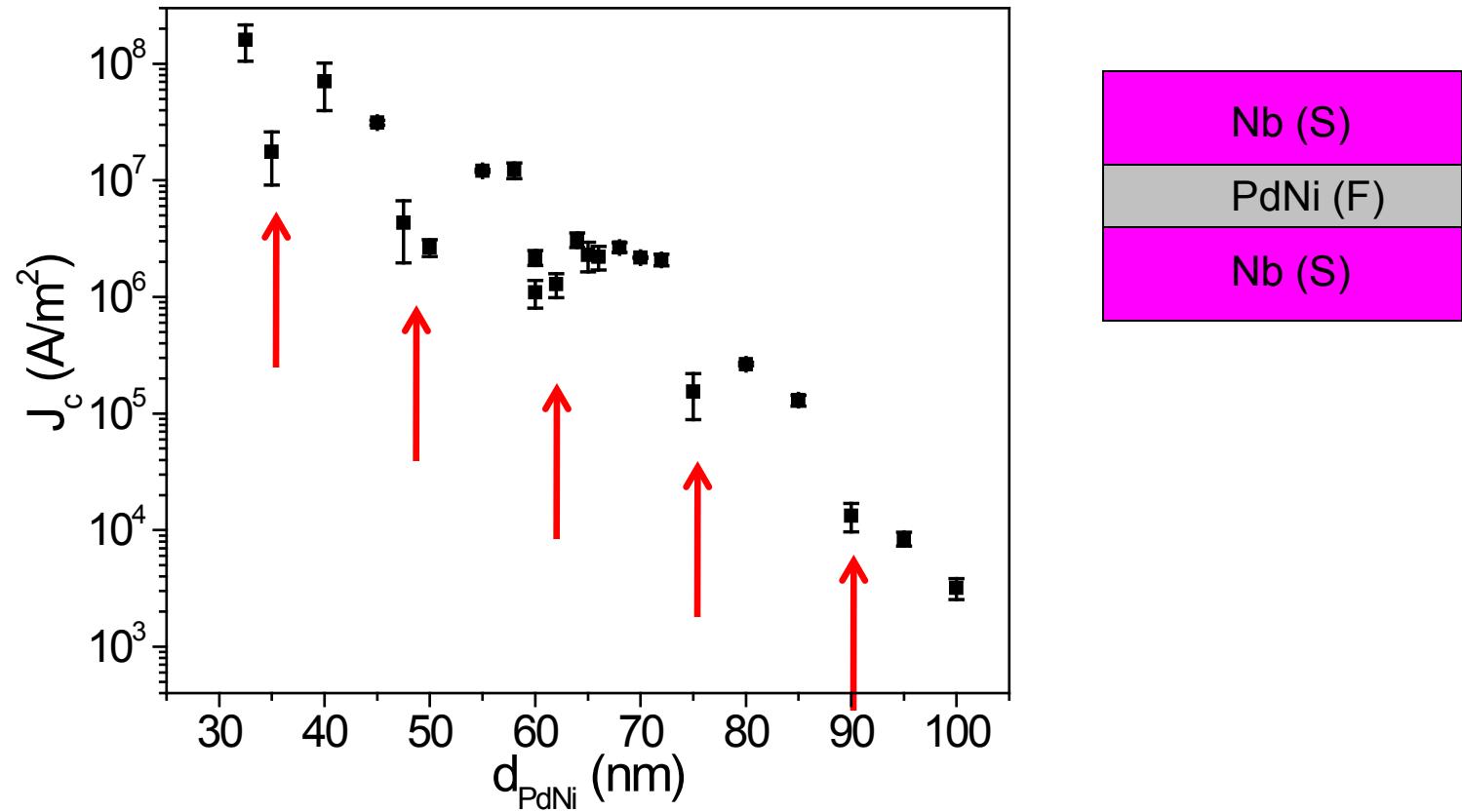
S/F/S junctions with a weak F: Pd₈₈Ni₁₂ alloy



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

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

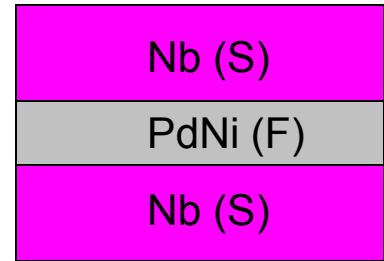
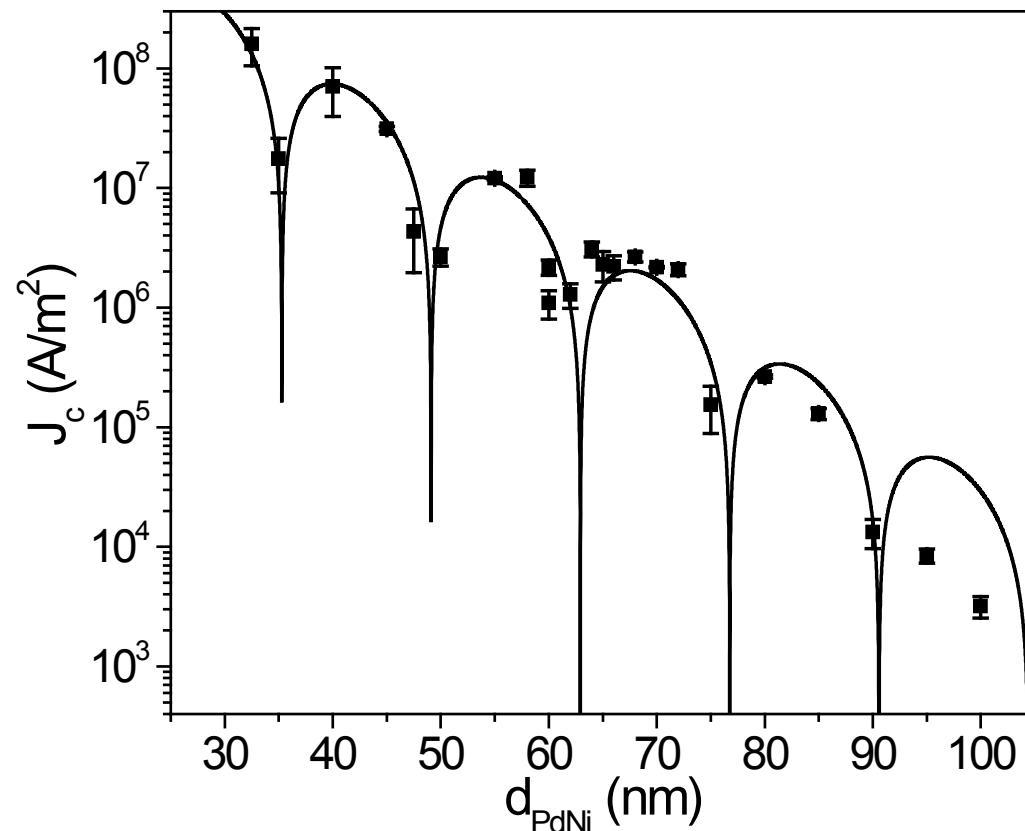
S/F/S junctions with a weak F: Pd₈₈Ni₁₂ alloy



Periodic minima in J_c

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

S/F/S junctions with a weak F: Pd₈₈Ni₁₂ alloy

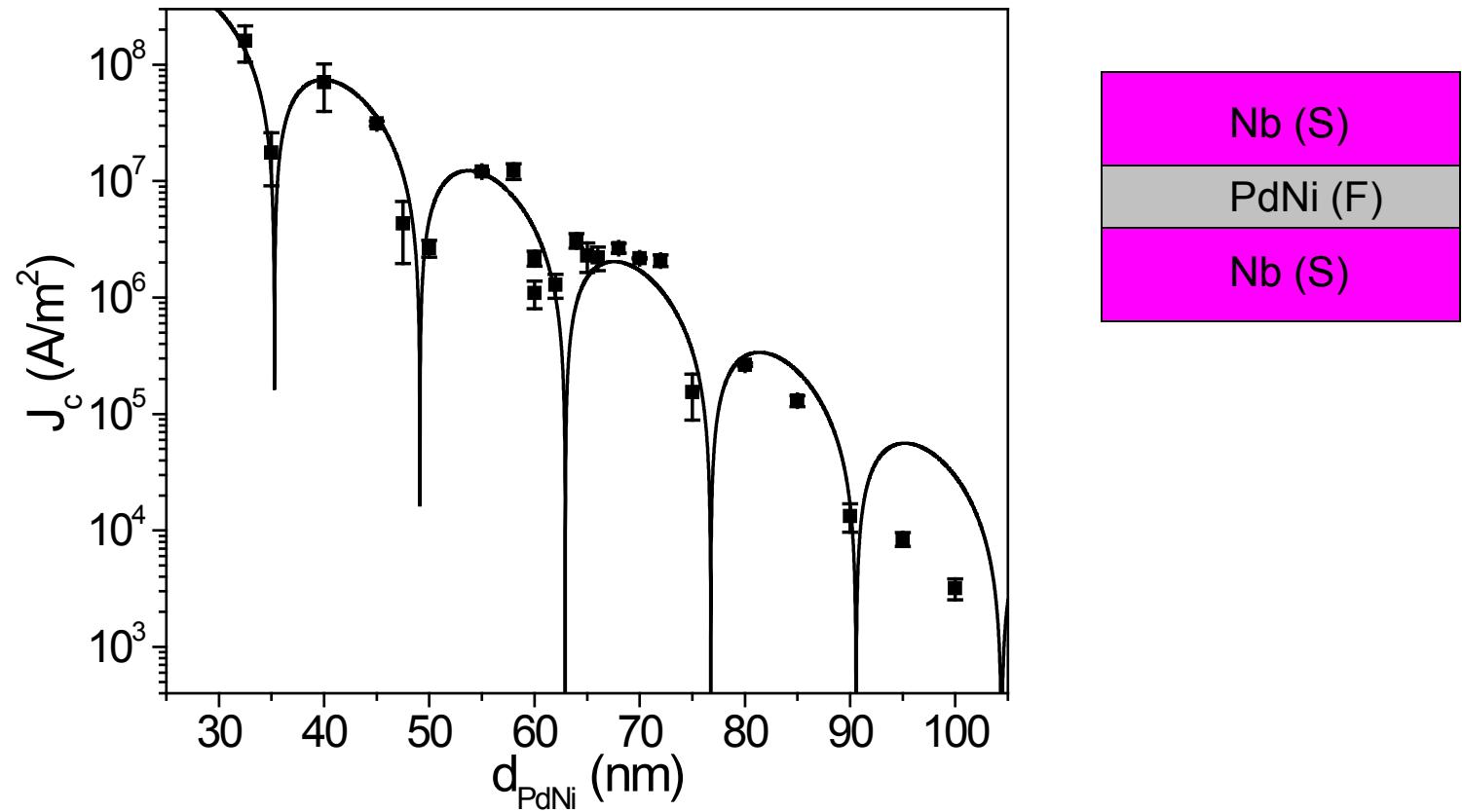


$$I_c \propto \exp(-x/\xi_{F1}) \cos(x/\xi_{F2})$$

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

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

S/F/S junctions with a weak F: Pd₈₈Ni₁₂ alloy



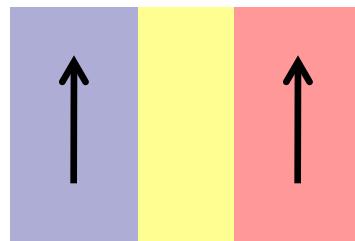
No sign of spin-triplet supercurrent!

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

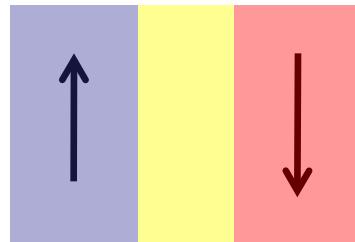
- Not enough non-collinear magnetization
- Magnetic inhomogeneity on wrong length scale
- Too much spin-flip and/or spin-orbit scattering

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

P state



AP state

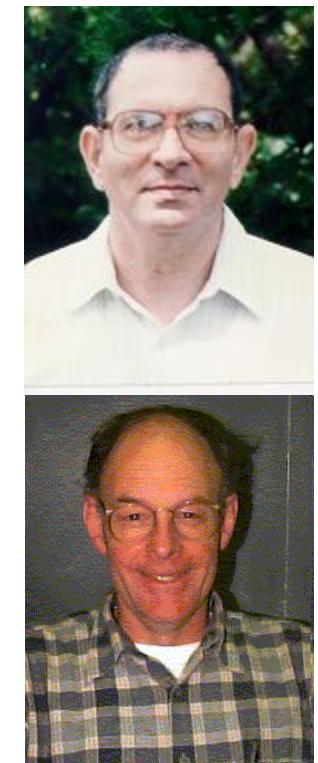


$$R_P < R_{AP}$$

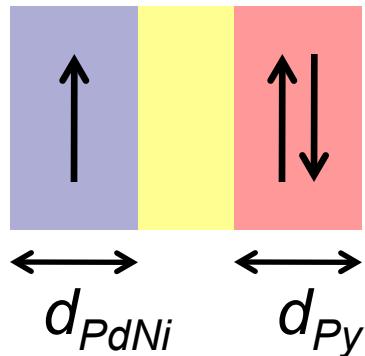
Albert Fert &
Peter Grunberg
2007 Nobel Prize



