Cooper pair spin collective phenomena in the $SrRuO_3$ / Sr_2RuO_4 heterostructure

Suk Bum Chung

University of Seoul

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서울시립대학교
UNIVERSITY OF SEOUL

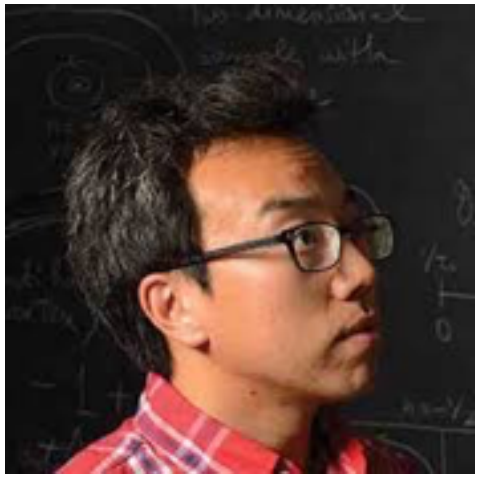
ibs 기초과학연구원
Institute for Basic Science

VERI TAS LUX MEA

Return to India after 30 years!!!

Collaborators

Y. Tserkovnyak
(UCLA)



Se Kwon Kim
(UCLA → U Missouri)

Kihoon Lee
(SNU/IBS-CCES)

Also: Hendrik Bluhm (Aachen), Eun-Ah Kim (Cornell) Tae Won Noh (SNU)
Raffi Budakian (Waterloo), Joonho Jang (MIT → SNU)

Works: **SBC**, S. K. Kim, K. Lee, and Y. Tserkovnyak, PRL (2018) **121**, 167001

SBC, H. Bluhm, and E-.A. Kim, PRL (2007) **99**, 197002

J.Jang, **SBC**, R. Budakian, *et al.*, Science (2011) **331**, 186

M. S. Anwar, T. W. Noh, Y. Maeno, *et al.*, Nat Comm (2016) **7**, 13220

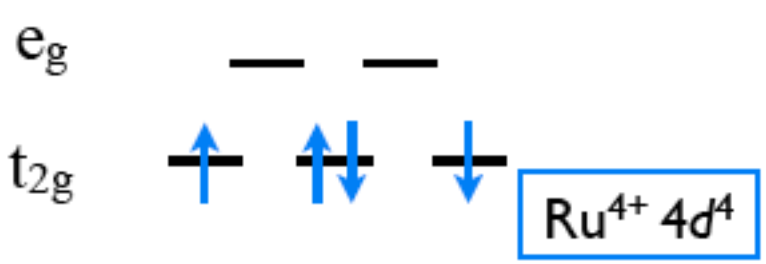
Acknowledgements to:

M. Sigrist, S. Raghu, S. Kivelson, S. Takei, M. S. Anwar, B. Kim, Y. Maeno

Sr_2RuO_4: normal state conventional...

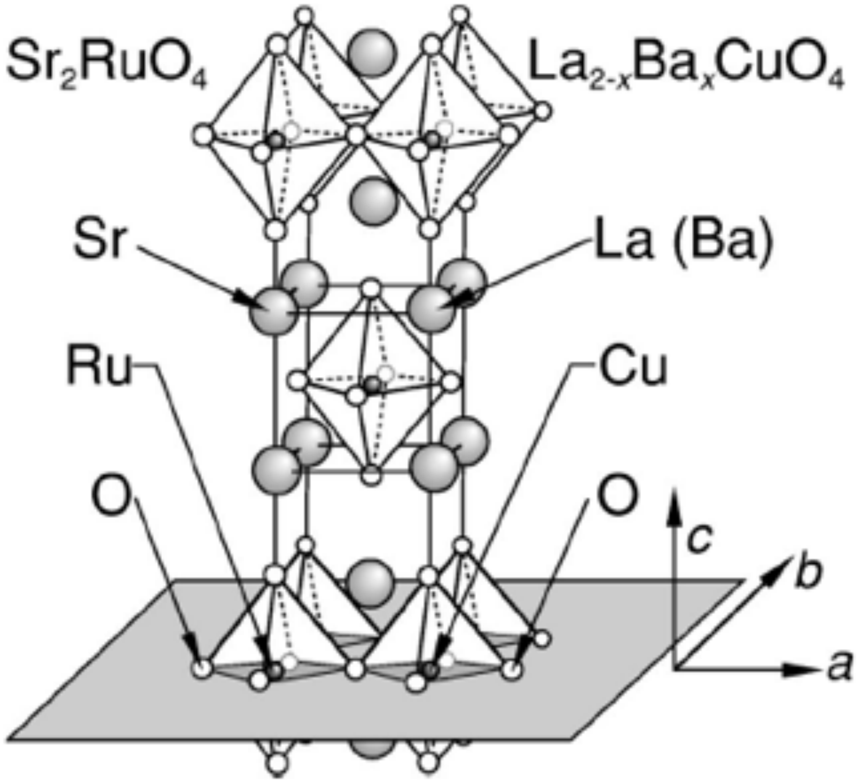
- Fermi liquid at low-T

- Multiple Fermi pockets from ruthenium orbitals



- Only SRO superconductor (T_c=1.5K)

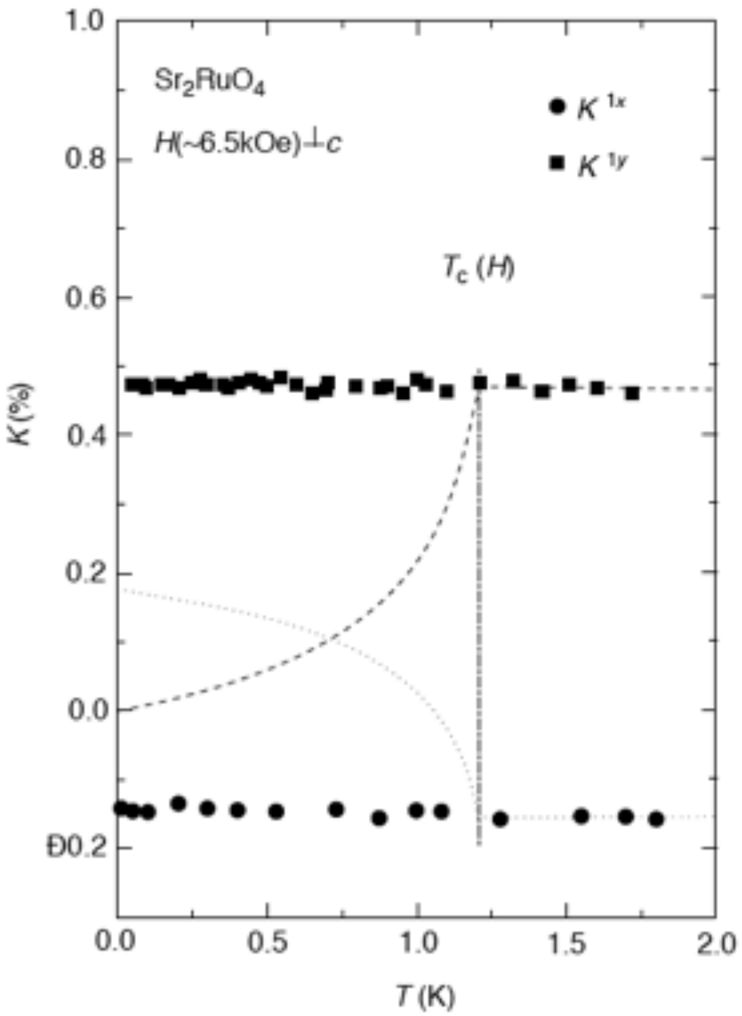
(Maeno *et al.* 1994 Nature)



(Mackenzie and Maeno 2003 RMP)

... but superconductivity most unconventional!

^{17}O Knight shift



(Ishida *et al*. 1998 Nature)

μ spin rotation

(Luke *et al*. 1998 Nature)

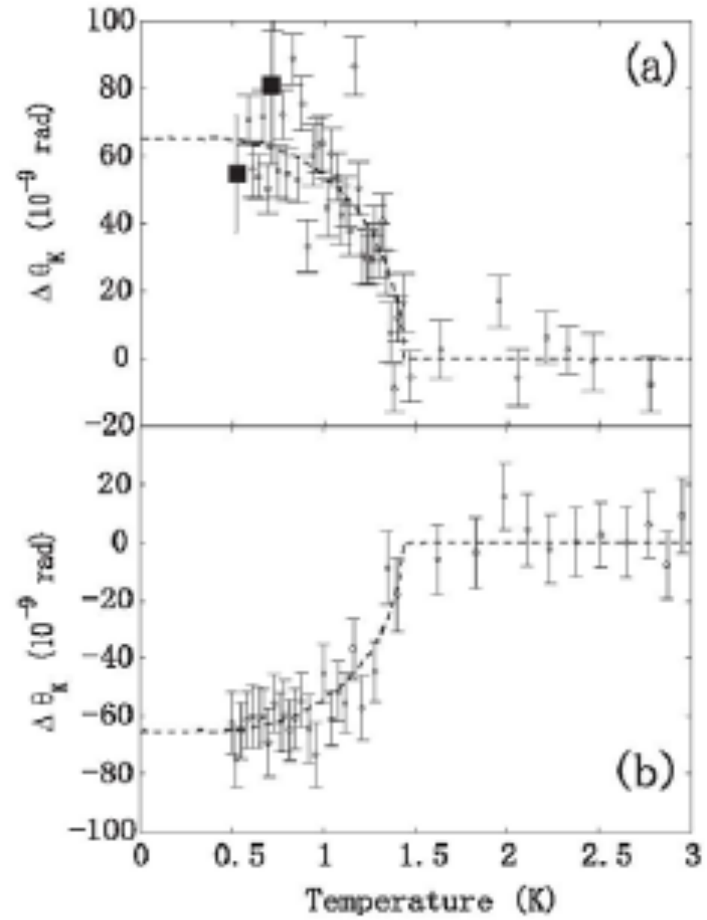
corner junction spontaneous flux

(Nelson *et al*. 2004 Science)

Josephson interferometry

(Kidwingira *et al*. 2006 Science)

Kerr rotation



(Xia *et al*. 2006 PRL)

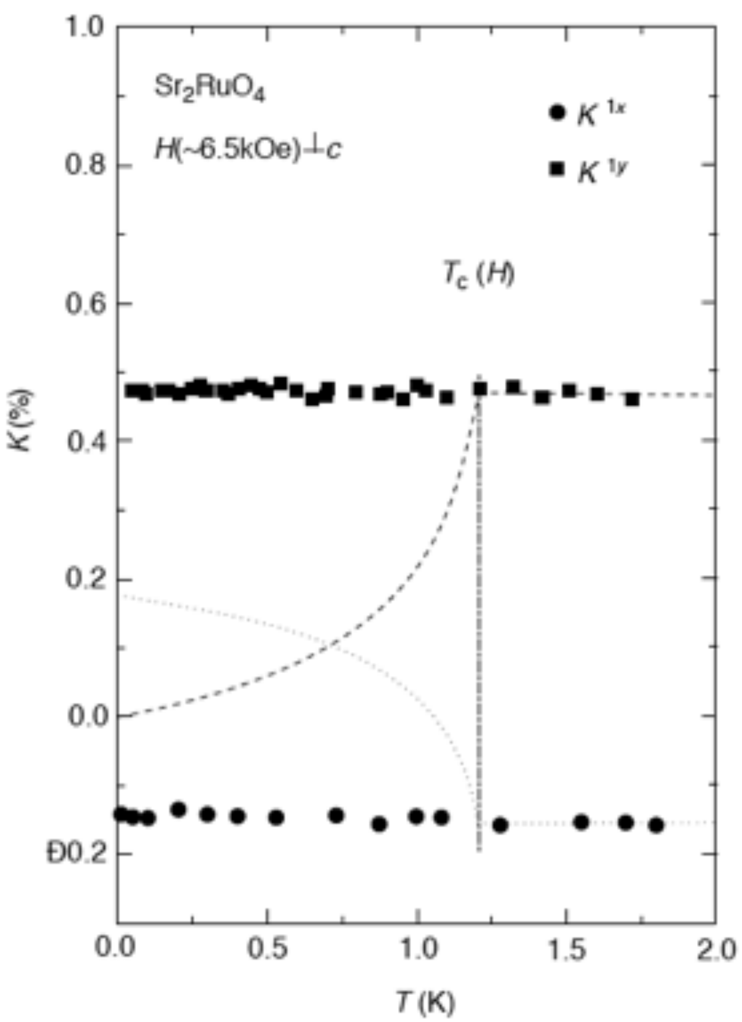
Spin-triplet Cooper pairing: **+** Time-reversal Symmetry Breaking

$$\Delta_{\alpha\beta}(\mathbf{k}) = i(\vec{d}(\mathbf{k}) \cdot \vec{\sigma}\sigma_2)_{\alpha\beta}$$

$$\vec{d}(\mathbf{k}) = (k_x + ik_y)\hat{\mathbf{z}}$$

… but superconductivity most unconventional!

