Spin-triplet supercurrent and controllable phase states in Josephson junctions containing ferromagnetic materials

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in collaboration with Northrop Grumman Corporation

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Outline

• Introduction: Proximity effect in S/N and S/F systems
• Theoretical prediction: spin-triplet pair correlations
• Experimental verification using S/F/S Josephson junctions
• Amplitude and phase control of spin-triplet supercurrent
• Work toward a superconducting magnetic memory
  • Phase control in a simpler system
• Summary and Future Prospects
Proximity effect: “leakage of Cooper pairs” from S to N or F

Electron wavefunctions dephase over time

\[ \tau \approx \frac{\hbar}{2\varepsilon} \]

Average over thermal distribution:

ballistic

\[ \xi_N = v_F \tau = \frac{\hbar v_F}{2\pi k_B T} \]

diffusive

\[ \xi_N = \sqrt{D_N \tau} = \sqrt{\frac{\hbar D_N}{2\pi k_B T}} \]

Ballistic

\[ \xi_F = Q^{-1} = \frac{\hbar v_F}{2E_{ex}} \]

diffusive

\[ \xi_F = \frac{\hbar D_F}{E_{ex}} \]

Proximity effect:

- S (Superconductor)
- N (Normal metal)
- F (Ferromagnet)

Electron wavefunction dephases over time due to scattering. The distance an electron can travel before dephasing is given by the mean free path, \( \xi \). In the ballistic regime, \( \xi \) is given by the Fermi velocity times the mean free path, \( \xi_F = Q^{-1} \), where \( Q = 2E_{ex}/\hbar v_F \). In the diffusive regime, \( \xi \) is given by the diffusion constant times the mean free path, \( \xi_N = \sqrt{D_N \tau} \), where \( D_N = \hbar D_N / (2\pi k_B T) \).
Proximity effect: S/N vs. S/F

\[ \xi_N = \sqrt{\frac{\hbar D_N}{2\pi k_B T}} \approx 0.1 - 1\mu m \]

\[ \Psi(x) = \Psi_0 \exp\left(-x / \xi_N\right) \]

\[ \xi_F \sim \sqrt{\frac{\hbar D_F}{E_{ex}}} \approx few nm \]

\[ \Psi(x) = \Psi_0 \cos\left(x / \xi_F\right) \exp\left(-x / \xi_F\right) \]
How to detect the oscillating pair correlation function?

1. Measure $T_c$ of S/F bilayers as a function of $d_F$

Jiang, Davidovic, Reich, Chien, PRL 74, 314 (1995).

Problem: interpretation controversial due to magnetic dead layers.
How to detect the oscillating pair correlation function?

2. 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?

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

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

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

Weak F: Cu$_{48}$Ni$_{52}$ alloy, vary $T$


Weak F: Pd$_{88}$Ni$_{12}$, vary $d_F$

Kontos, Aprili ... 89, 137007 (2002)
How to detect the oscillating pair correlation function?

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

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

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

More Cu$_{48}$Ni$_{52}$ alloy

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Robinson, Piano, Burnell, Bell, Blamire, PRL 97, 177003 (2005)
2001 Prediction: *spin-triplet* pair correlations

i) are long-ranged in $F$

ii) can be induced by noncollinear magnetization


S/F/S Josephson junction can carry spin-triplet supercurrent

Bergeret, Volkov & Efetov, PRL 86, 4096 (2001)
Houzet & Buzdin, PRB 76, 060504(R) (2007):

Non-collinear magnetizations convert pairs from spin-singlet to spin-triplet

Signature of spin-triplet supercurrent:

\[
\log(I_c) = \frac{\hbar D_F}{E_{ex}}
\]

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

\[
\xi_F^T = \sqrt{\frac{\hbar D_F}{2\pi k_B T}}
\]
Sample Fabrication

1. Sputter S/F/S
2. Pattern pillars with photo or e-beam lithography

Image reversal photoresist

Au

Nb

F

Nb bottom contact

Si Substrate

0.3 – 40 µm
Sample Fabrication

1. Sputter S/F/S
2. Pattern pillars with photo or e-beam lithography
3. Ion mill
4. Deposit SiOx

Sample Fabrication

Si O\textsubscript{x}

Au

F

 Nb

Nb bottom contact

Si Substrate

0.3 – 40 \textmu m
Sample Fabrication

1. Sputter S/F/S
2. Pattern pillars with photo or e-beam lithography
3. Ion mill
4. Deposit SiOx
5. Liftoff
6. Deposit top Nb contact