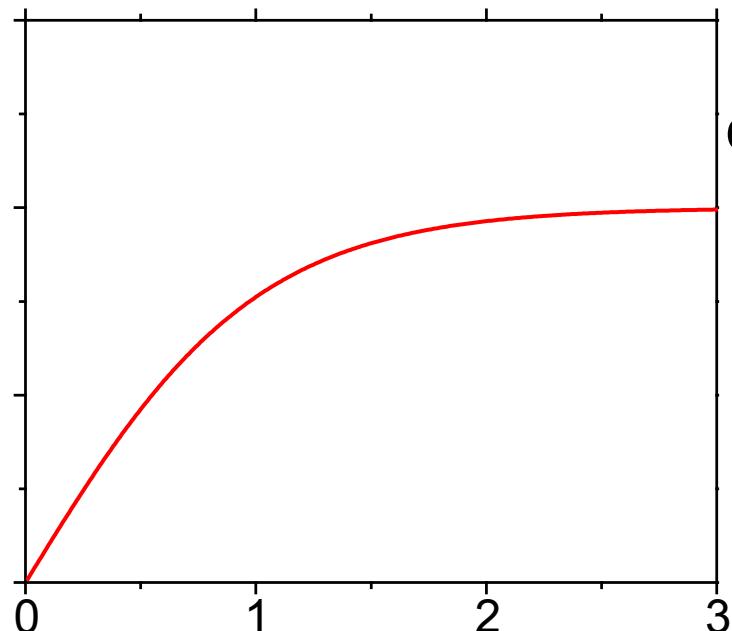
Jack Bass
& Bill Pratt



Dependence of GMR signal on d_{PdNi}



Fix d_{Py} , vary d_{PdNi}

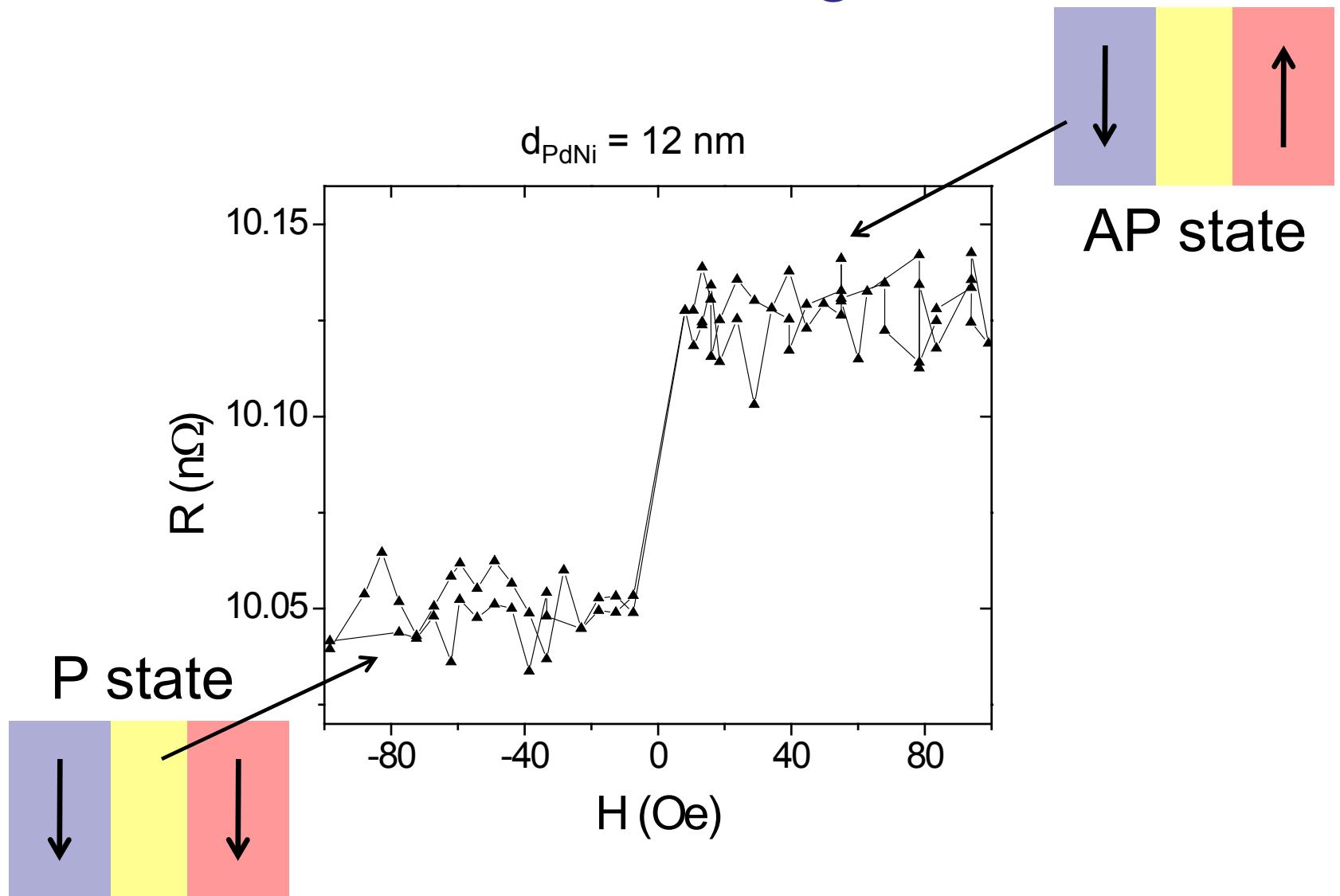


If $d_{PdNi} < l_{PdNi}^{sf}$ $A\Delta R \propto \beta_{PdNi} \rho_{PdNi}^* d_{PdNi}$

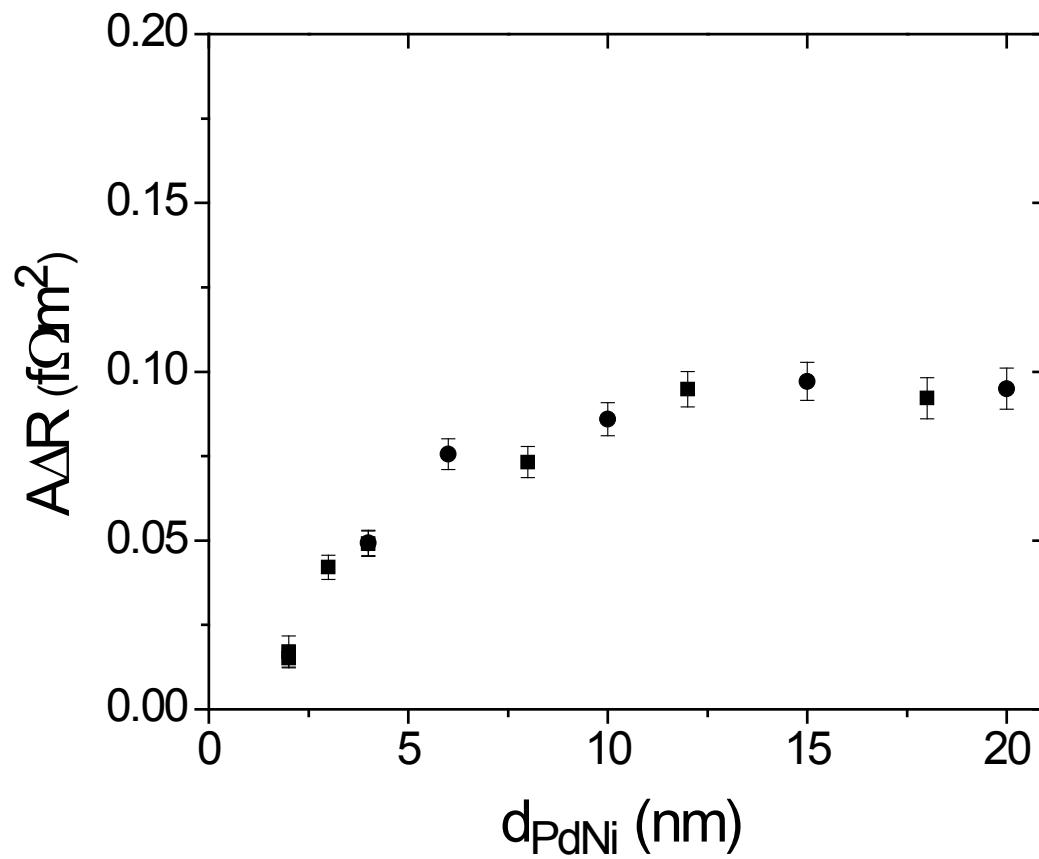
If $d_{PdNi} > l_{PdNi}^{sf}$ $A\Delta R \propto \beta_{PdNi} \rho_{PdNi}^* l_{PdNi}^{sf}$