^{17}O Knight shift



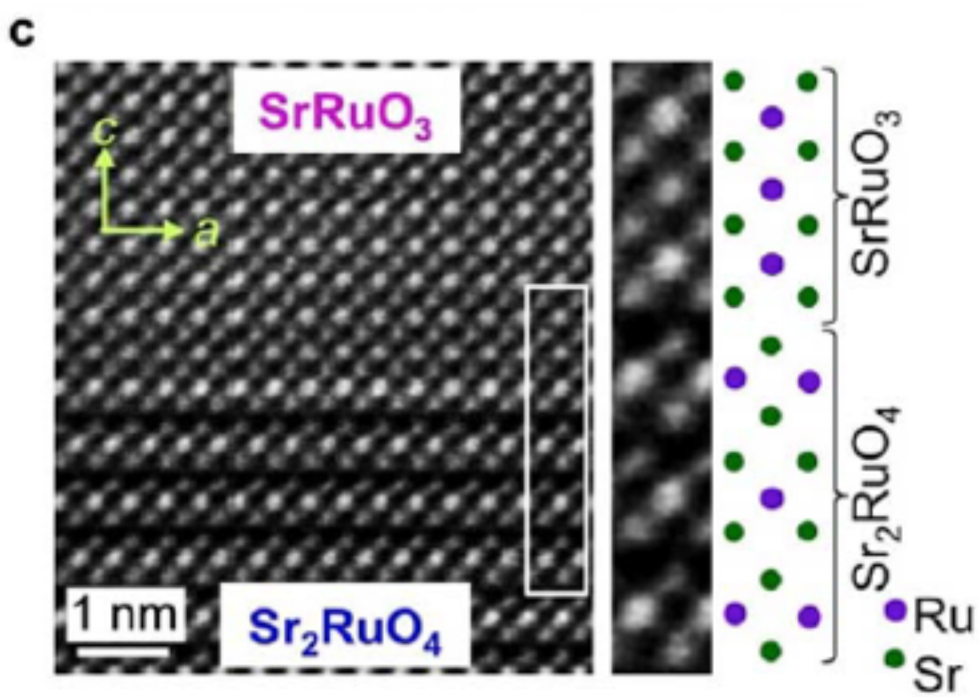
(Ishida *et al*. 1998 Nature)

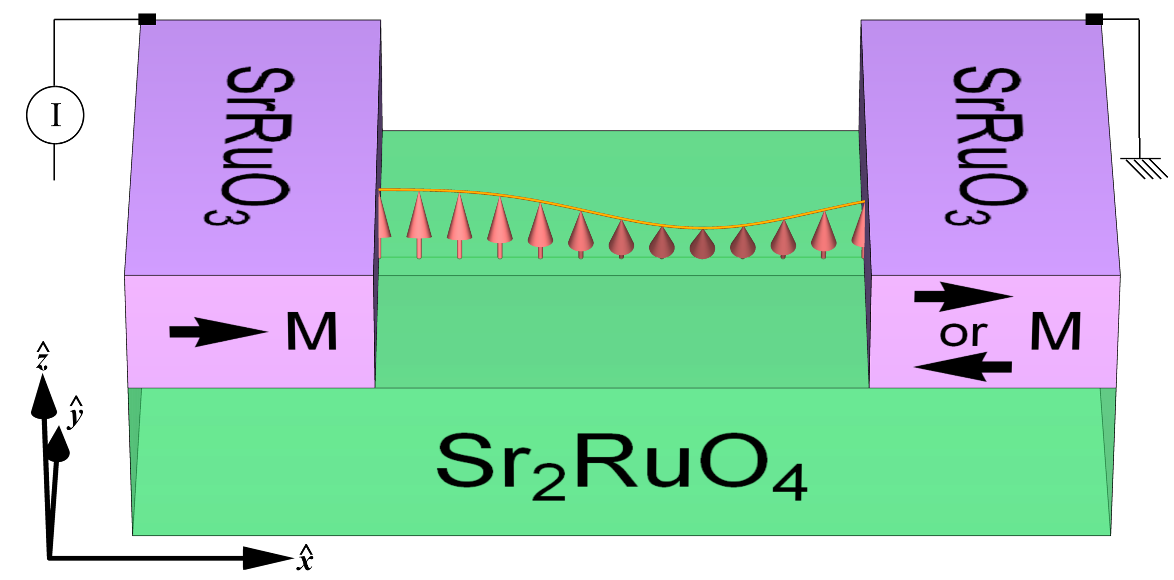
- Zero spin susceptibility for Cooper pairs in conventional superconductor
  ← spin singlet pairing gives us S=0 pairs

- Unchanged spin susceptibility across T_c indicates equal-spin pairing, i.e. S=1 (spin triplet) pairing

- Corroboration needed for spin rotational symmetry breaking:
  ⇒ any spin collective phenomena arising from spin triplet pairing?

Sr_2RuO_4 spin controlled by $SrRuO_3$





- Coupling of $SrRuO_3$ ferromagnetism to spin rotational symmetry breaking of Sr_2RuO_4 spin-triplet superconductivity $\Rightarrow$ generate spin supercurrent & spin wave

- Voltage bias controlled spin supercurrent / wave

Outline

- Spin supercurrent from spin triplet pairing in Sr_2RuO_4
  - ▶ Example: Half-quantum vortex

- Spin collective phenomena in $SrRuO_3$ / Sr_2RuO_4 junction
  - ▶ $SrRuO_3$ / Sr_2RuO_4 heterostructure
  - ▶ 2-terminal junction as spin valve
  - ▶ Spin collective mode generation via AC bias

- Ongoing experimental efforts

Broken spin-rotational symmetry for spin triplet

- Possible to parametrize by the 3-component d-vector:

(Leggett 1975 RMP; Sigrist and Ueda 1991 RMP; Mackenzie and Maeno 2003 RMP)

$$\Delta_{\alpha\beta}(\mathbf{k}) = \begin{bmatrix} \Delta_{\uparrow\uparrow} & \Delta_{\uparrow\downarrow} \\ \Delta_{\downarrow\uparrow} & \Delta_{\downarrow\downarrow} \end{bmatrix} \equiv \begin{bmatrix} -d_x + id_y & d_z \\ d_z & d_x + id_y \end{bmatrix}$$
$$\equiv i(\vec{d}(\mathbf{k}) \cdot \vec{\sigma}\sigma_y)_{\alpha\beta}$$

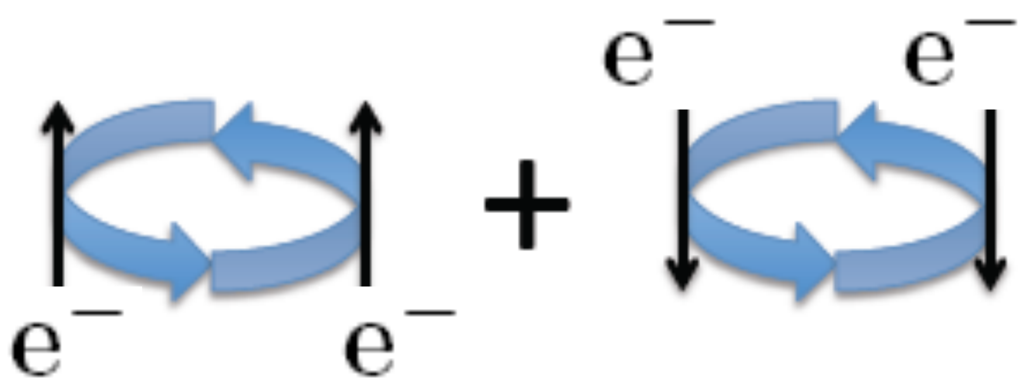
$\leftarrow \vec{d}$ is a 3-component real vector up to a U(1) overall phase, in absence of condensate spin-polarization

- Order parameter rotation is generated by spin: $[S^i, \hat{d}^j] = i\epsilon^{ijk}\hat{d}^k$
  - ▶ $\mathbf{S} \cdot \hat{d}$ rotation is a residual symmetry
  - ▶ the d-vector alignment implies breaking of SO(3) / SO(2) = S_2 symmetry

Spin supercurrent from spin-triplet pairing (1)

$$\Delta_{\alpha\beta}(\mathbf{k}) = \begin{bmatrix} \Delta_{\uparrow\uparrow} & 0 \\ 0 & \Delta_{\downarrow\downarrow} \end{bmatrix} \equiv \begin{bmatrix} -d_x + i d_y & 0 \\ 0 & d_x + i d_y \end{bmatrix}$$
$$\equiv \; i(\vec{d}(\mathbf{k}) \cdot \vec{\sigma}\sigma_y)_{\alpha\beta}$$

✳ consider the case with zero spin-polarization
i.e. the d-vector is real after factoring out exp(iθ)



$$\vec{d} \perp \hat{\mathbf{z}} \;\rightarrow\; \hat{d} = (\cos\alpha, \sin\alpha, 0)$$

- Phase factors of -exp[i(θ-α)] for |↑↑⟩ pairs
  +exp[i(θ+α)] for |↓↓⟩pairs

- Nonzero gradient of α with ∇θ - 2πe**A**/h = 0 means counter-flowing supercurrent from |↑↑⟩ pairs and |↓↓⟩ pairs

⇒ $-\nabla\alpha \propto J_{\uparrow\uparrow} - J_{\downarrow\downarrow}$ = (S_z spin supercurrent)

Spin supercurrent from spin-triplet pairing (2)

$$\Delta_{\alpha\beta}(\mathbf{k}) = \begin{bmatrix} \Delta_{\uparrow\uparrow} & \Delta_{\uparrow\downarrow} \\ \Delta_{\downarrow\uparrow} & \Delta_{\downarrow\downarrow} \end{bmatrix} \equiv \begin{bmatrix} -d_x + i d_y & d_z \\ d_z & d_x + i d_y \end{bmatrix}$$

$$\equiv i(\vec{d}(\mathbf{k}) \cdot \vec{\sigma}\sigma_y)_{\alpha\beta}$$

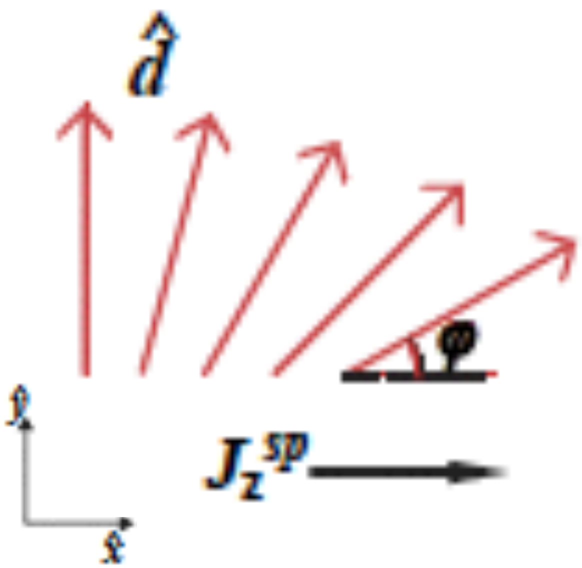
✳ consider the case with zero spin-polarization
i.e. the d-vector is real after factoring out exp[iθ]