Sample Fabrication Diagram:

- SiOx
- Nb top contact
- Nb bottom contact
- Au
- F
- Si Substrate

0.3 – 40 µm
Low sample resistance: 100 $\mu\Omega$ – 10 m$\Omega$

→ Measure with SQUID-based potentiometer

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

I-V characteristic of overdamped Josephson junction

$I_c^-$ $I_c^+$

supercurrent

$I_c \equiv$ critical current
Problem: Large-area S/F/S junctions with strong ferromagnets ⇒ distorted Fraunhofer patterns

\[ 2\lambda_L + d_F \]

\[ H_{\text{ext}} \]

\[ 2R \]

\[ d_{\text{Co}} = 5 \text{ nm}, \ 2R = 40 \mu\text{m} \]

Random Fraunhofer pattern due to complex domain configuration
Solution: Co/Ru/Co synthetic antiferromagnet cancels flux and restores Fraunhofer pattern
How to generate spin-triplet supercurrent

\[ I_{cN} (\text{nV}) \]

\[ D_{\text{Co}} (\text{nm}) \]

Fix \( D_{\text{Co}} = 20 \text{ nm} \) and vary \( d_{F'} \).

Control amplitude of triplet with $d_{F'}$, $d_{F''}$

Microscopic mechanism for triplet generation
(M. Eschrig)

\[ |0,0\rangle = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle) \]

\[ |\psi\rangle = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle e^{iQx} - |\downarrow\uparrow\rangle e^{-iQx}) \]

\[ = \frac{1}{\sqrt{2}} \left[ (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle) \cos(Qx) + i (|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle) \sin(Qx) \right] \]

\[ = |0,0\rangle_z \cos(Qx) + |1,0\rangle_z \sin(Qx) \]

rotate basis:

\[ |1,0\rangle_z = \frac{\sin(\theta)}{\sqrt{2}} |1,1\rangle_\theta + \cos(\theta) |1,0\rangle_\theta - \frac{\sin(\theta)}{\sqrt{2}} |1,-1\rangle_\theta \]

\[ |1,1\rangle_\theta = |\uparrow\uparrow\rangle_\theta \]

\[ |1,-1\rangle_\theta = |\downarrow\downarrow\rangle_\theta \]
Triplet contribution to the critical current is observed only for $d_x \approx (0.5–2.5)\xi_F$

increase $\sim \sin^2(Qx)$; decrease $\sim e^{-x/\xi_F}$

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

$\xi_F^{Ni} < \xi_F^{PdNi} < \xi_F^{CuNi}$
Earlier observation of triplet:


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

Reproduced in 2010 by J. Aarts:

Spin-triplet supercurrents observed by others in 2010


2. Sprungmann et al., PRB 82, 060505 (2010)

Heusler alloy Cu$_2$MnAl
Spin-triplet supercurrents observed by others in 2010

3. Anwar et al., PRB 82, 100501 (2010)

First step toward control of supercurrent: magnetize the samples
(Expect $I_c$ to drop by reducing non-collinear magnetization?)
$I_c$ increases!

Why does $I_c$ increase?

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

Spin-flop transition optimizes 90° angle between $\mathbf{M}_{\text{Co}}$ and $\mathbf{M}_{\text{Ni}}$ and aligns Ni magnetizations.

Magnetic configuration confirmed at NIST by SEMPA (McMorran & Unguris) and PNR (Ginley & Borchers); see Klose et al., Phys.Rev. Lett. 108, 127002 (2012)
Other probes of spin-triplet correlations: $T_c$ of S/F/F’ trilayers

Next step: Control spin-triplet supercurrent by controlling magnetic states

**On/Off**

- "Off" (Singlet only)
- "On" Triplet

- Rotate \( \mathbf{M} \) by 90°
- Requires only one junction
- Need two external field coils

**Phase Change**

- 0
- \( \pi \)

- Rotate \( \mathbf{M} \) by 180°
- Measure with SQUID (interference)
- Need one external field coil
Desire single-domain magnets:
Fabricate submicron S/F/S Josephson junctions