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

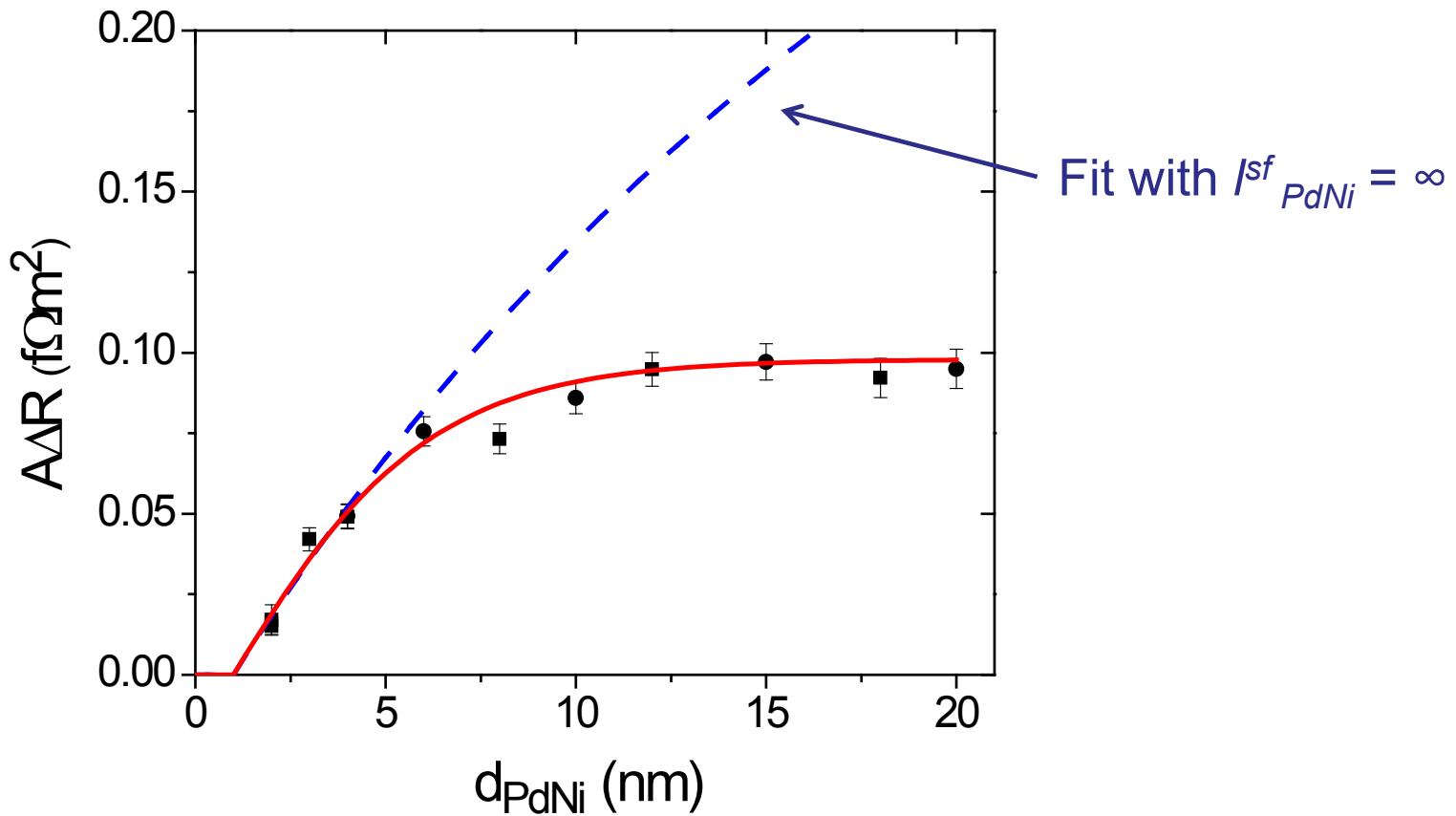
Raw GMR signal



GMR signal vs. d_{PdNi}



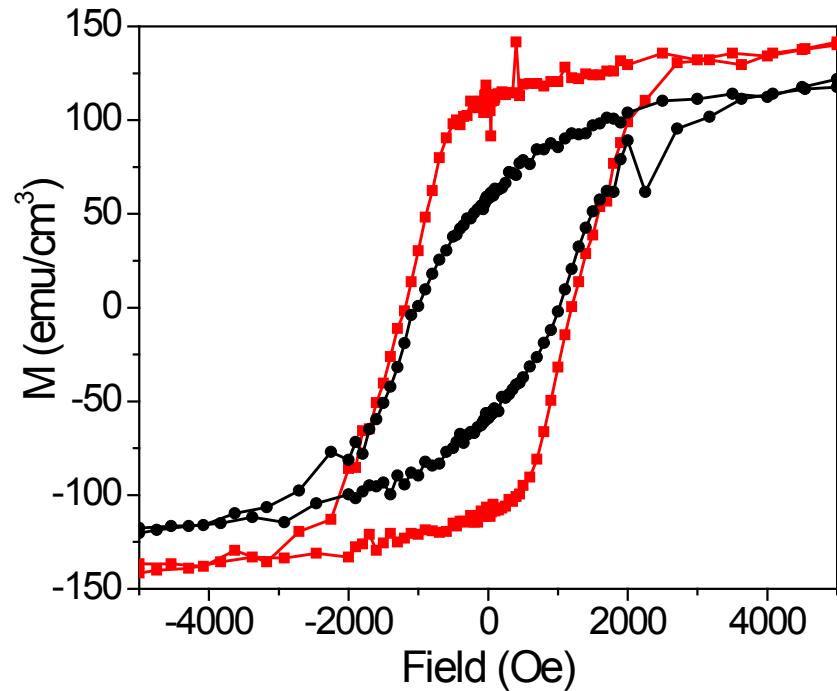
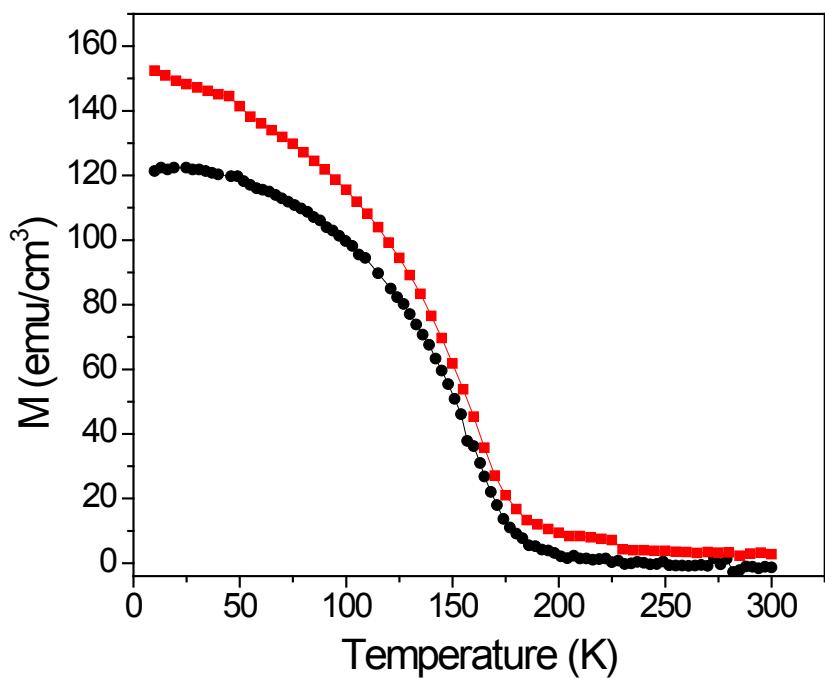
Fit to Valet-Fert theory



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!!

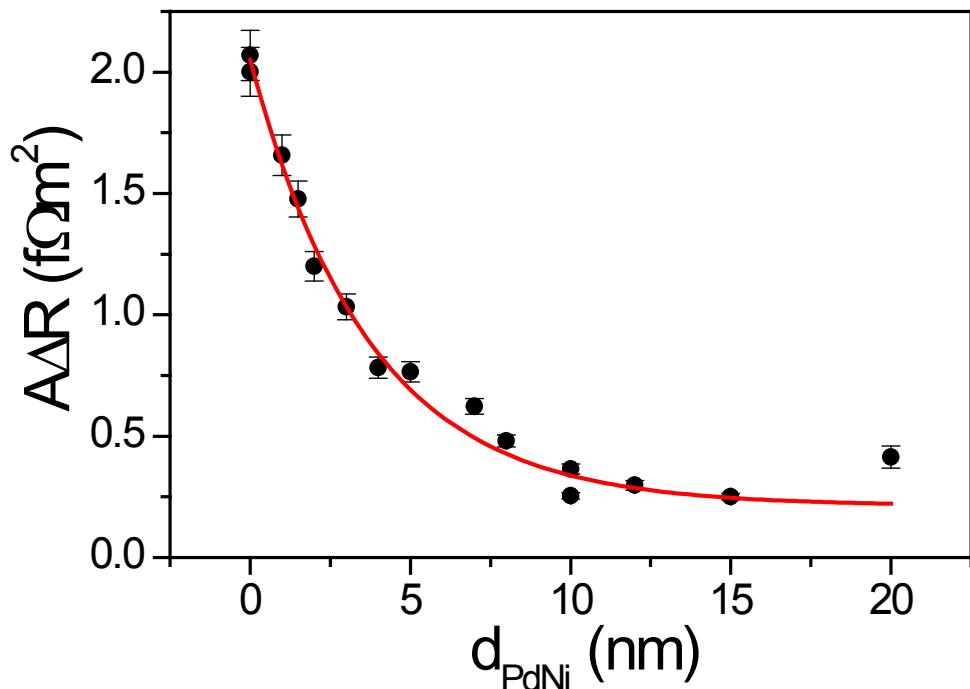
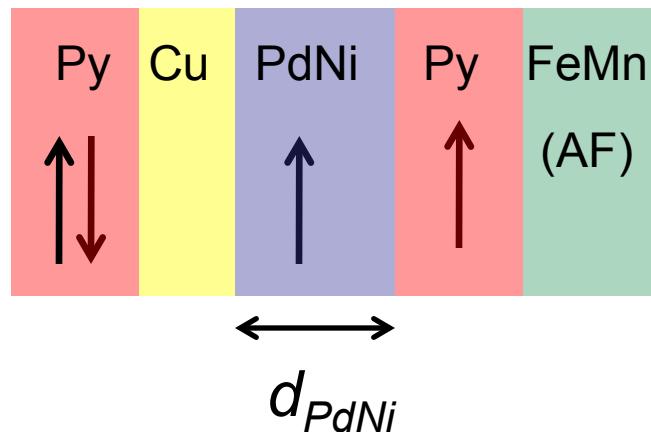
(violates assumptions about P and AP states in GMR experiment)

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

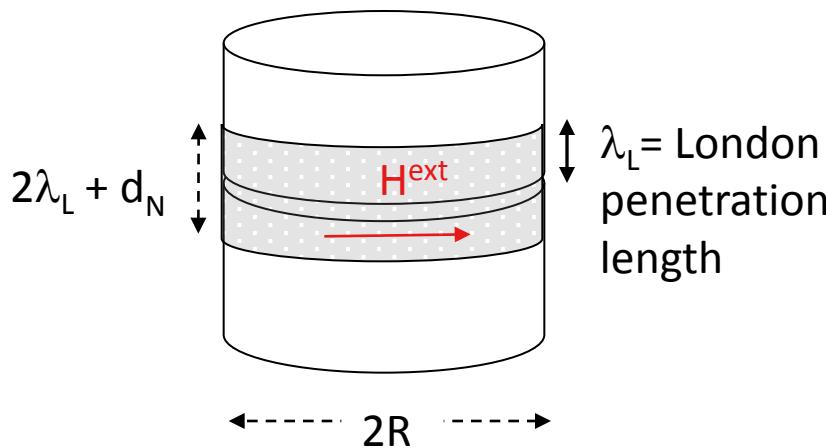


Try a different approach:

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

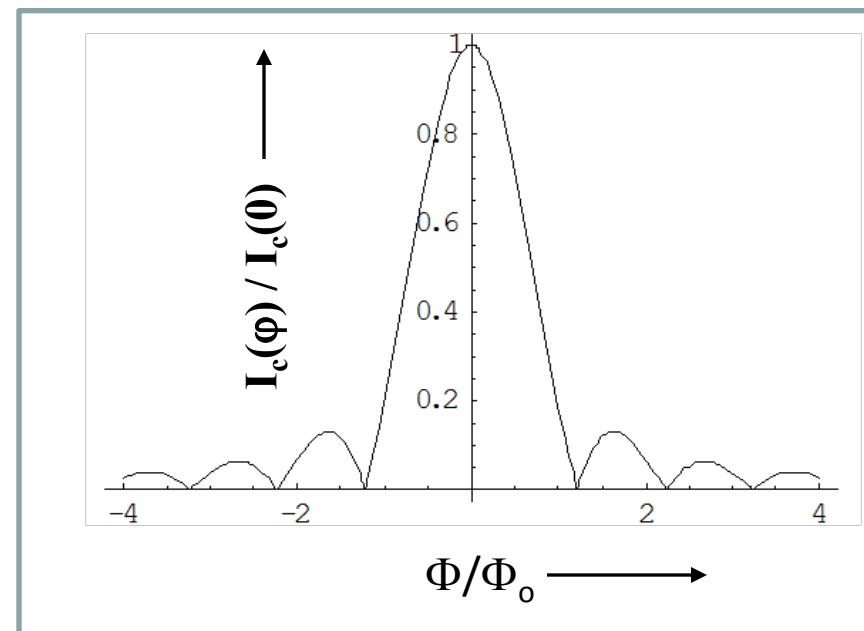
Aside: Characterization of Josephson Junctions by the Fraunhofer pattern

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



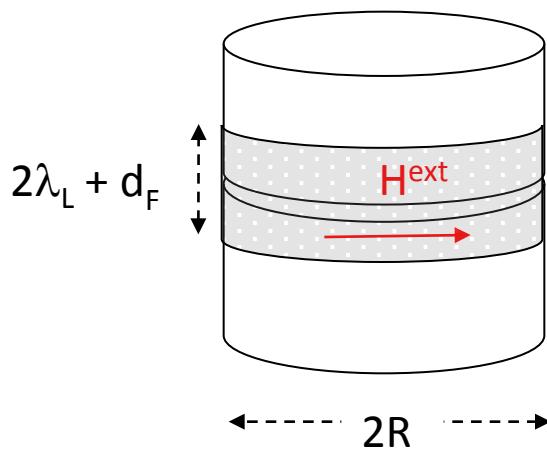
$$I_C(\Phi) = I_C(0) \frac{2 \times J_1\left(\frac{\pi\Phi}{\Phi_o}\right)}{\frac{\pi\Phi}{\Phi_o}}$$