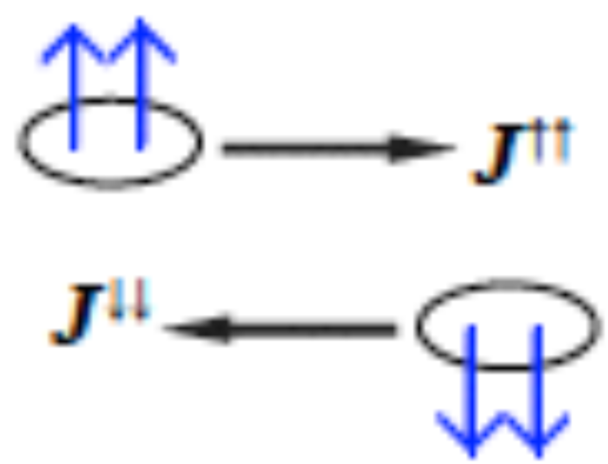
- $\nabla\theta - 2\pi e\mathbf{A}/h \propto \mathbf{J}_s$ (charge supercurrent)

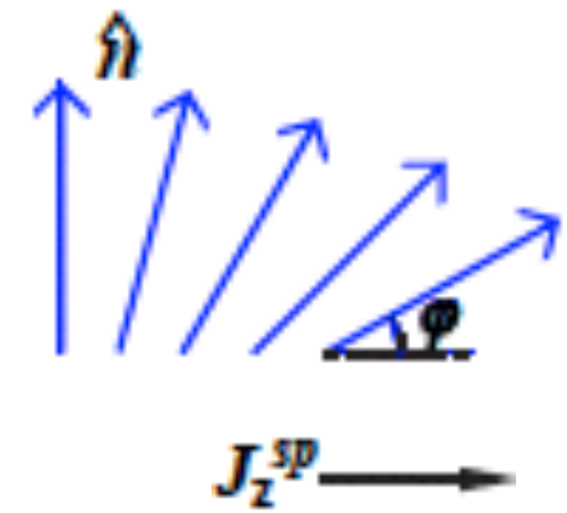
- Spin supercurrent arises with gradient of the d-vector
$\Rightarrow \epsilon^{\alpha\beta\gamma}\hat{d}^{\beta}\nabla\hat{d}^{\gamma} \propto \mathbf{J}_{sp}^{\alpha}$ (supercurrent of S^{α} spin)

Analogy to antiferromagnetic insulator

(**SBC**, Lee, Kim, Tserkovnyak 2018 PRL)



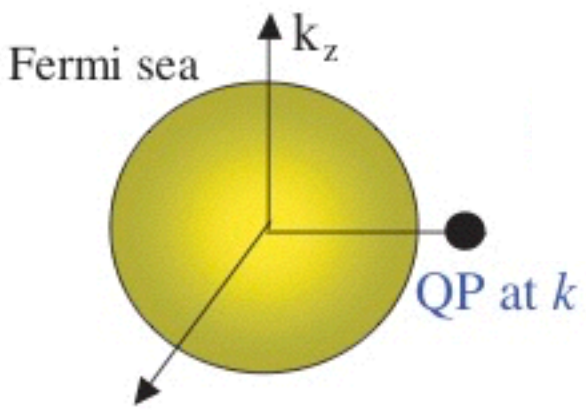




- d-vector winding means Cooper pair spin current, i.e. $|\uparrow\uparrow\rangle$ / $|\downarrow\downarrow\rangle$ counterflow
- Néel vector winding in AFM insulator means coherent magnon current

$\Rightarrow$ coherent transport by Goldstone boson due to spontaneous breaking of continuous symmetry

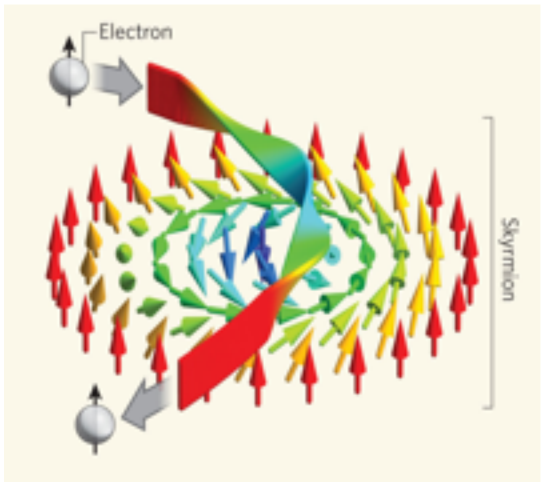
Suppressing dissipation in transport



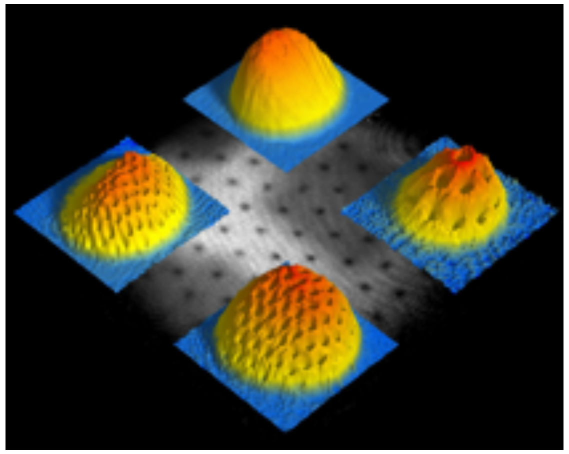
- Transport through excited states in metal:
  ▶ entropy increase
  → thermal dissipation
  → spin current exponentially suppressed

- Adiabatic transport through the ground state:
  ⇒ NO single-particle dissipation!!

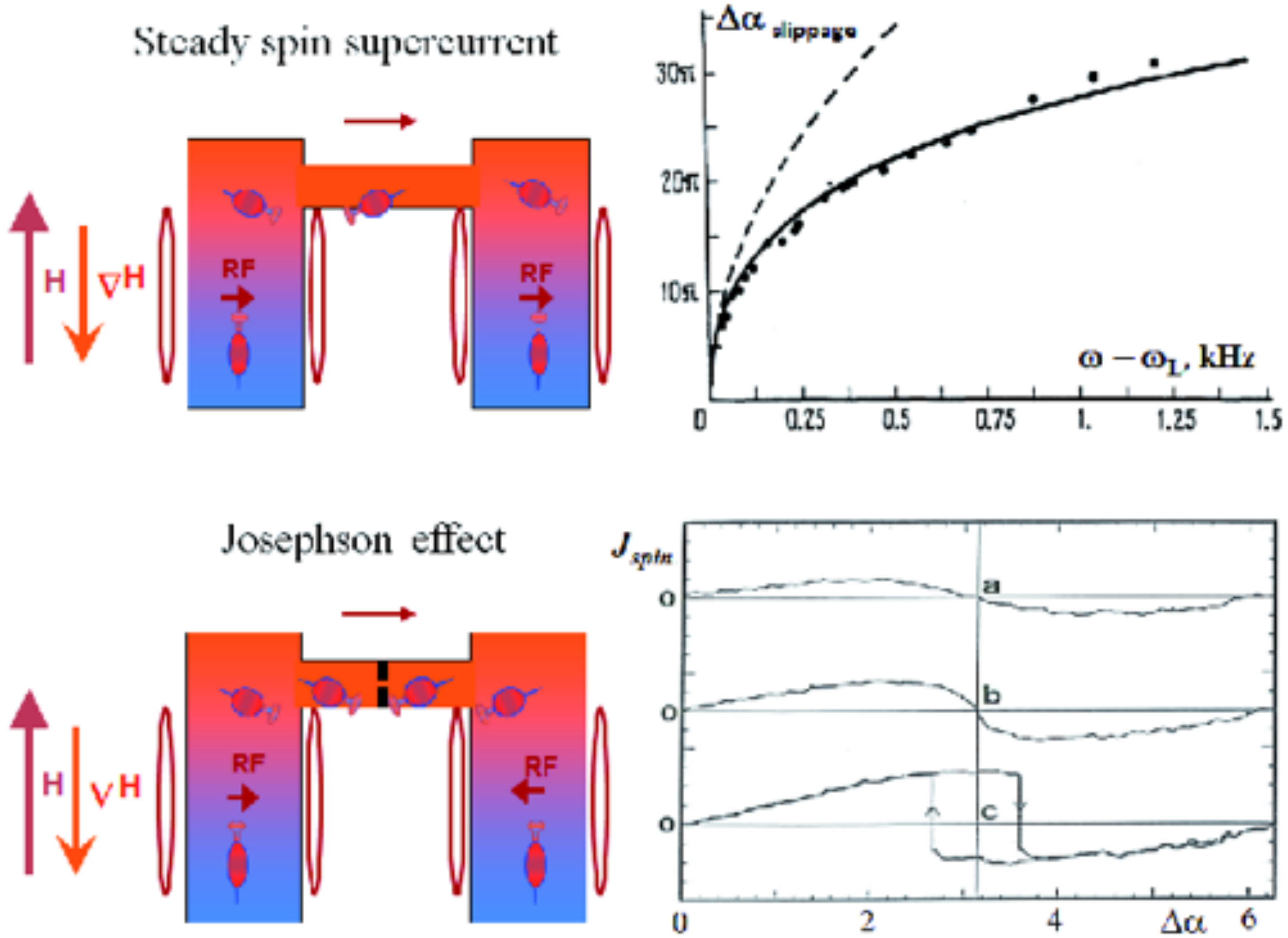
ex) ▶ Berry phase in band insulator
(Pancharatnam Proc Indian Acad Sci 1956;
Berry Proc R Soc 1984)



▶ Superfluid transport in BEC



Long-known spin current in 3He superfluid…



(Fomin 1984 JETP Lett; Borovik-Romanov, Bunkov *et al.* 1984 *ibid*)

- Carried by spin-triplet Cooper pairs

- No exponentially suppression in the bulk
  ⇒ spin / mass superfluidity co-existing!

Outline

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  - ▶ Example: Half-quantum vortex

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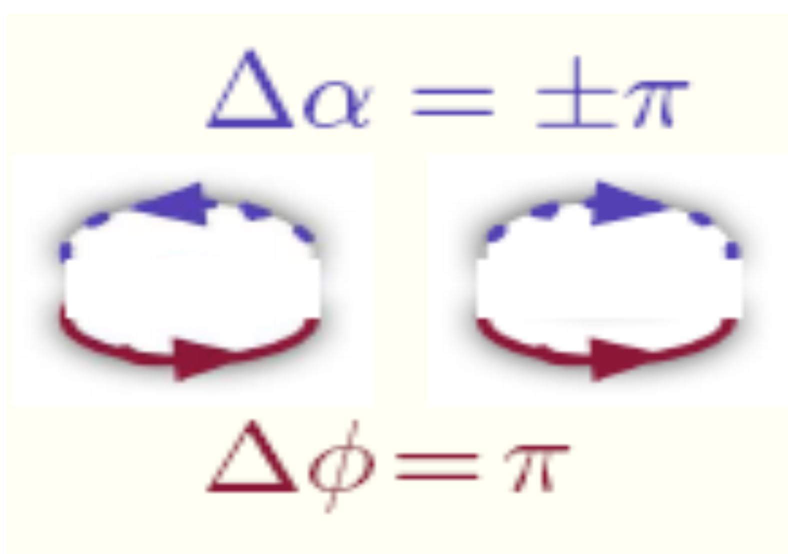
  - ▶ $SrRuO_3$ / Sr_2RuO_4 heterostructure
  - ▶ 2-terminal junction as spin valve
  - ▶ Spin collective mode generation via AC bias

- Ongoing experimental efforts

Half-quantum vortex (HQV) require spin current!