- ma-N 2401 negative e-beam resist
- Junctions patterned by Ar ion milling

Diagram:
- Nb
- Cu
- F
- Cu

Side view:
- $H_{app}$
- 0.50 $\mu$m
- 1.26 $\mu$m

Top view:
- $H_{applied}$
- M
Fraunhofer pattern for junction with $F = \text{Ni}_{73}\text{Fe}_{21}\text{Mo}_6$

Airy pattern is shifted in opposite direction of $\mathbf{M}$

F layer is **single-domain** in remnant state

I_c vs NiFeMo thickness

Data fit to:

\[ I_c R_N = V_0 e^{-\frac{d_F}{\xi_{F1}}} \cos \left( \frac{d_F}{\xi_{F2}} - \phi \right) \]

0 - \pi transition occurs
at \( d_{NiFeMo} = 2.25 \) nm

\( I_c \) decays rapidly with
\( d_{NiFeMo} \cdot \xi_{F1} = 0.48 \) nm

Next step: Control spin-triplet supercurrent by controlling magnetic states

On/Off

- "Off" (Singlet only)
- "On" Triplet
- Rotate $\mathbf{M}$ by 90°
- Requires only one junction
- Need two external field coils

Phase Change

- 0
- 0π
- Rotate $\mathbf{M}$ by 180°
- Measure with SQUID (interference)
- Need one external field coil
Cartoon Representation ("Py" = NiFe)

- Orthogonal magnetizations – "High" state

- Non-orthogonal magnetizations – "Low" state

Martinez, Pratt & Birge, PRL 116, 077001 (2016)
Rotate Py Magnetization to turn “off”

Measurement at 0-field

Slowly increase $H_\perp$ to rotate $M_{py}$

Measure at 0-field

Martinez, Pratt & Birge, PRL 116, 077001 (2016)
Rotate Py back to turn “on”

Slowly increase $H_{||}$ to rotate $M_{py}$ back again

Measure at 0-field

Martinez, Pratt & Birge, PRL 116, 077001 (2016)
Turn on and off repeatedly
Next step: Control spin-triplet supercurrent by controlling magnetic states

On/Off

“Off” (Singlet only)

“On” Triplet

- Rotate $\mathbf{M}$ by 90°
- Requires only one junction
- Need two external field coils

Phase Change

0

\[ \pi \]

- Rotate $\mathbf{M}$ by 180°
- Measure with SQUID (interference)
- Need one external field coil
Controllable $0-\pi$ switching with spin-triplet supercurrent

[Co/Pd] multilayers have strong perpendicular magnetic anisotropy.

Central Ru layer creates a synthetic antiferromagnet, to reduce stray field at domain walls

Ni(1.6) has fixed in-plane magnetization direction

NiFe(1.25) is in-plane free layer

Glick et al. (in preparation)
SQUID design & measurement protocol

• SQUIDs have one ellipse with aspect ratio = 2 and one hex bit with aspect ratio = 3, both have area = 0.5 μm².

• Measurement protocol:
  • Initialize bits at -1500 Oe, then remove trapped flux
  • Apply set fields $H_{set} = +5$ Oe, $+10$ Oe, $+15$ Oe, etc. until magnetic state switches
  • After each set field, measure $I_c(I_{flux})$
  • Apply set fields $H_{set} = -5$ Oe, $-10$ Oe, etc. to return to initial state
  • All measurements are performed at $H_{set} = 0$.
  • Keep $|H_{set}|$ small enough so that only the elliptical bit switches (minor loop).
Controllable $0-\pi$ switching with spin-triplet supercurrent

Data will be re-inserted after publication (Glick et al. in preparation)
This work is not just of academic interest!
Energy-Efficient Superconducting Computing—Power Budgets and Requirements

D. Scott Holmes, Senior Member, IEEE, Andrew L. Ripple, and Marc A. Manheimer
A superconducting computer needs memory…

Josephson Magnetic Random Access Memory (JMRAM)
Northrop Grumman Corporation

Memory cell is SQUID loop

One junction has two stable states for “0” and “1”

Magnetic states are written using standard MRAM techniques
Recall: $I_c$ of S/F/S Josephson junction oscillates with $d_F$