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

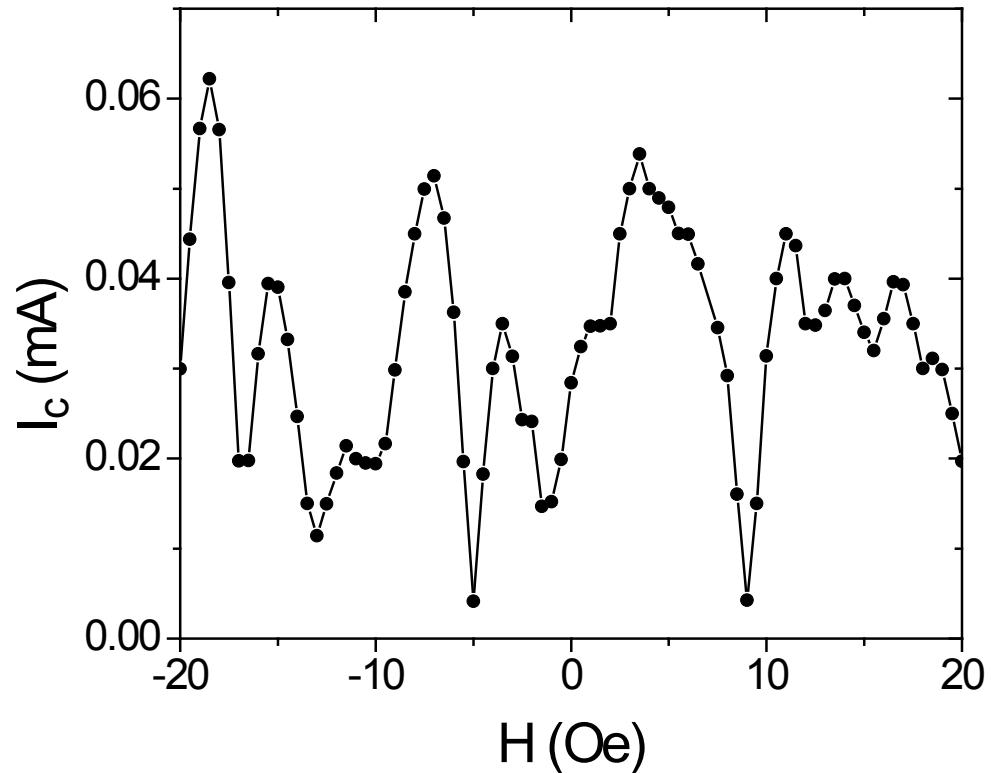


Airy diffraction pattern for a circular junction

Large-area Nb/Co/Nb junctions

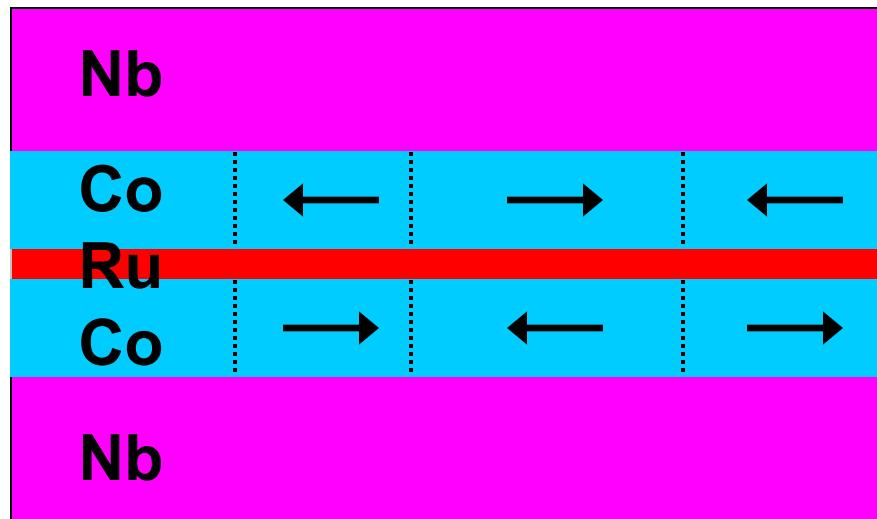


Nb/Co/Nb, $d_{\text{Co}} = 5 \text{ nm}$, $2R = 40 \mu\text{m}$



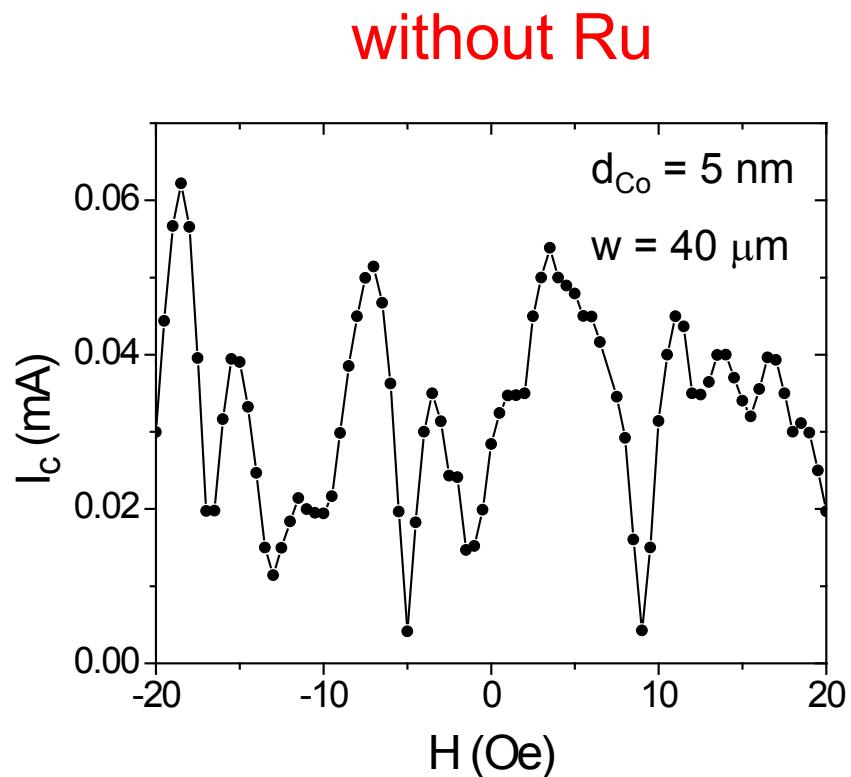
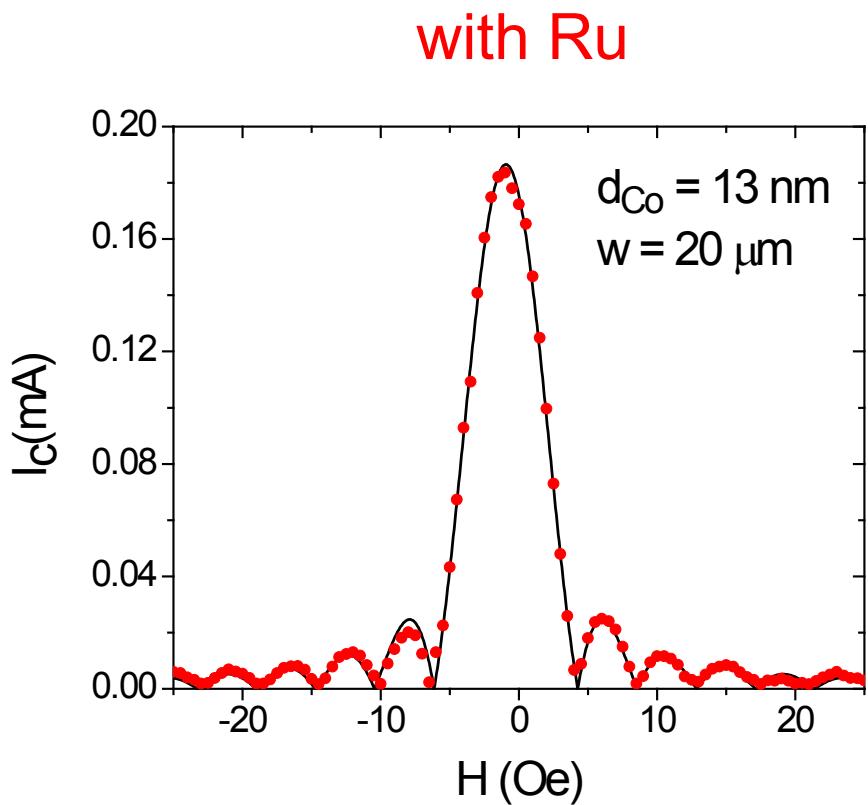
Random Fraunhofer pattern due
to complex domain configuration

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

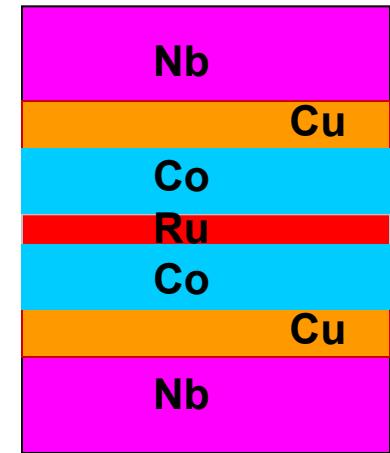
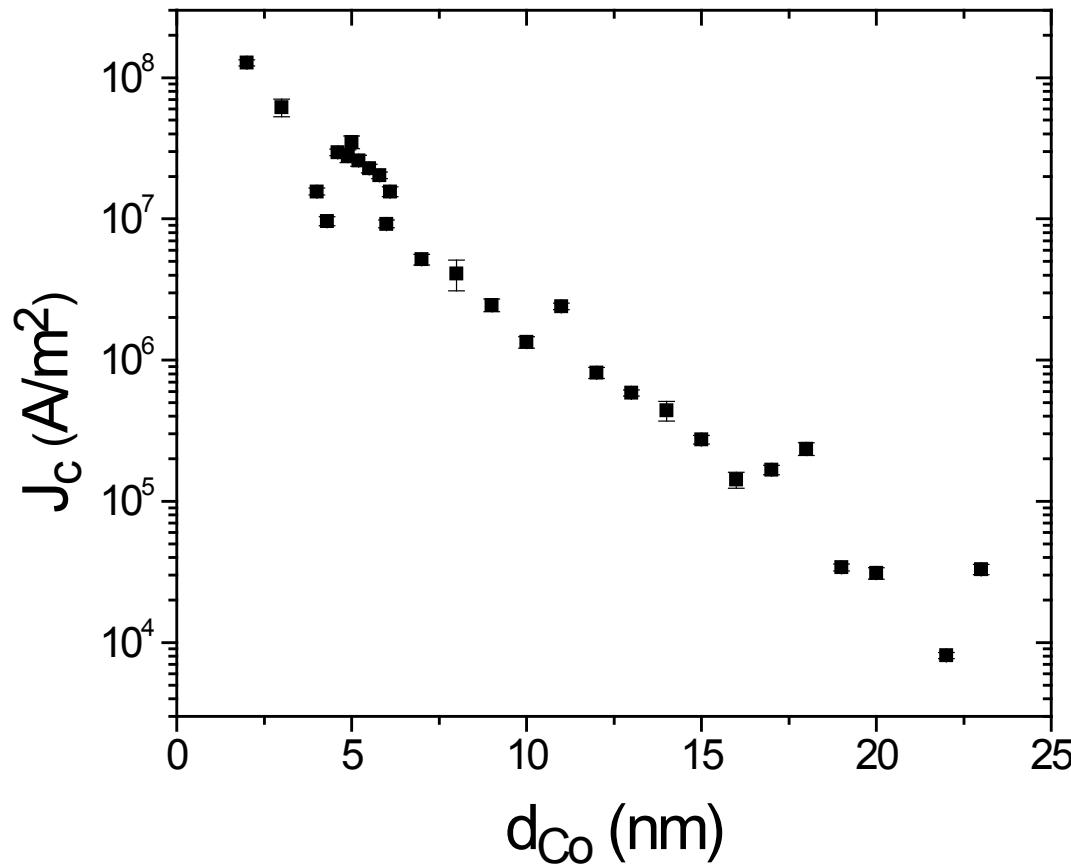


H.A.M. vandenBerg *et al.*, J. Mag. Magn. Mat. 165, 524 (1997).

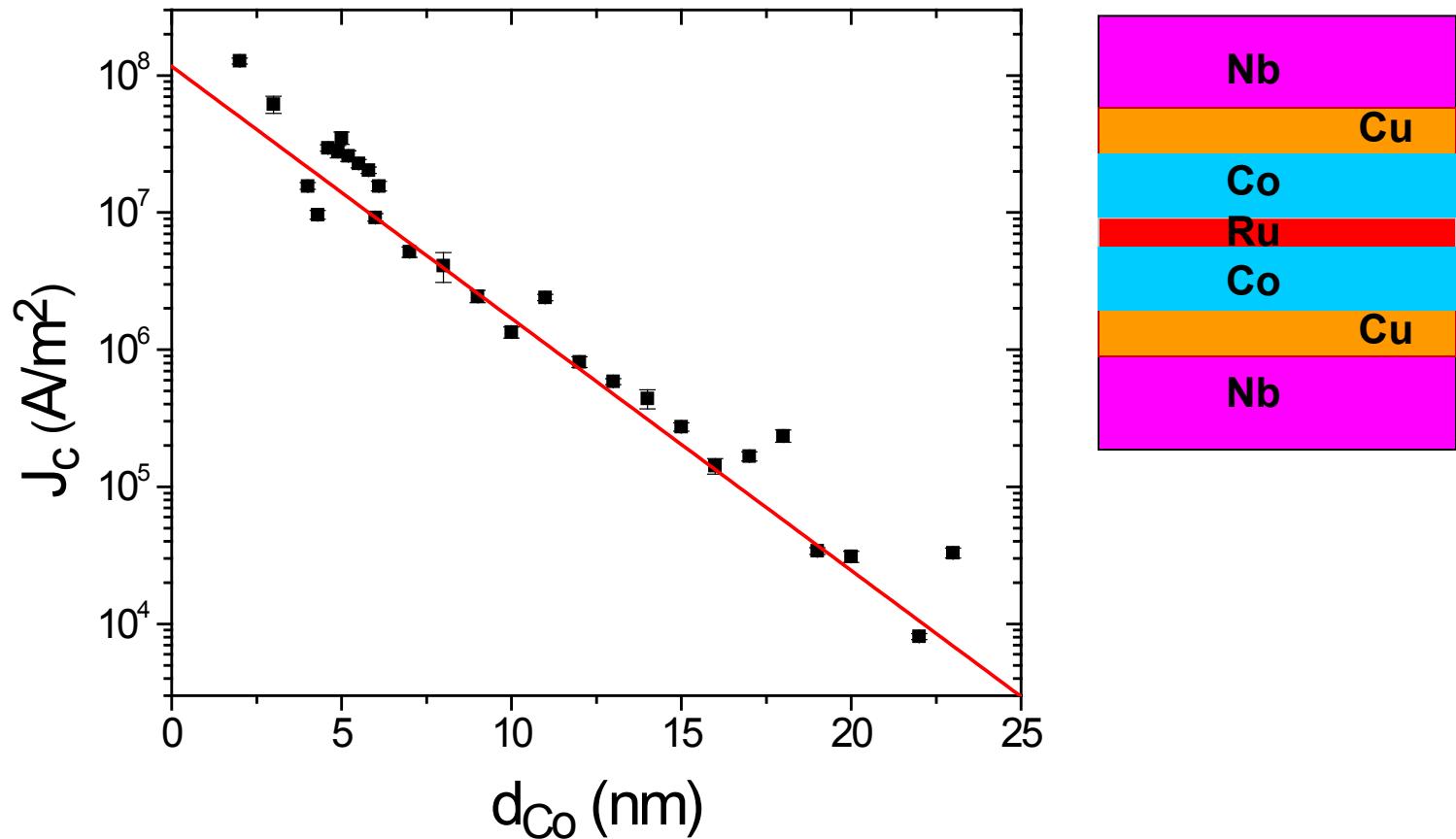
Co/Ru/Co synthetic antiferromagnet restores Fraunhofer pattern!