- Vortex flux in conventional superconductor: $h/2e$ (Abrikosov 1953)
  ←2π phase winding for single-valued pair wave function



- For two component condensate, $h/4e$ vortex is allowed
  ←HQV≡(π overall + π relative) winding
  (Salomaa, Volovik 1985 PRL)

  ⇒ spin supercurrent circulation!

HQV Energetics - General

In the London limit,

$$f_{\rm grad}^{\rm 2D} = \frac{1}{2}\left(\frac{\hbar}{2m}\right)^2\left[\rho_{\rm s}\left(\nabla_\perp\phi - \frac{2e}{\hbar c}\mathbf{A}\right)^2 + \rho_{\rm sp}(\nabla_\perp\alpha)^2\right] + \frac{1}{8\pi}(\nabla\times\mathbf{A})^2.$$

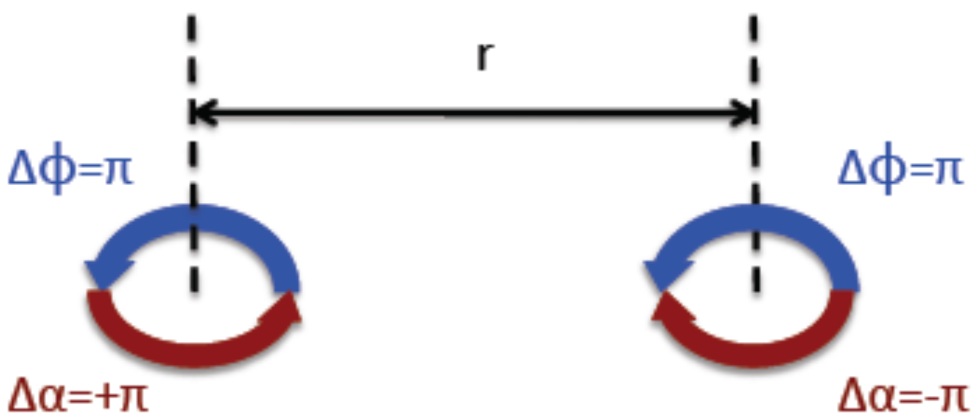
- (screened charge current) + (unscreened spin current)
  ϕ winding α winding

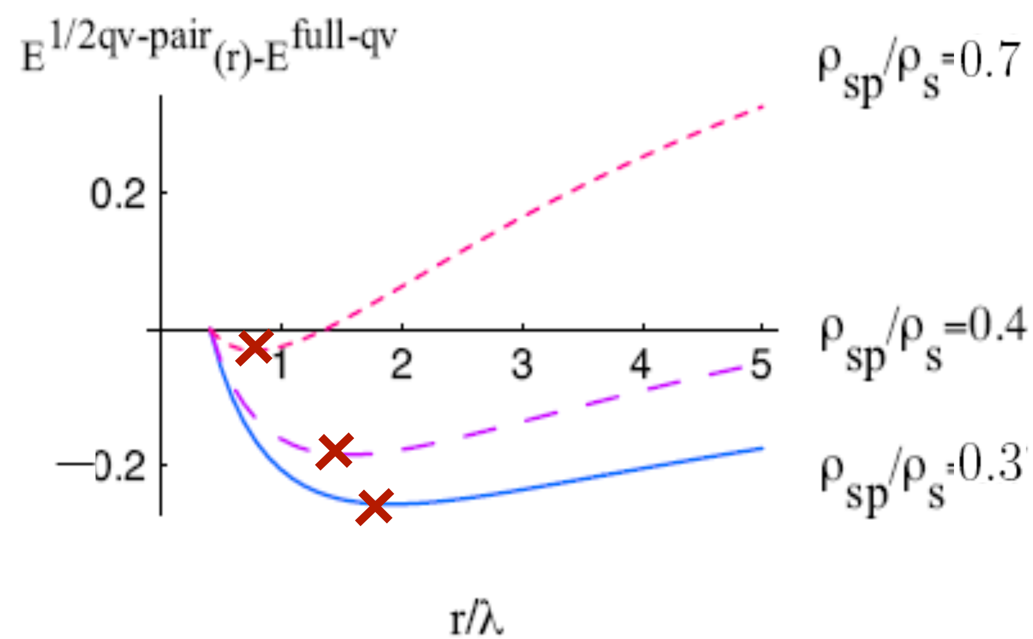
- α winding: energy cost ∝ log L (L - system size)
  smaller ϕ vorticity may NOT reduce energy cost

(Babaev 2005 PRL)

Energy Competition - Magnetic vs. Spin

(**SBC**, Bluhm, Kim 2007 PRL)





- Energy competition in HQV pair:

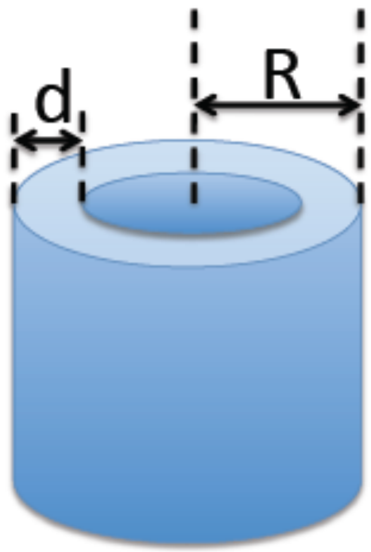
  screened magnetic repulsion

  vs. *unscreened spin attraction*

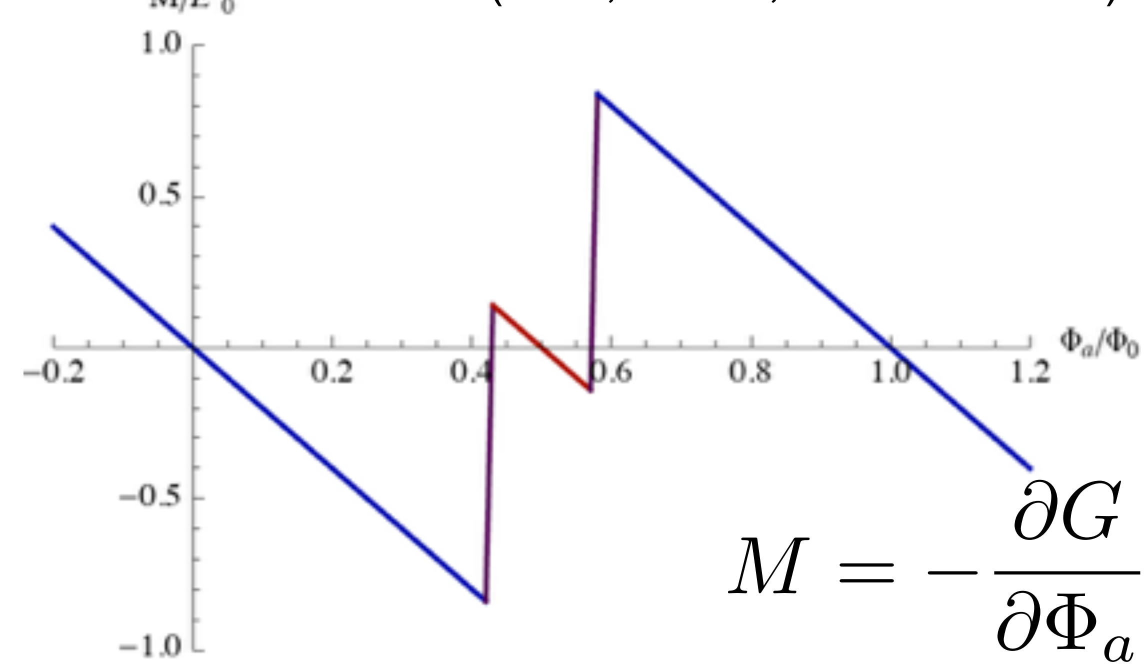
- Long-wavelength spin wins → confined HQV in the bulk

  (Kee, Kim & Maki 2000 PRB)

HQV in Mesoscopic Sample



(**SBC**, Bluhm, Kim 2007 PRL)

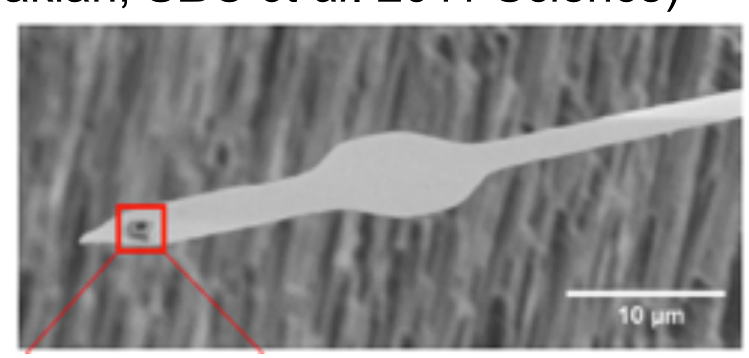


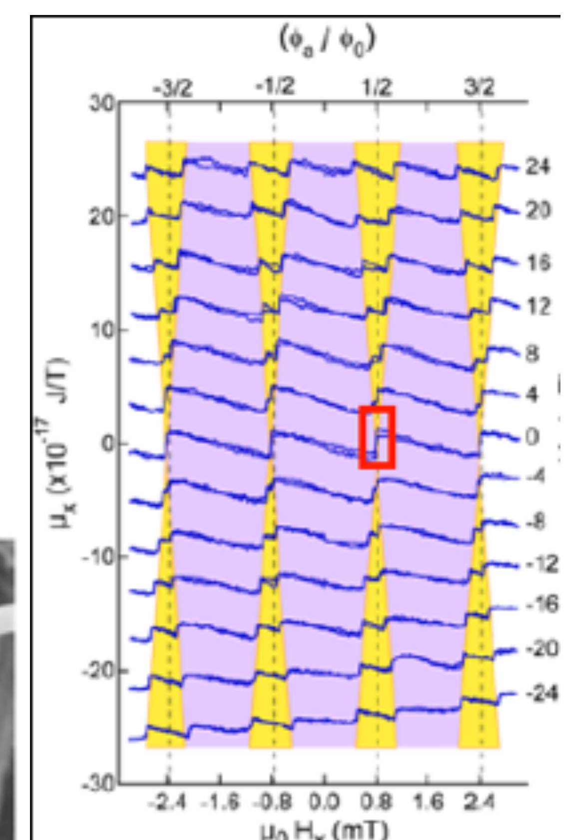
$$M = -\frac{\partial G}{\partial \Phi_a}$$

- Flux entry at $\Phi_a = (n + 1/2)\, h/2e$
  ⇒ Predict ‘double saw-tooth’ magnetization curve.

- Experimental confirmation!