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

Weak F: Cu$_{48}$Ni$_{52}$ alloy

Strong F: Co

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

Robinson, Piano, Burnell, Bell, Blamire, PRL 97, 177003 (2005)
JMRAM memory cell junction

Spin-valve memory element

S/F/S: $I_c$ vs $d_F$

$S/F_1/N/F_2/S$: fixed $d_{F_1}$ & $d_{F_2}$

Control phase rather than amplitude:
JJ acts as passive phase shifter; no need for large $I_cR_N$

Demonstration of $0 - \pi$ switching of $S/F_1/N/F_2/S$ spin-valve Josephson junctions

$F_1$ = fixed layer: Ni (1.2 nm)

$F_2$ = free layer: NiFe (1.5 nm)

Switch magnetization with in-plane field

JJ's with different aspect ratios switch at different applied magnetic fields

On-chip current line couples magnetic flux into SQUIDs
Major loop data show switching of both JJs

Gingrich, Niedzielski, Glick, et al., Nat. Phys. 12, 564 (2016)
Data cuts for the four magnetic states

\[ I_{c^{\text{ave}}} = \frac{(I_{c^+} - I_{c^-})}{2} \]

Gingrich, Niedzielski, Glick, et al., Nat. Phys. 12, 564 (2016)
I_c(Φ) curves have tilted ratchet shape when loop inductances and/or critical currents are asymmetric.

I_c^+(Φ) and I_c^-(Φ) oscillations are asymmetric when L_1 ≠ L_2 & I_{c1} ≠ I_{c2}

I_c^+ and I_c^- shift by equal amounts and in opposite directions along the Φ axis.

Analyze I_c^+ and I_c^- peak shifts to extract JJ phase shifts.
Quantitative fits to SQUID modulation data for the four magnetic states

\[ I_{c^{\pm}} = \frac{I_{c^+} - I_{c^-}}{2} \]

Gingrich, Niedzielski, Glick, et al., Nat. Phys. 12, 564 (2016)
Quantitative Analysis Consistently Assigns the Inductance and Critical Currents of Each State

<table>
<thead>
<tr>
<th>state</th>
<th>$I_{c1}$ (mA)</th>
<th>$I_{c2}$ (mA)</th>
<th>$L_1$ (pH)</th>
<th>$L_2$ (pH)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\pi - \pi$</td>
<td>0.292</td>
<td>0.217</td>
<td>5.73</td>
<td>11.38</td>
</tr>
<tr>
<td>0 - $\pi$</td>
<td>0.565</td>
<td>0.203</td>
<td>5.64</td>
<td>11.33</td>
</tr>
<tr>
<td>0 - 0</td>
<td>0.567</td>
<td>0.419</td>
<td>5.63</td>
<td>11.55</td>
</tr>
<tr>
<td>$\pi - 0$</td>
<td>0.294</td>
<td>0.420</td>
<td>5.71</td>
<td>11.56</td>
</tr>
<tr>
<td>ave</td>
<td>5.68</td>
<td>11.46</td>
<td></td>
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<tr>
<td>$\sigma$</td>
<td>0.05</td>
<td>0.12</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

FastHenry simulations:

$L_1 \approx 7$ pH, $L_2 \approx 13$ pH

Fitting parameters from independent fits of 4 magnetic states are highly consistent

- Exception: critical current of JJ #2 changes slightly in $\pi$ state when JJ #1 switches from $\pi$ to 0 state

Gingrich, Niedzielski, Glick, et al., Nat. Phys. 12, 564 (2016)
What needs to be done

• Memory
  – Optimize performance of magnetic materials
    • Lower $M_{\text{sat}} \Rightarrow$ lower $E_{\text{switch}}$
    • Reduce extrinsic sources of anisotropy in thin films
    • Find better material for fixed layer (Ni has issues)
    • Minimize underlayer roughness
  – Develop read/write electronics & interface to SFQ logic
• Make the rest of the computer!
Summary

• Experimental observation of long-range supercurrent in S/F/S Josephson junctions:
  – requires three F layers
  – can control either supercurrent amplitude or junction ground-state phase by changing the magnetic state

• Magnetic Josephson junctions may be useful for ultra-low-power cryogenic memory
  – phase control achieved with a spin-valve JJ