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



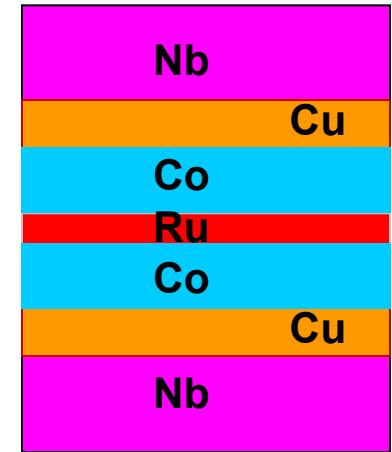
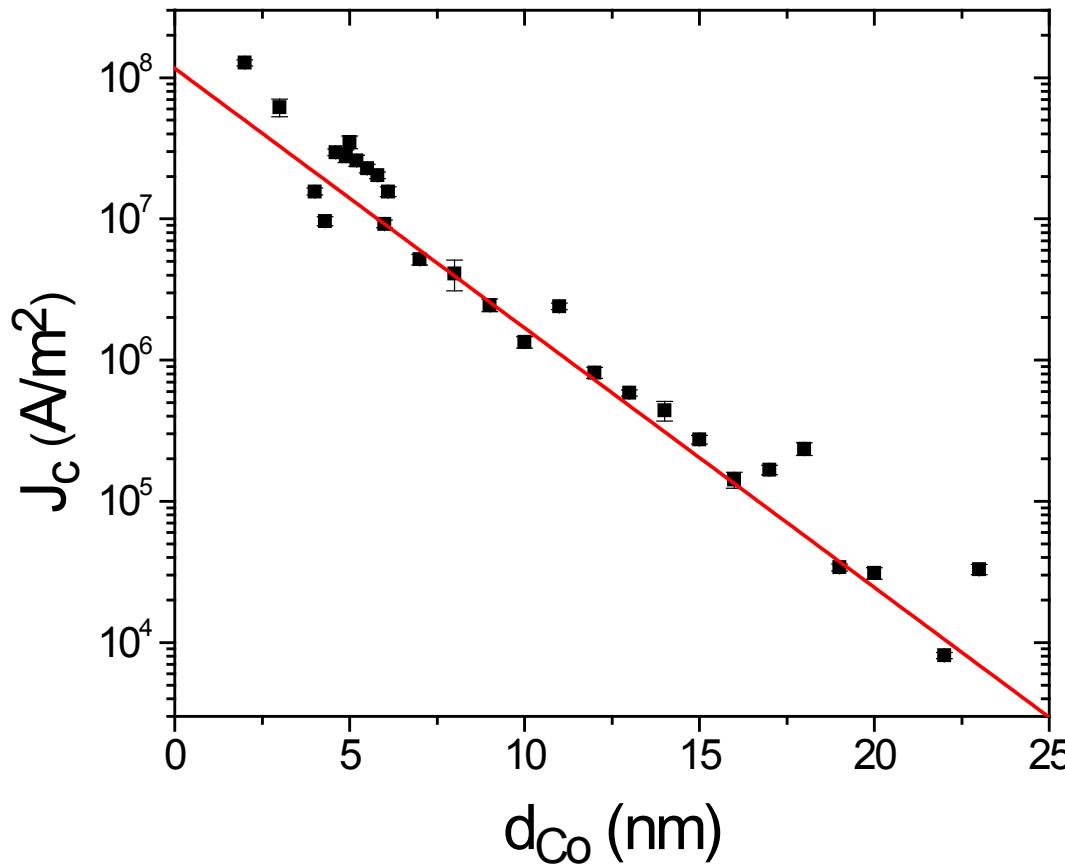
S/F/S junction with a strong F: Co



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



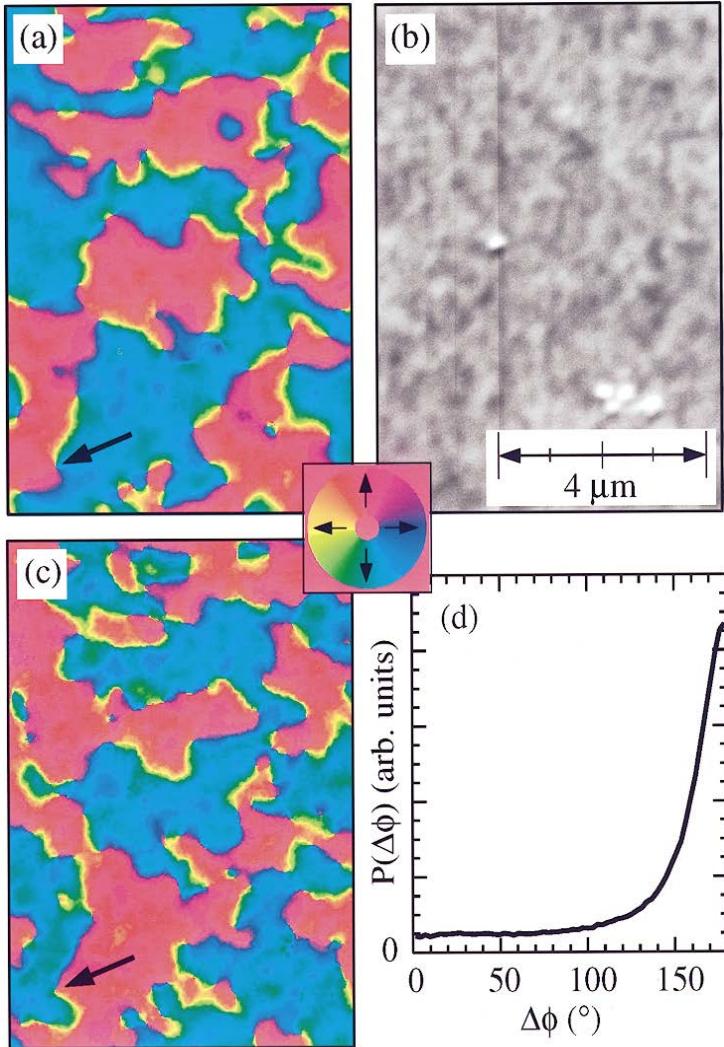
No sign of spin-triplet supercurrent!

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

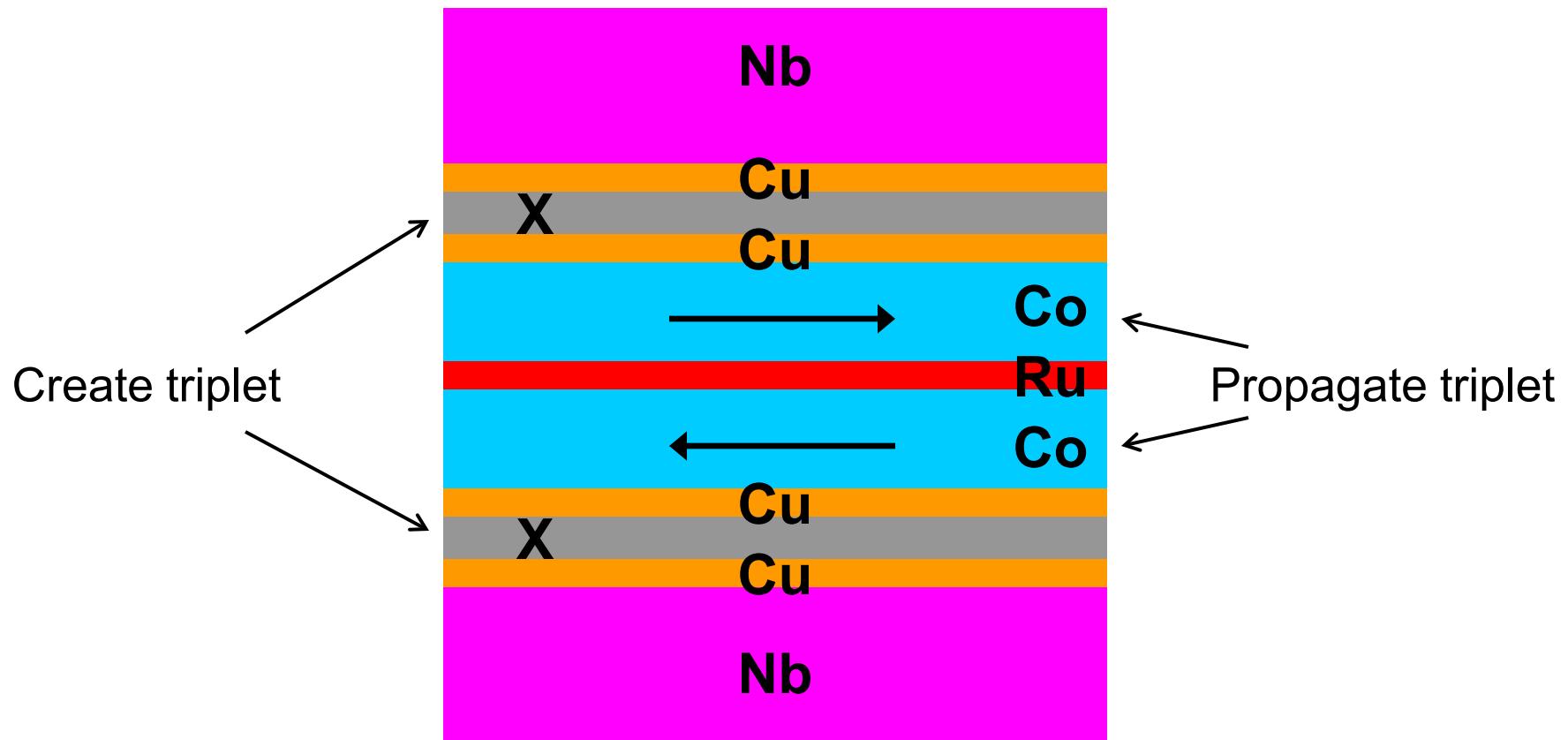
Domains are large ~ 3 μm

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

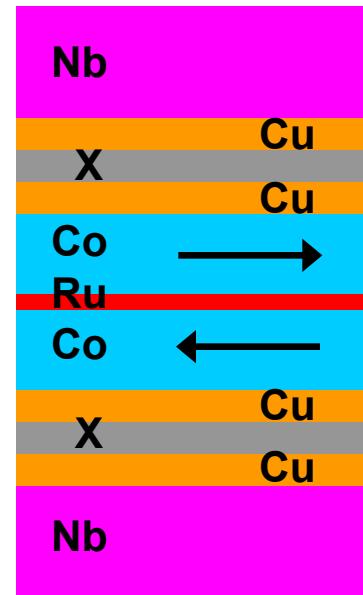
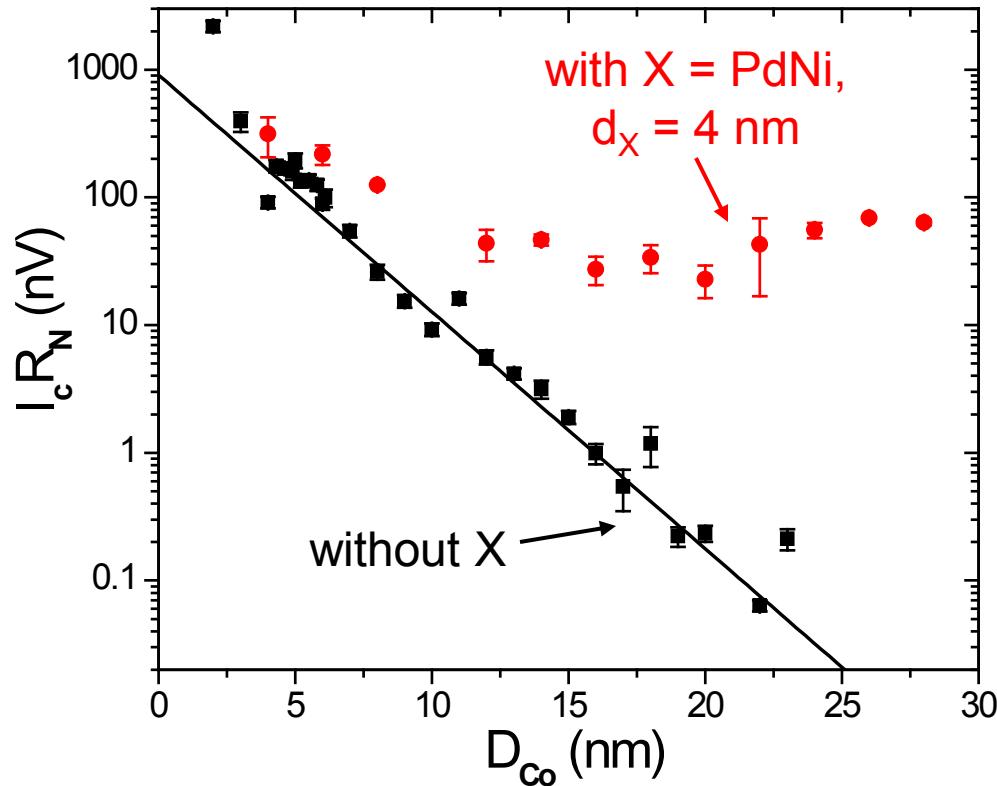
New Idea: combine best of two materials



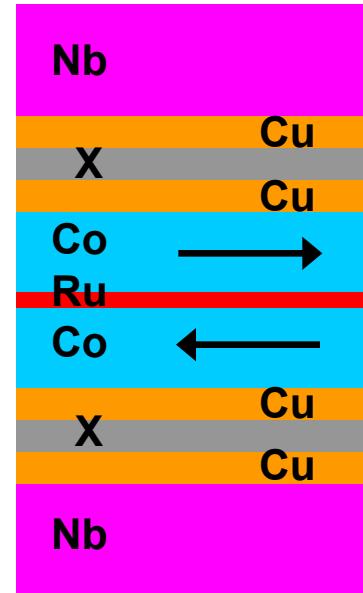
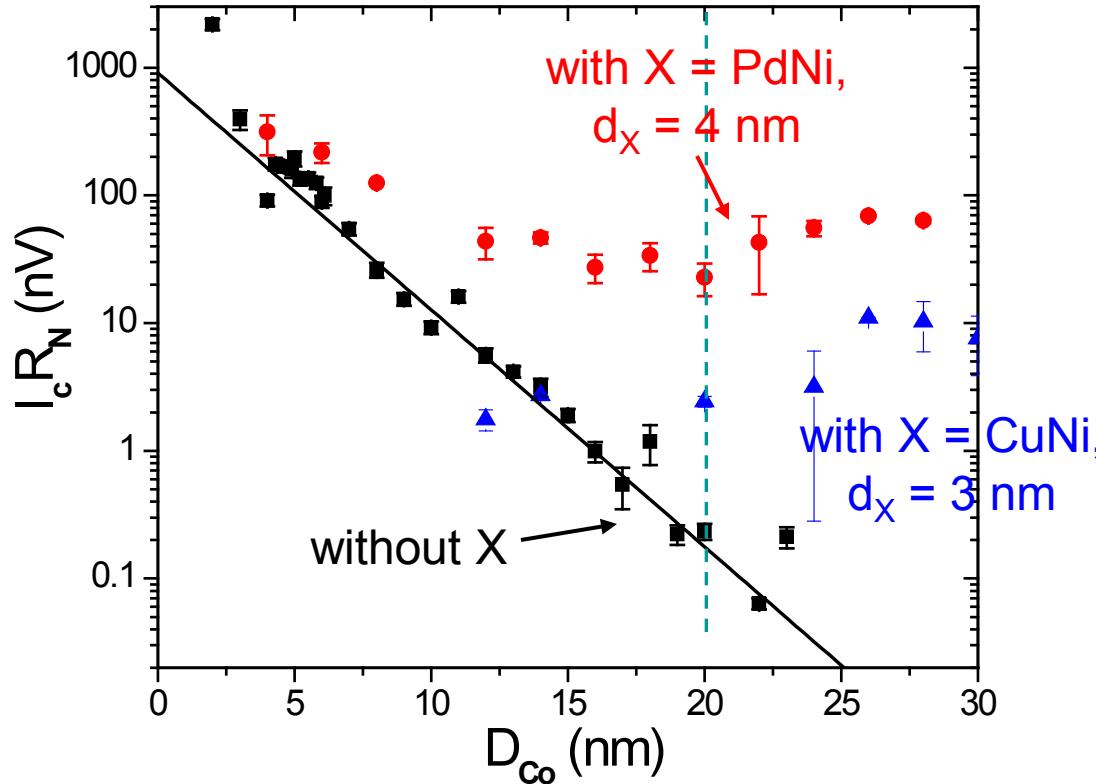
$X = \text{PdNi or CuNi alloy}$

(Cu buffer layers magnetically isolate X from Co)

Finally, the triplet appears!