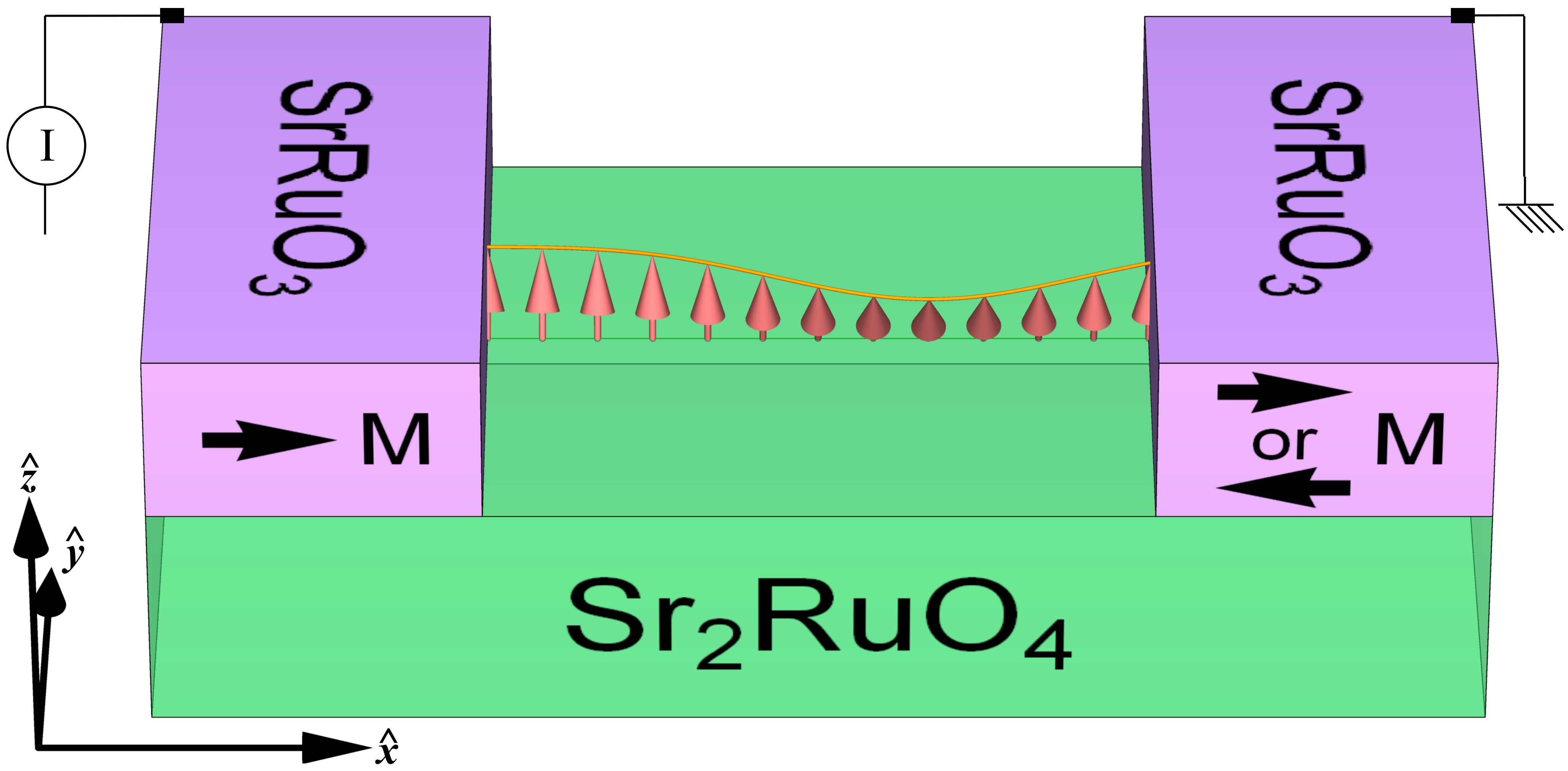
(Jang, Budakian, **SBC** *et al*. 2011 Science)





⇒ How can we generate spin supercurrent in the bulk Sr_2RuO_4?

Single platform for detecting both spin supercurrent and spin collective mode:



Outline

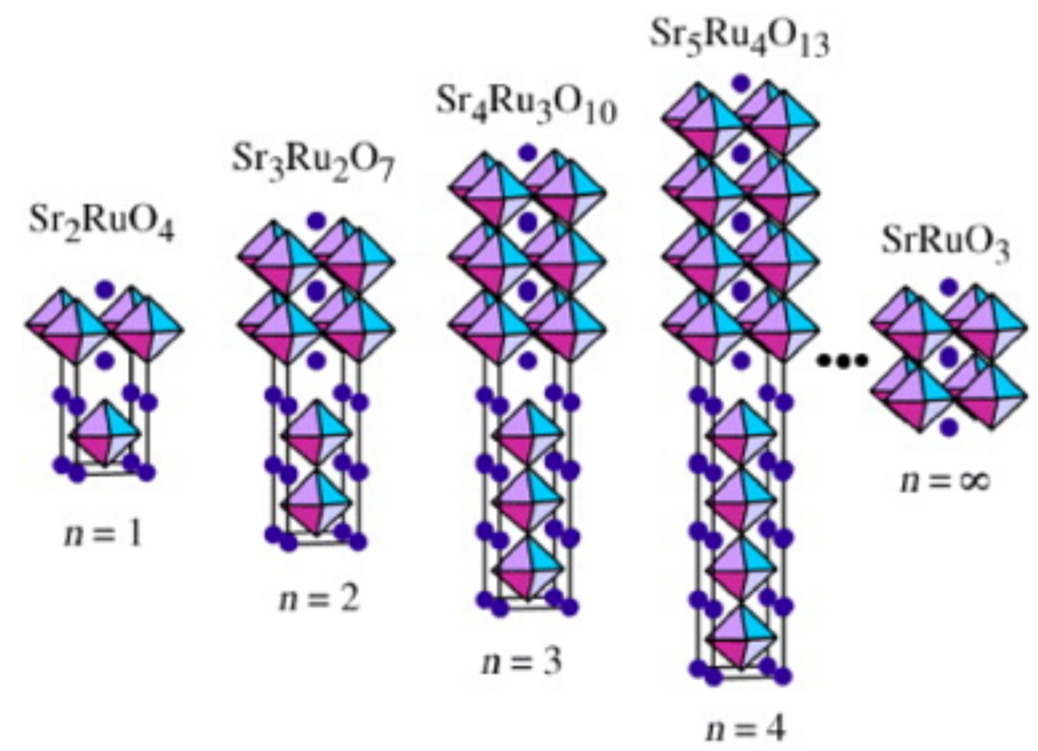
- Spin supercurrent from spin triplet pairing in Sr_2RuO_4
  - ▶ Sr_2RuO_4 spin supercurrent in half-quantum vortex

- Spin collective phenomena in $SrRuO_3$ / Sr_2RuO_4 junction
  - ▶ $SrRuO_3$ / Sr_2RuO_4 heterostructure
  - ▶ 2-terminal junction as spin valve
  - ▶ Spin collective mode generation via AC bias

- Ongoing experimental efforts

$SrRuO_3$: ferromagnetic metal

- Similar structure to Sr_2RuO_4 except for all apical O shared by Ru
  ▶ 1.3% lattice mismatch in (001) direction



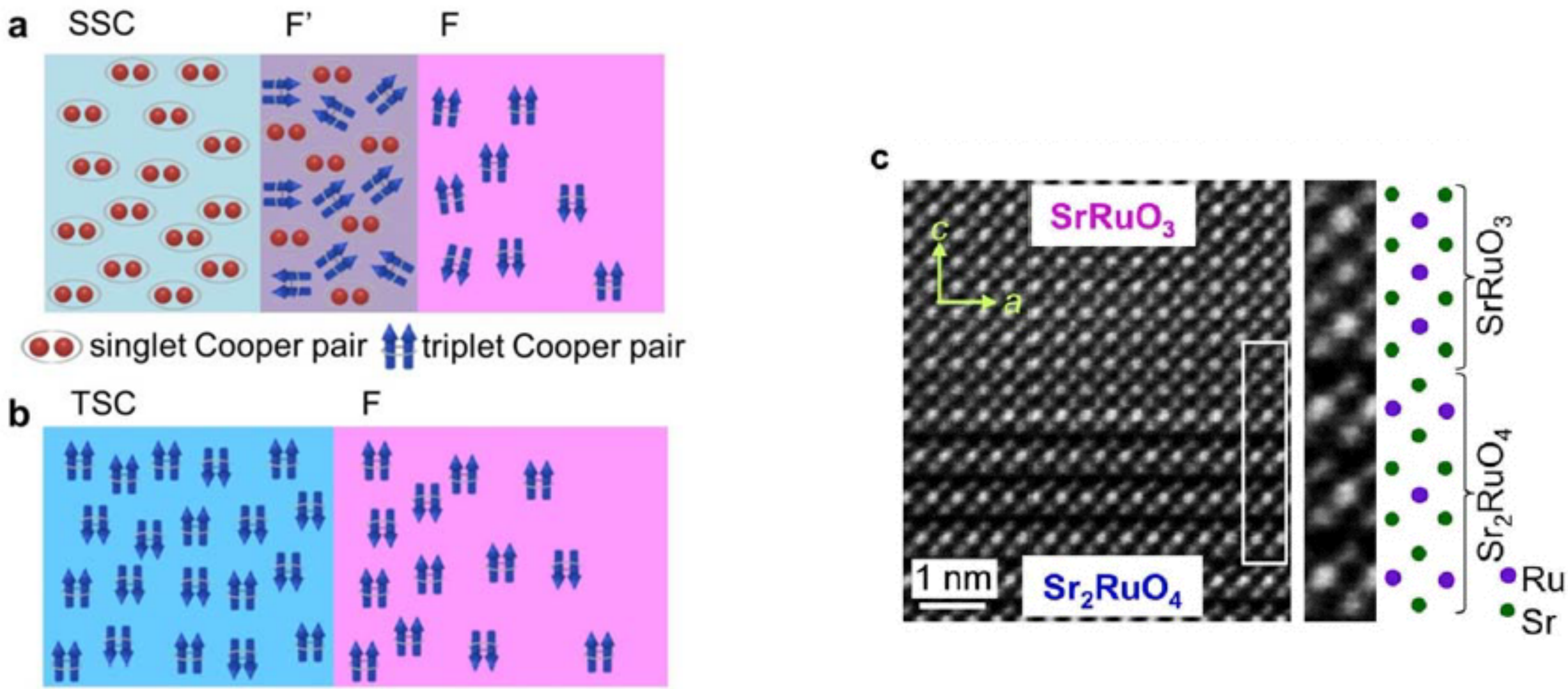
- Thin film remains ferromagnetic w/ 3:1 transport spin polarization

(Nadgorny Eom *et al*. 2003 Appl Phys Lett;
Raychaudhuri Mackenzie Beasley *et al*. 2003 PRB)

- Metallic with Curie temperature of 160K
  (c.f. T_c=1.5K for Sr_2RuO_4)
  ⇒ NO effect of Sr_2RuO_4 superconductivity on magnetization

Characteristics of SRO 214/113 interface

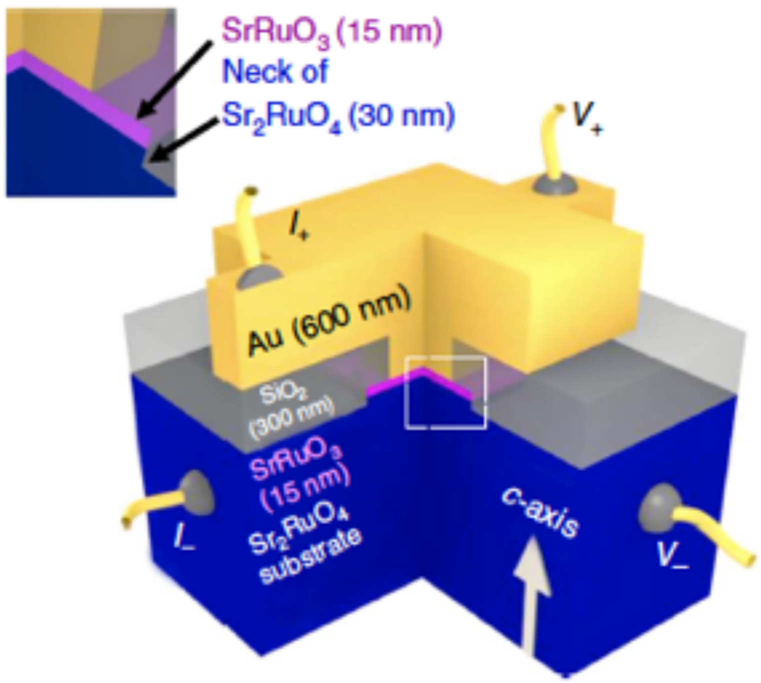
(Anwar, Lee, Noh, Maeno *et al*. 2016 Nat Comm)



- FM/SC junction may NOT require impurity for proximity effect
- 113/214 interface is impurity-free even on the atomic scale