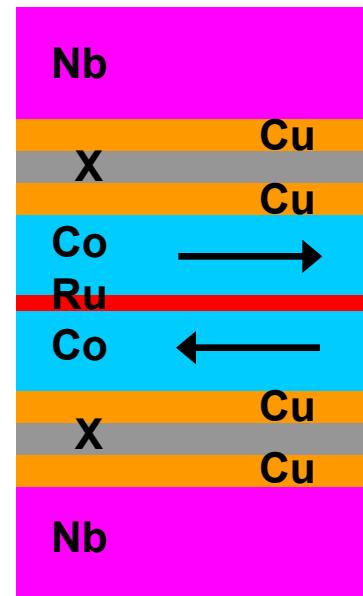
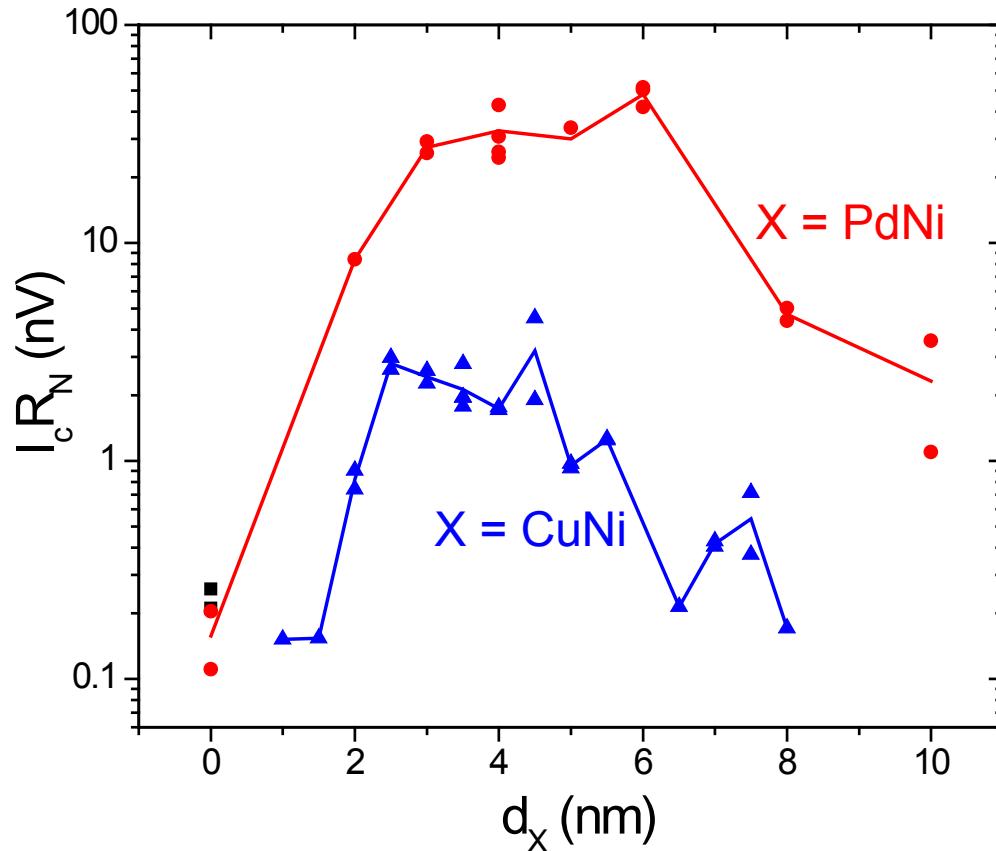
Finally, the triplet appears!



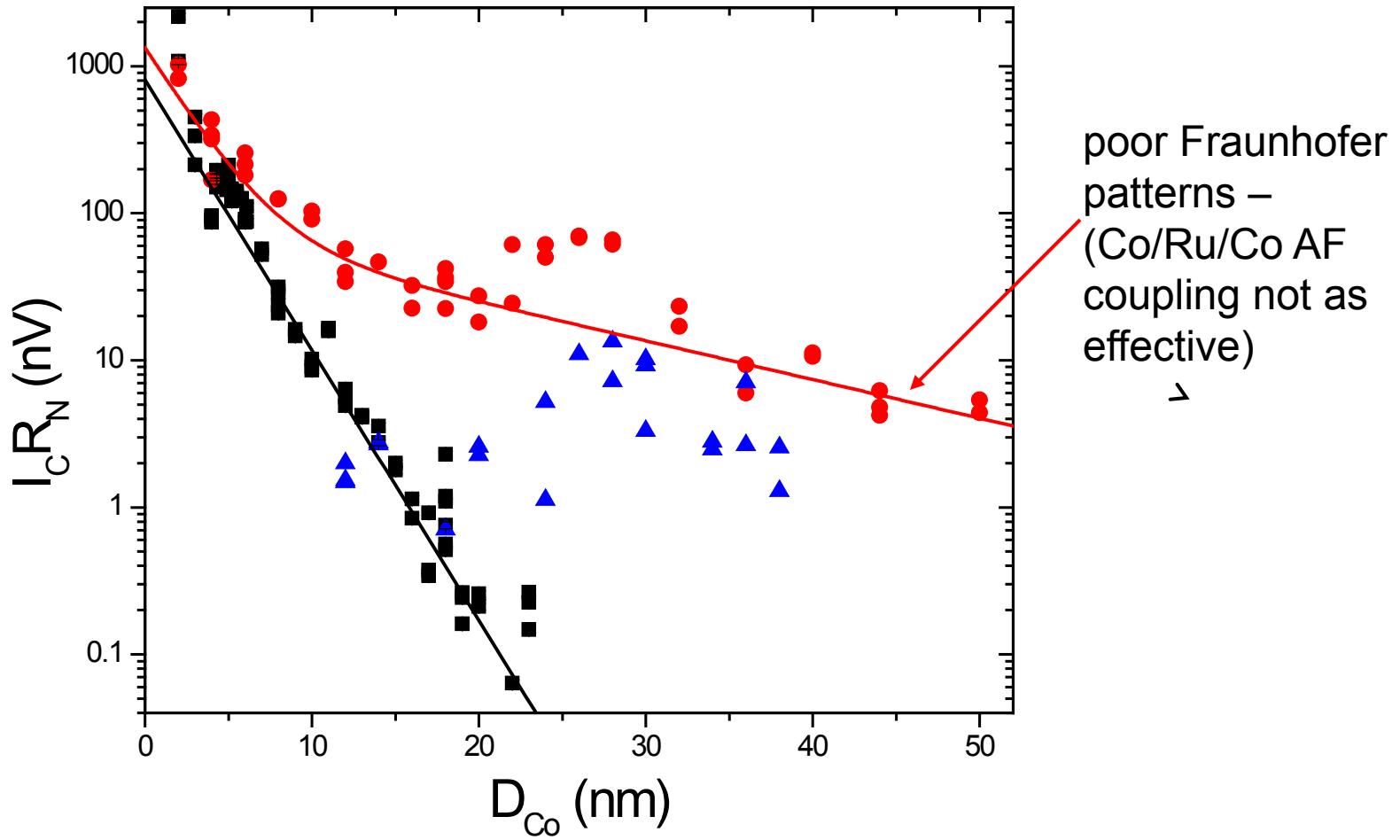
Fix $D_{Co} = 20$ nm and vary d_X

Khaire, Khasawneh, Pratt, & Birge, Phys. Rev. Lett. **104**, 137002 (2010)

Control amplitude of triplet with d_X

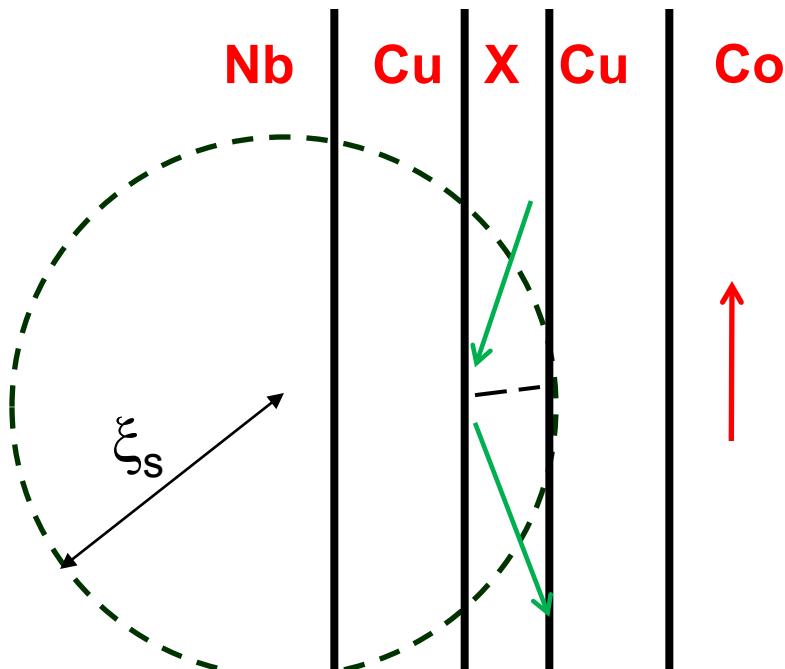


What happens with thicker Co layers?