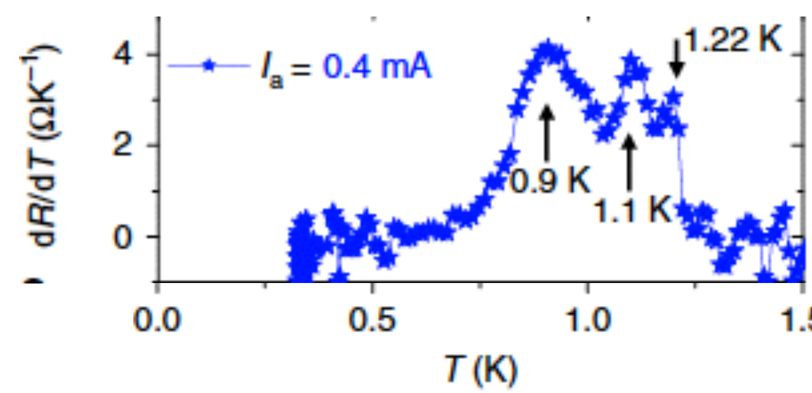
SRO 214/113 proximity effect measurement

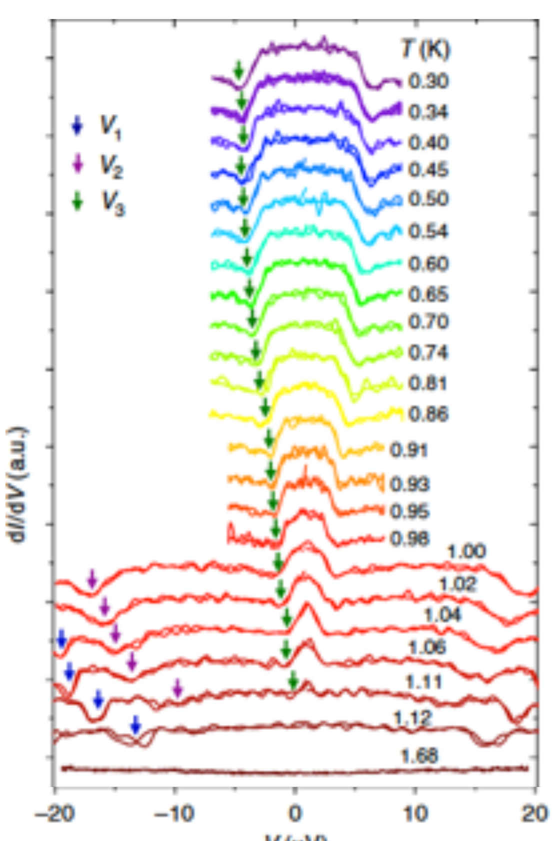
(Anwar, Lee, Noh, Maeno *et al*. 2016 Nat Comm)



- Measured junction voltage V for the given applied current I

- Multiple transition seen on dR/dT as indication of proximity effect onset

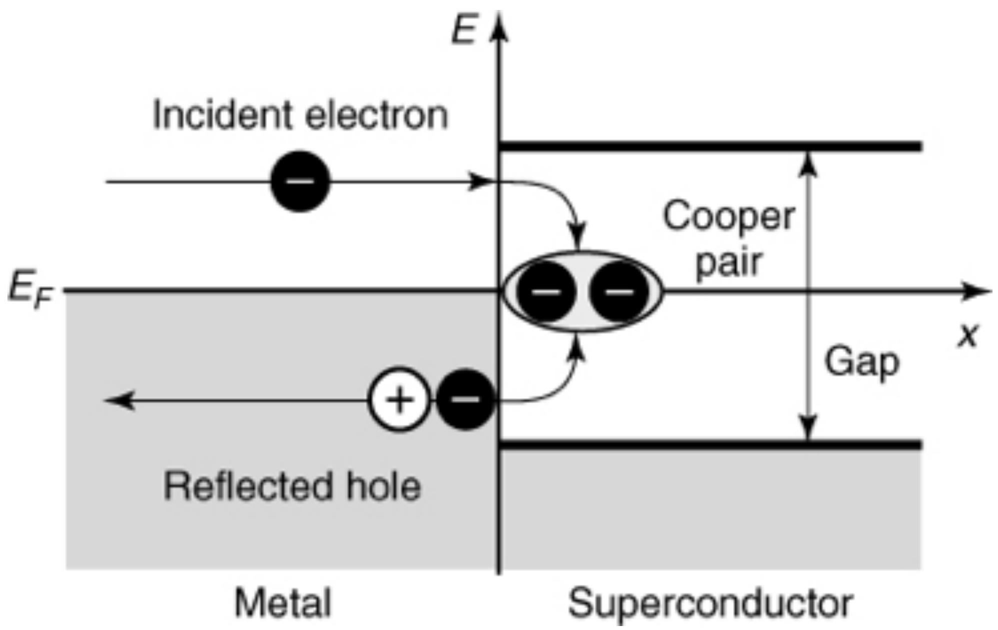




- dI/dV measurement to obtain the pairing gap at each interface

Andreev reflection in spin-triplet superconductor

- Andreev reflection always occurs between superconductor and metal



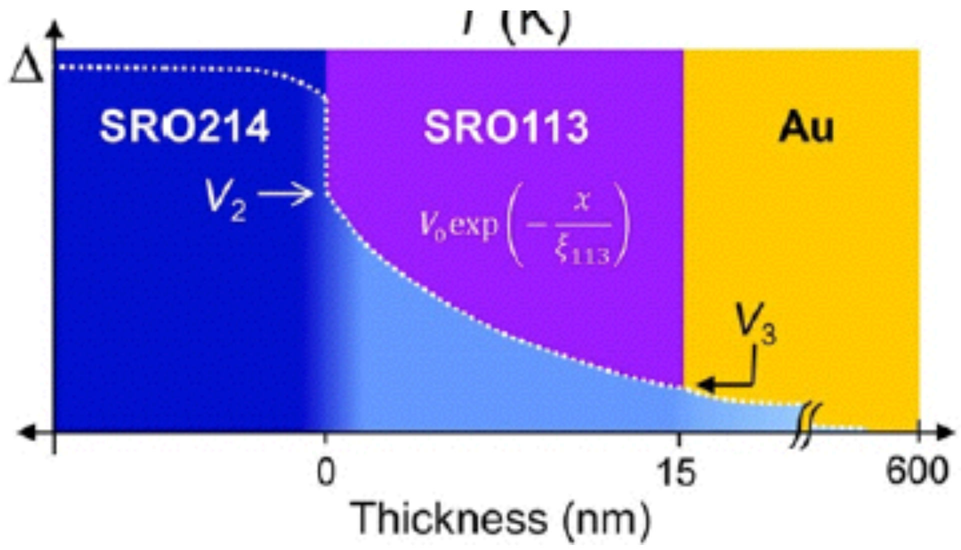
▶ electron near Fermi level gets reflected as hole

▶ electron / hole equal superposition localized at the interface

- Andreev reflection at the metal / spin-triplet SC interface leads to formation of equal-spin Cooper pairs in spin-triplet SC
  ⇒ In absence of spin-flip scattering, an FM metal injects spin-polarized supercurrent into spin-triplet SC

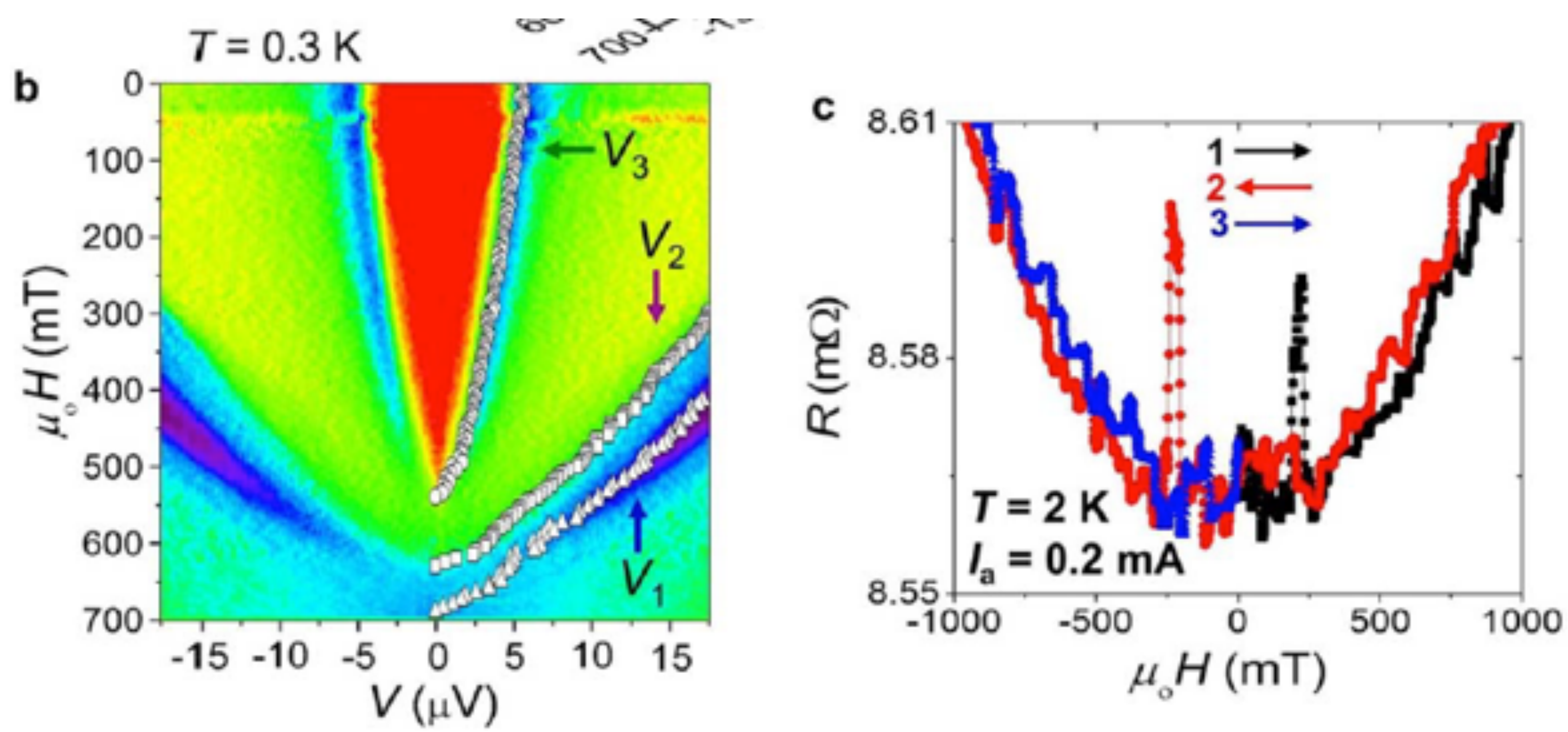
Evidence for triplet pairing induced in 113

(Anwar, Lee, Noh, Maeno *et al*. 2016 Nat Comm)



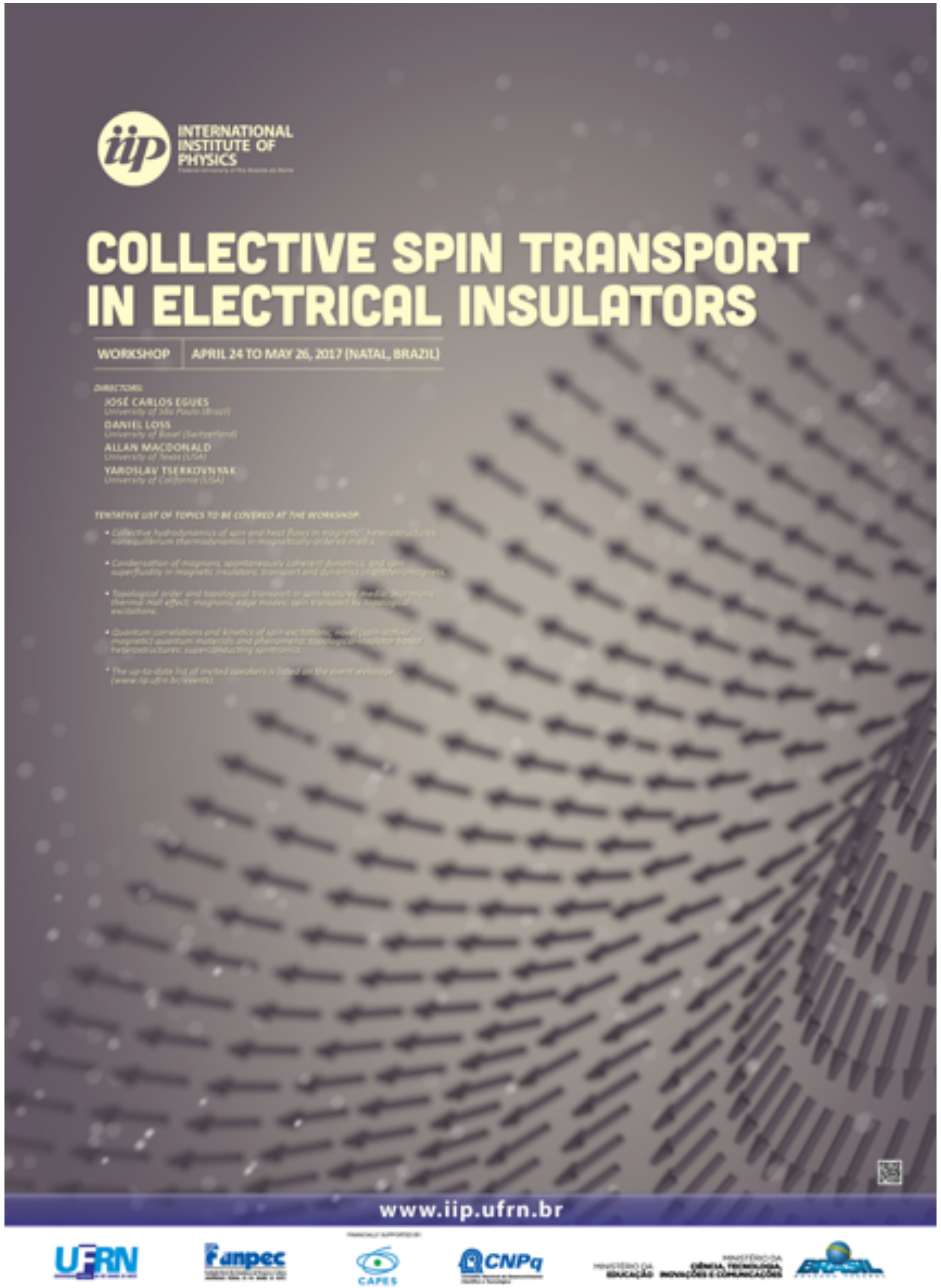
- Cooper pair decay length in 113 is very long

  → Andreev reflection, not tunneling at 113/214 interface



- Proximity effect intrinsic, e.g. unaffected by 113 domain switching

⇒ proximity effect NOT collective phenomenon…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  - ▶ Sr_2RuO_4 spin supercurrent in half-quantum vortex
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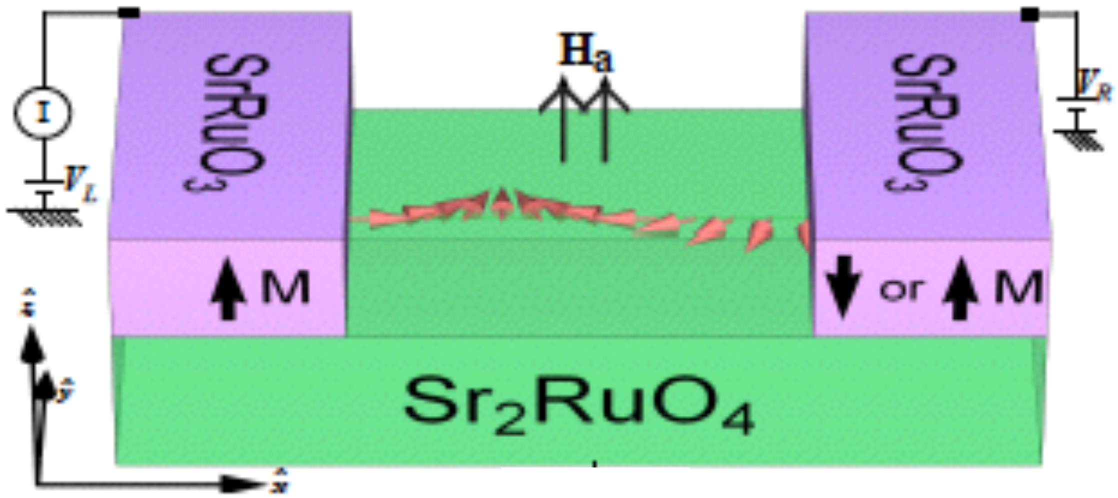
Spin supercurrent proposal for SRO 214/113

(**SBC**, Lee, Kim, Tserkovnyak 2018 PRL)



- For collinear 113 magnetization, independent Andreev conductance for spin-up / spin-down:

$$I^{\sigma}_{L,R} = \pm g^{\sigma\sigma}_{L,R}(V_{L,R} - \hbar\partial_t\varphi_\sigma/2e)$$

- Cooper pairs injected from 113 leads are spin-polarized as parametrized by $p_{L,R} = \sum_\sigma \sigma g^{\sigma\sigma}_{L,R} / \sum_\sigma g^{\sigma\sigma}_{L,R}$ (≈0.5 estimated for 113)

  → spin-polarized supercurrent, i.e. spin supercurrent along with charge super current

Spin transport from d-vector

- The effective easy-plane d-vector Hamiltonian gives, as the equation of motion, the local Cooper pair spin conservation:

$$H_{eff} = \frac{1}{2}\int d\mathbf{r}[A(\nabla\alpha)^2 + \gamma^2 S_z^2/\chi]$$ (with $[\alpha, S_z]=i\hbar$)

$$\Rightarrow \quad \partial_t S_z + \nabla\cdot\mathbf{J}_{sp}^z = 0$$ (with $\mathbf{J}_{sp}^{\alpha} = -A\nabla\alpha$)

- Finite lifetime can be introduced phenomenologically for imbalance between |↑↑〉 and |↓↓〉, while retaining spin ordering

i.e. $$\partial_t S_z + \nabla\cdot\mathbf{J}_{sp}^z = -\frac{1}{\tau}S_z$$ (with $1/\boldsymbol{\tau} = \tilde{\alpha} n\hbar\boldsymbol{\gamma}_e^2/\boldsymbol{\chi}$)

(Tserkovnyak, Brataas, Bauer, Halperin 2005 RMP)

- Cooper pair spin current inverse distance decay w/ finite lifetime $\longleftrightarrow$ quasi-particle spin current exponentially suppressed beyond spin diffusion length

SRO Cooper pair spin valve

(**SBC**, Lee, Kim, Tserkovnyak 2018 PRL)

- Boundary condition for charge & spin supercurrent:

$$\sum_\sigma (I_L^\sigma - I_R^\sigma) = 0, \qquad \sum_\sigma \sigma(I_L^\sigma - I_R^\sigma) = 2\tilde{\alpha} e \Omega_s S L$$

- No distance cutoff for supercurrent dependence on conductance spin polarization difference between two leads

$$I^c = \sum_\sigma I^\sigma = I_0 \left[1 - \frac{g_L g_R (p_L - p_R)^2}{(g_L + g_R)(g_L + g_R + g_\alpha) - (p_L g_L + p_R g_R)^2}\right]$$

(where $g_\alpha = 4\tilde{\alpha} n S L e^2/\hbar$)

- Magnetoresistance for beyond spin diffusion length proportional to condensate fraction → increase with 1-T/T_C (Romeo, Citro 2013 PRL)

⇒ d-vector Hamiltonian convenient for collective spin dynamics

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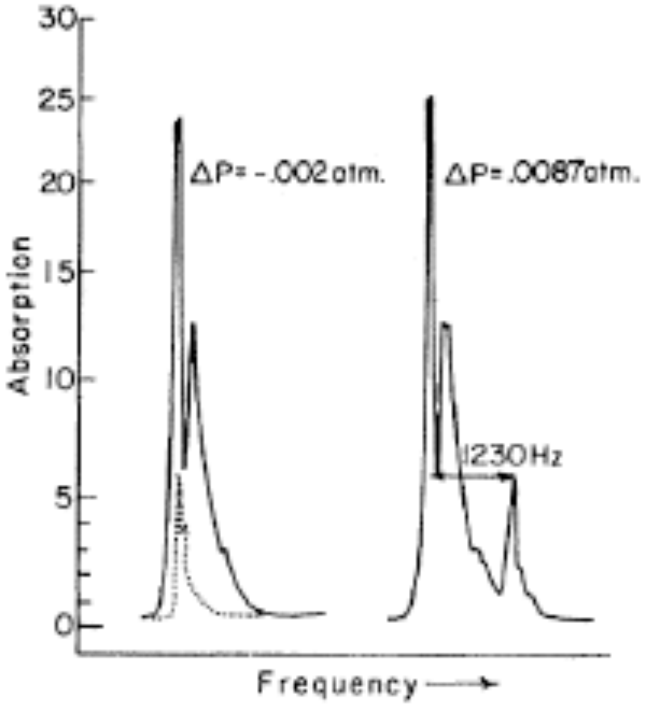
Symmetry breaking leads to collective modes

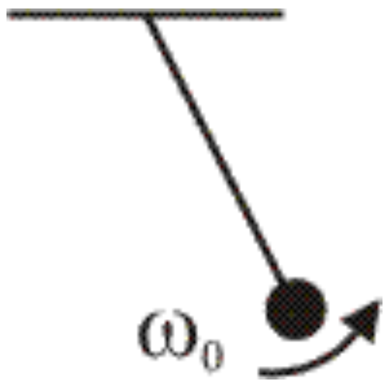
- Crystal (broken translational symm) → phonons
  Magnet (broken rotational symm) → spin wave



- ^3He: 1st known example of
  spin-triplet Cooper pairing
  ⇒ NMR resonance signature of
  spin collective mode confirmed
  spin-triplet pairing
  (Osheroff, Lee, Richardson *et al*. 1972 PRL;
  A J Leggett 1974 Ann Phys; Maki, Ebisawa 1974 PRL)

- NO analogous resonance detected in Sr_2RuO_4 superconductor

d-vector collective motion



$$H[\hat{\mathbf{d}}, \mathbf{S}] = \frac{1}{2\chi_{\rm sp}}\mathbf{S}^2 + \Gamma_0 \hat{\mathbf{d}}_{\parallel}^2$$

→ oscillation of relative phase between |↑↑〉 and |↓↓〉

- For the (d_y,S_x) mode in x-axis spin quantization (Leggett 1975 RMP)
  - ▶ Cooper pair spin state exp(iα) |↑↑〉 + exp(-iα) |↓↓〉 with α=0 at the ground state
  - ▶ $d_y \approx \alpha$ means we can take the relative phase as the pendulum angle

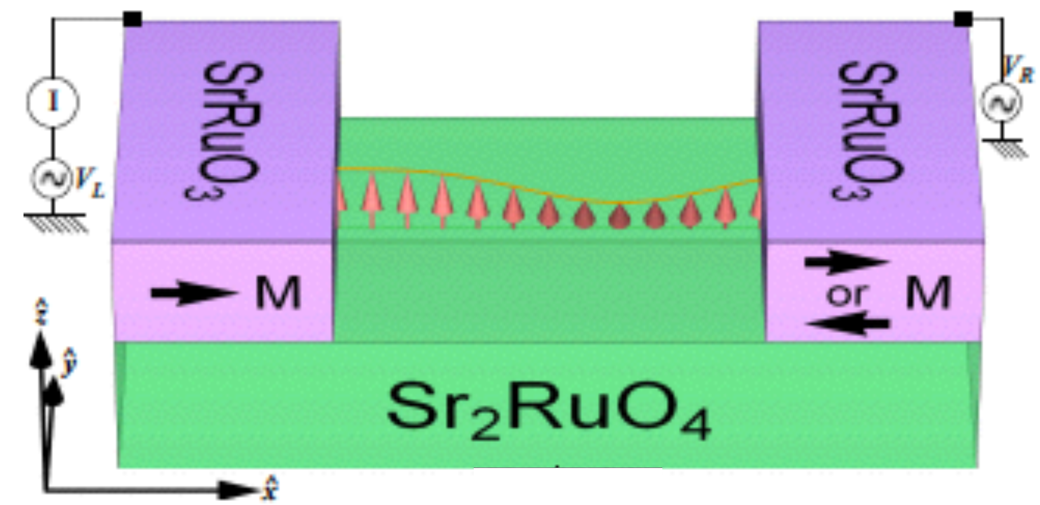
- SRO 113/214 can induce oscillation of relative phase between |↑↑〉 and |↓↓〉