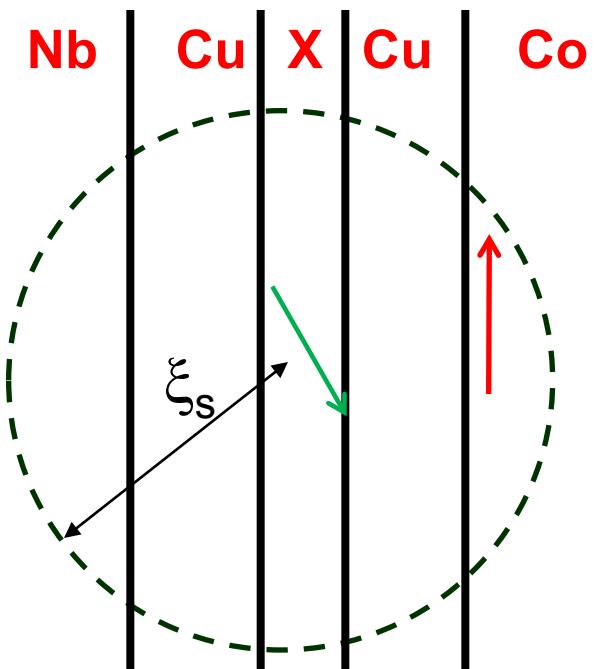


Mechanism for generating triplet

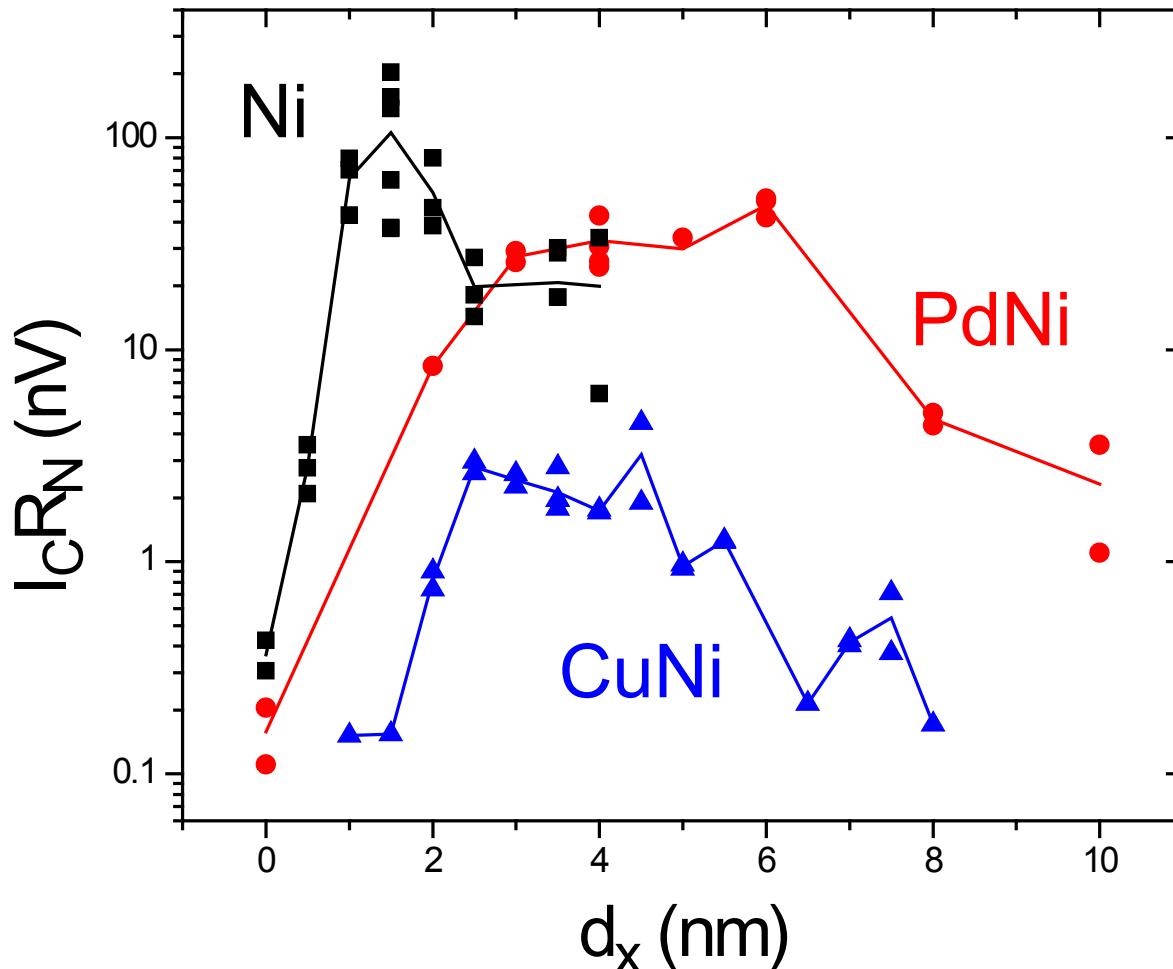
A. Cooper pairs feel non-collinear M between X-layer domains



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

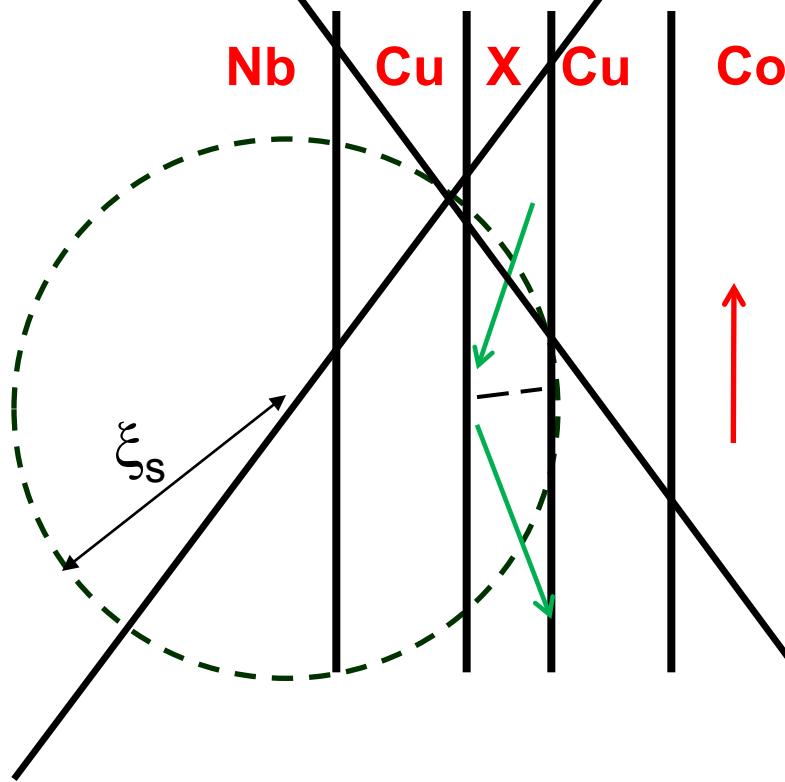


Pure Ni works well for X layer (no need for inhomogeneous X layer)

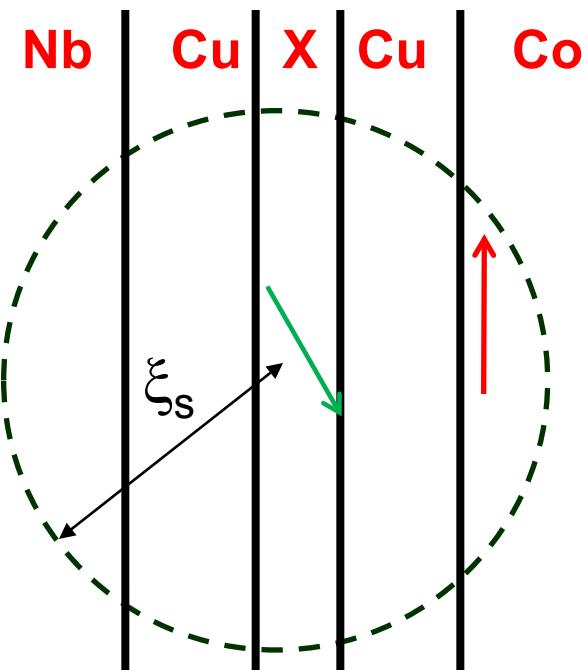


Mechanism for generating triplet

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

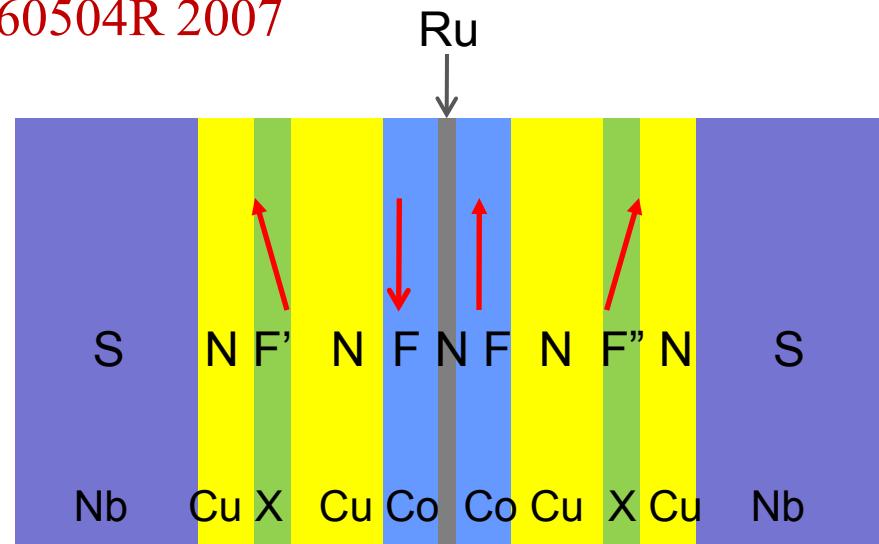
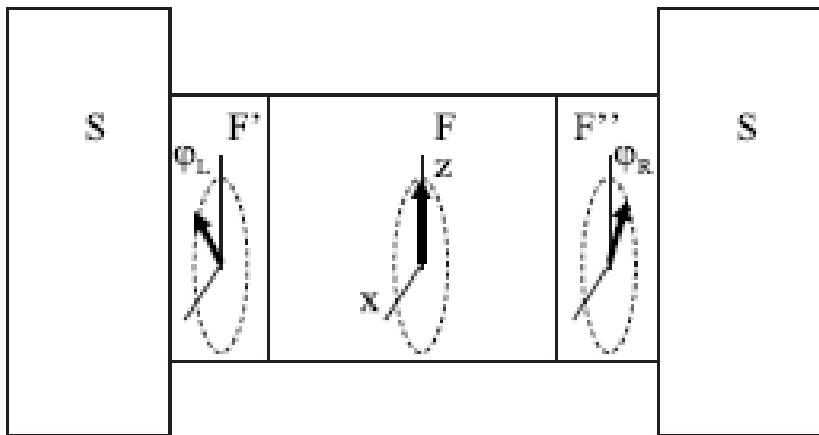


B. Cooper pairs feel non-collinear M between X and Co layers.



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
F₁

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

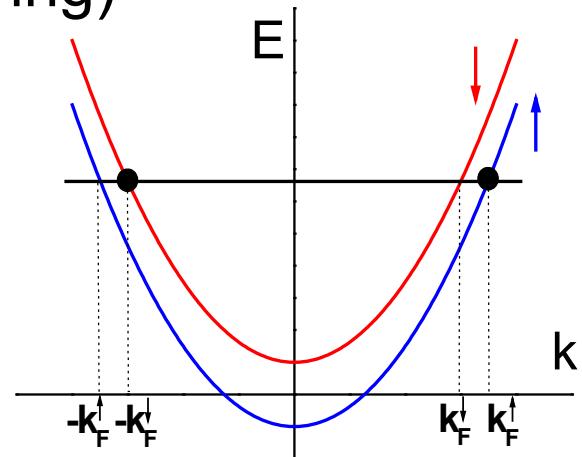
$$Q = k_F^\uparrow - k_F^\downarrow$$

S \uparrow
F₁ \nearrow
F₂

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

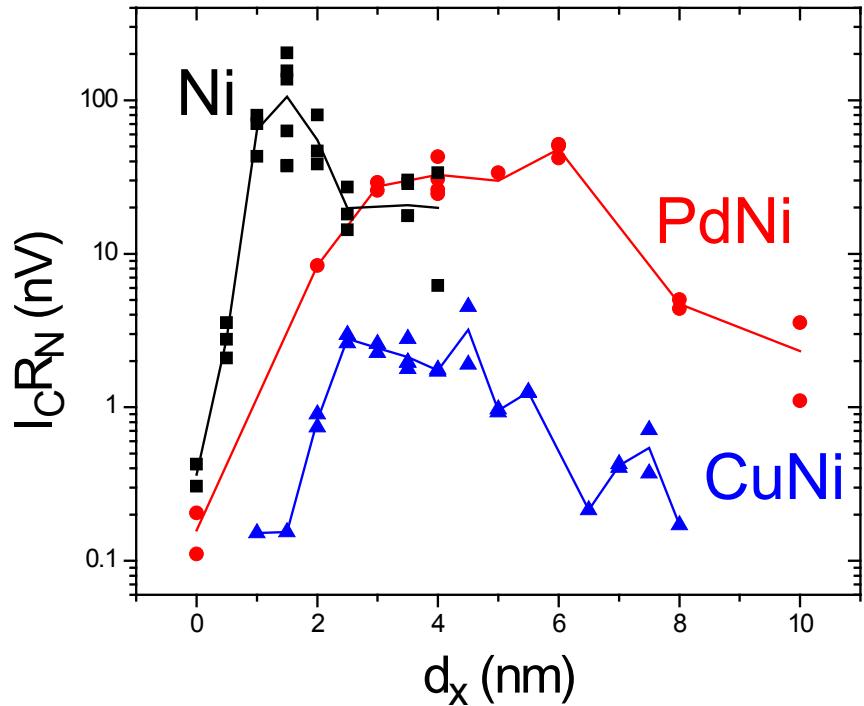
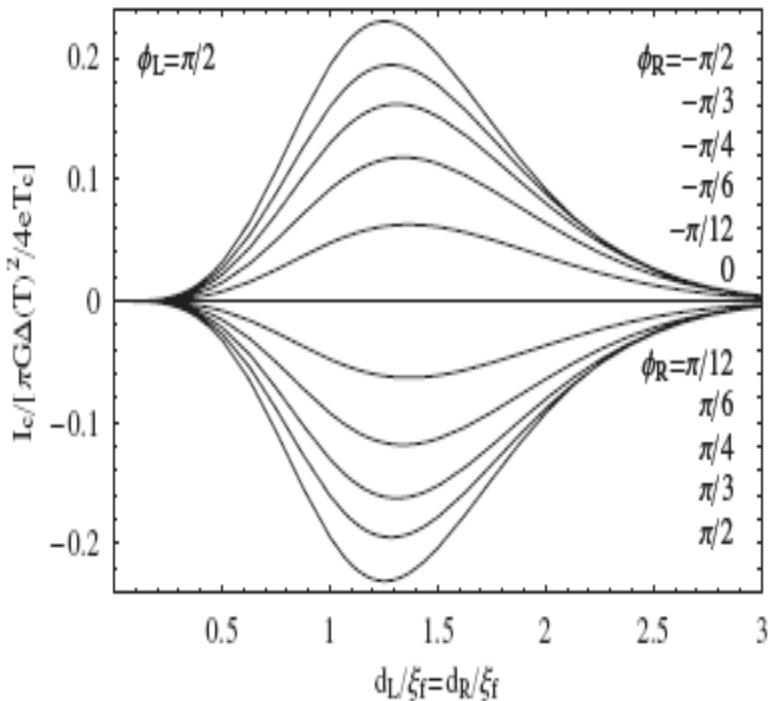
short-range triplet component

long-range triplet components



Optimization of triplet generation

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

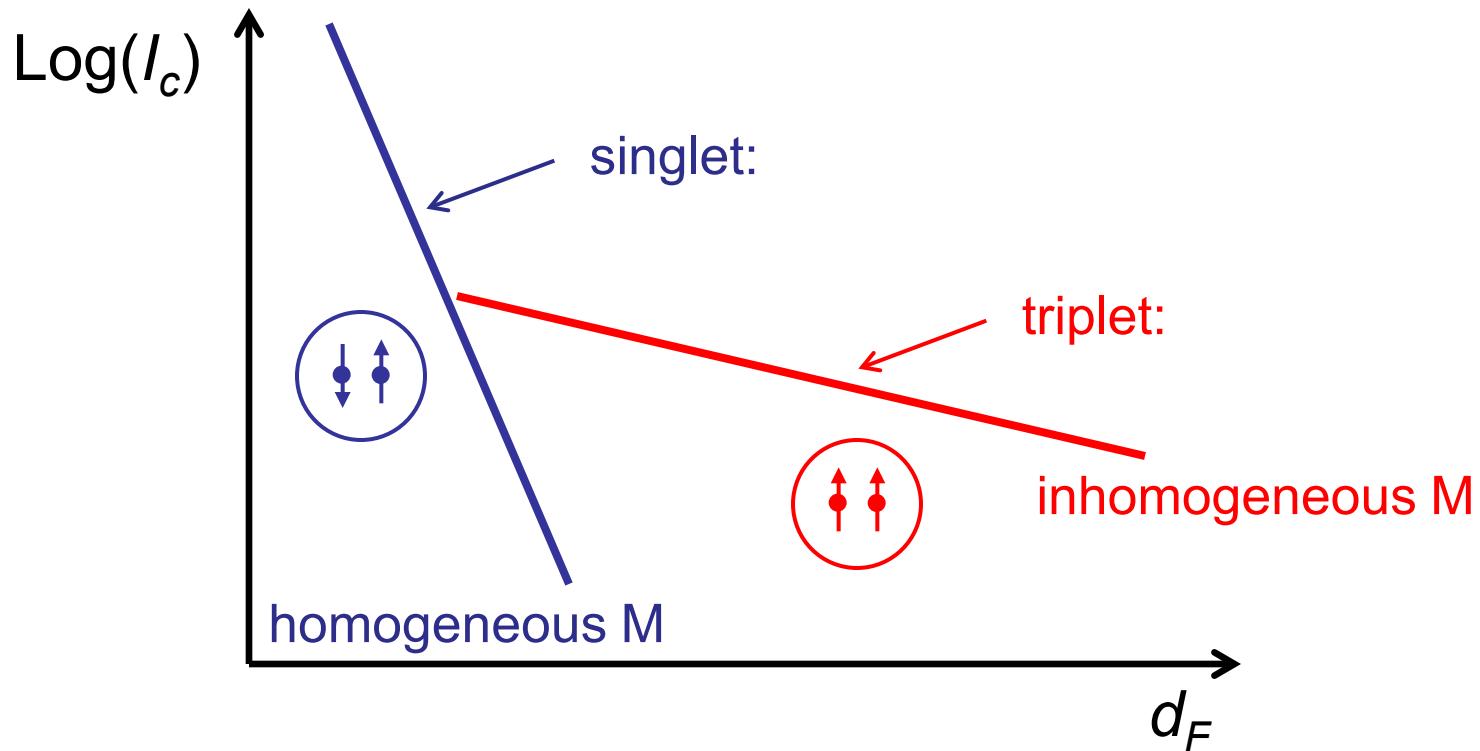


Triplet contribution to the critical current
is observed only for $d_x \approx (0.5–2.5)\xi_F$

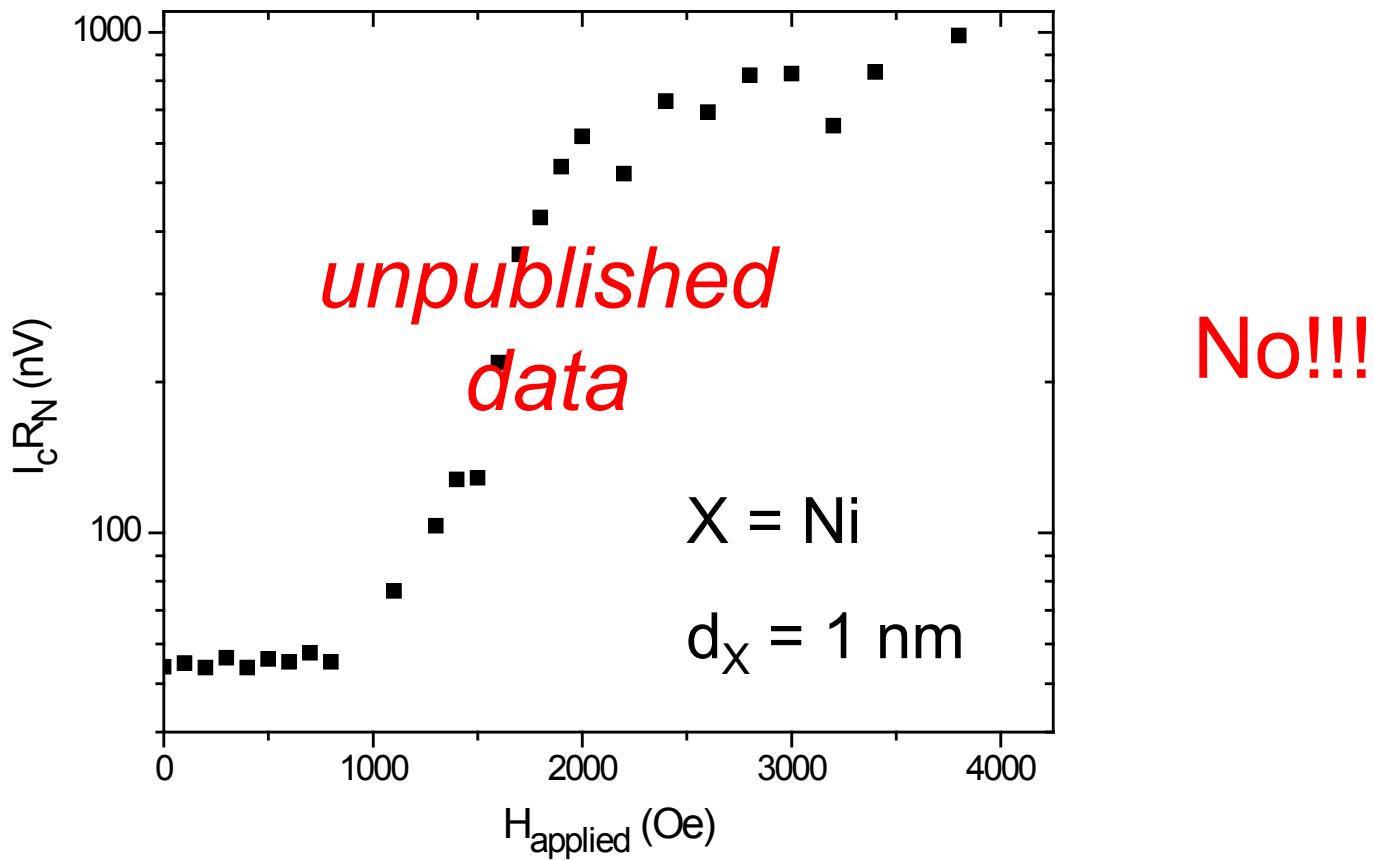
$$E_{ex}^{Ni} > E_{ex}^{PdNi} > E_{ex}^{CuNi}$$

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

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

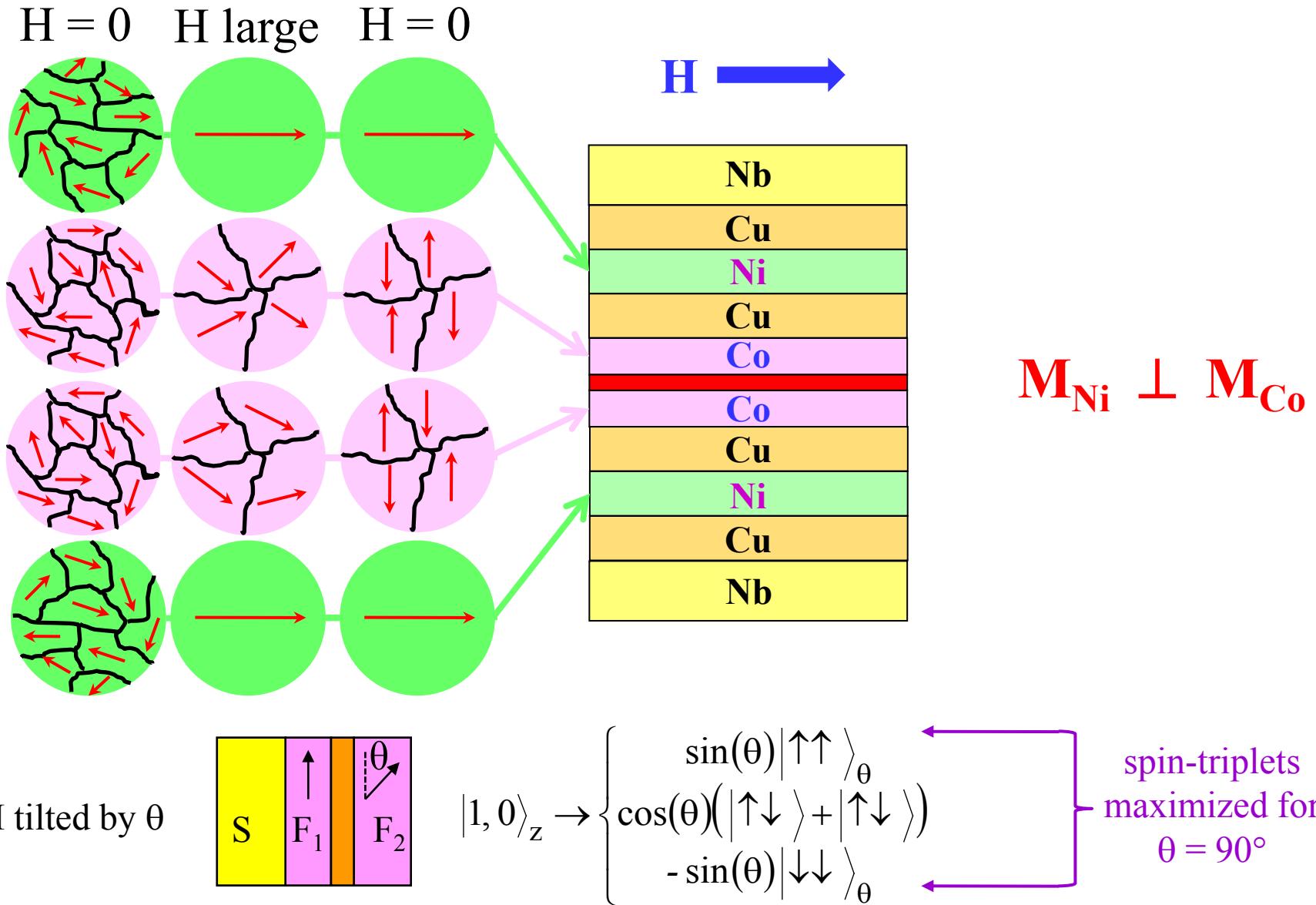


Does triplet disappear after we magnetize the samples?



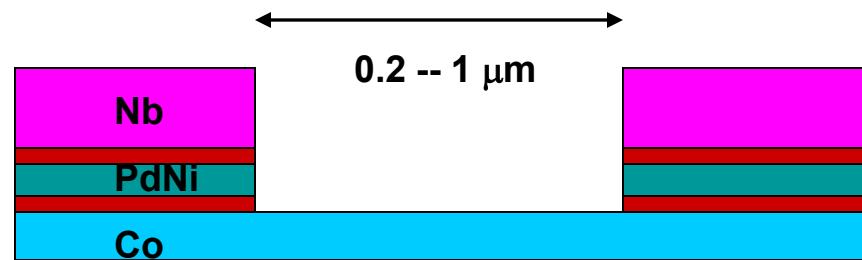
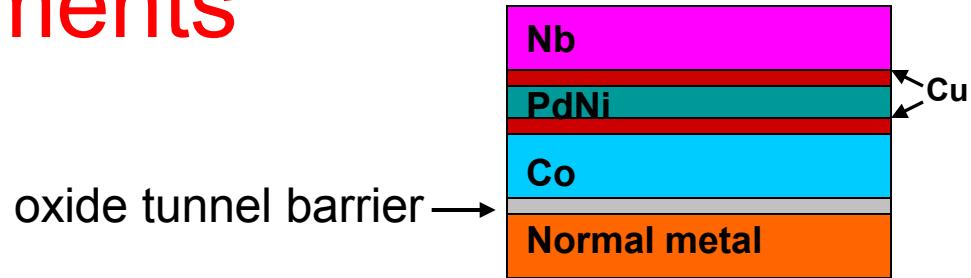
Why does I_c increase?

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



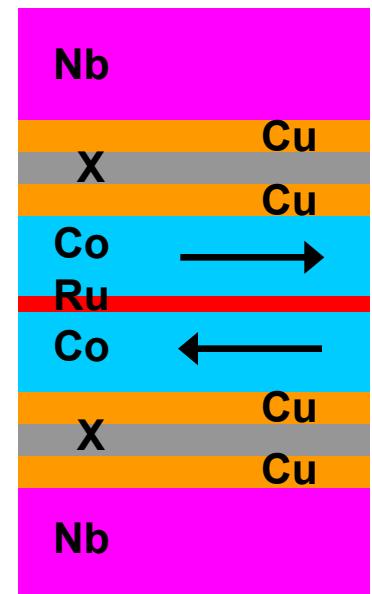
Future Experiments

- 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).