SRO spin mode detection by 214/113 junction

(**SBC**, Lee, Kim, Tserkovnyak 2018 PRL)

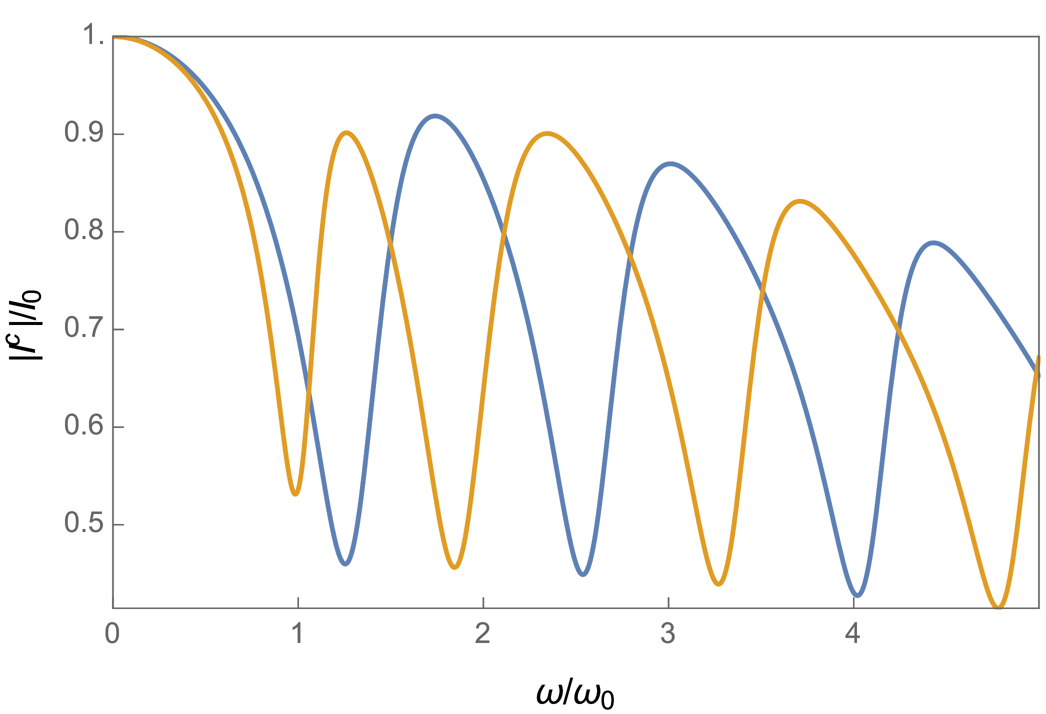
- Spin-polarized AC supercurrent injection excites spin collective mode

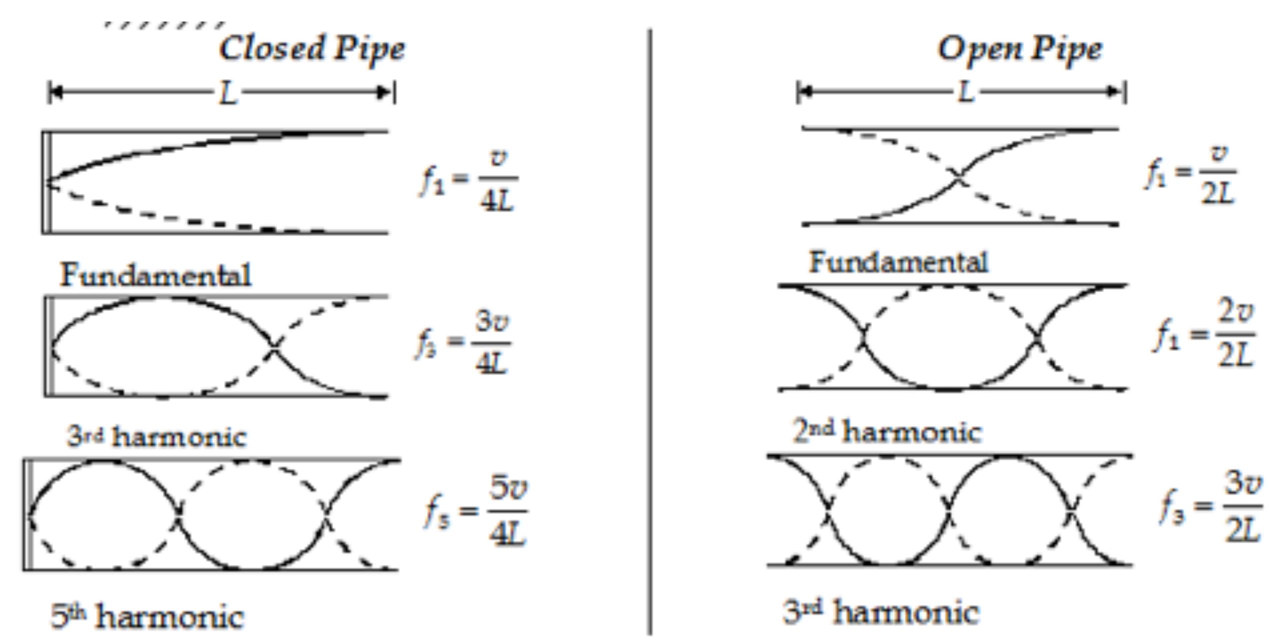
  ⇒ electrically generated spin wave



- Current maxima indicates resonant frequency for spin wave

  → maxima / minima dependent on magnetization alignment





Blue: parallel 113 magnetization

Orange: opposite 113 magnetization

Outline

- Spin supercurrent from spin triplet pairing in Sr_2RuO_4
  - ▶ Sr_2RuO_4 spin supercurrent in half-quantum vortex

- Spin collective phenomena in $SrRuO_3$ / Sr_2RuO_4 junction
  - ▶ $SrRuO_3$ / Sr_2RuO_4 heterostructure
  - ▶ 2-terminal junction as spin valve
  - ▶ Spin collective mode generation via AC bias

- Ongoing experimental efforts

EunKyo Ko
(SNU/IBS-CCES)

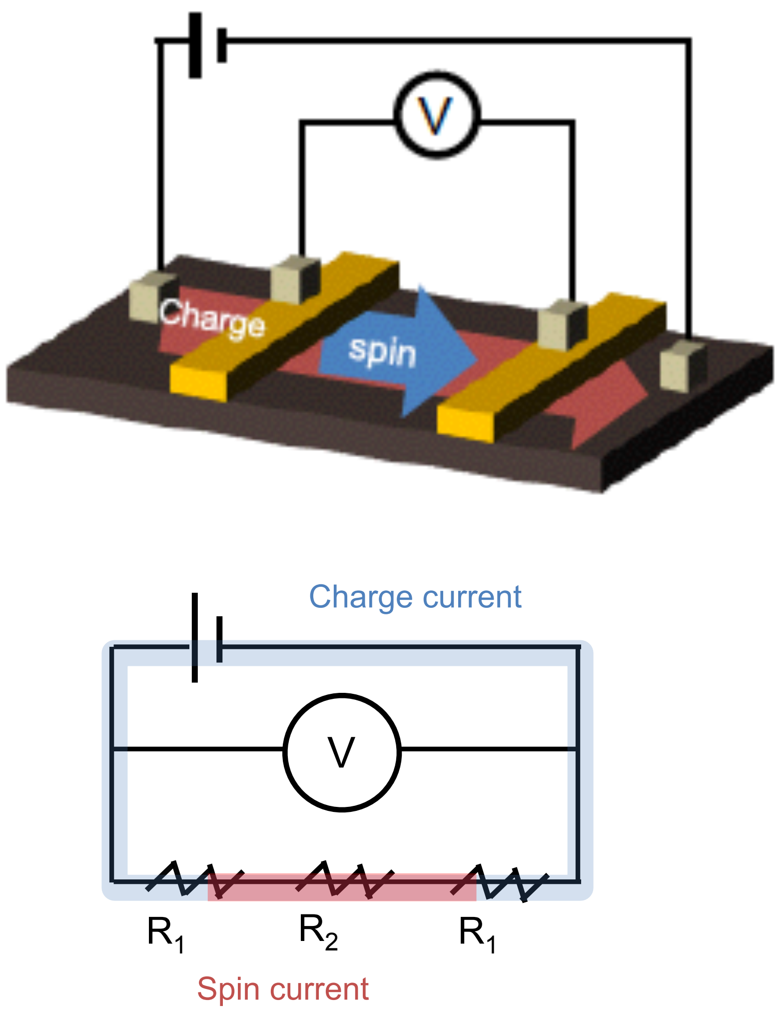
Bongju Kim
(SNU/IBS-CCES)

Tae Won Noh
(SNU/IBS-CCES)

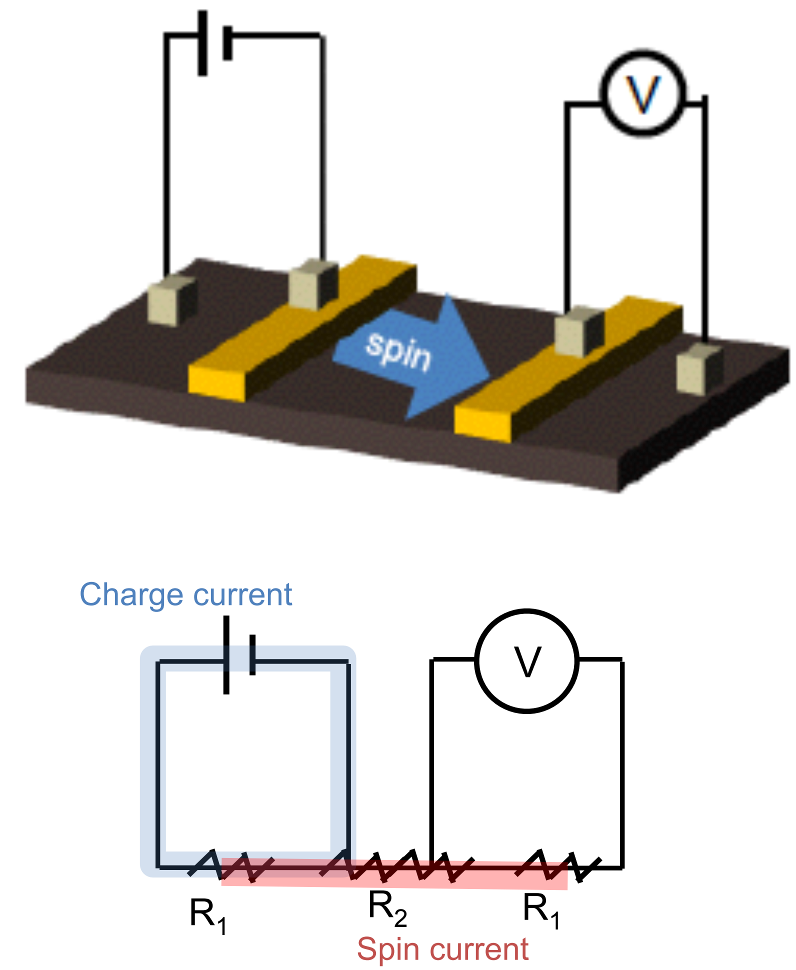
Non-local setup

(Ko, B Kim, **SBC**, Noh *et al* in progress)

▶ local measurement



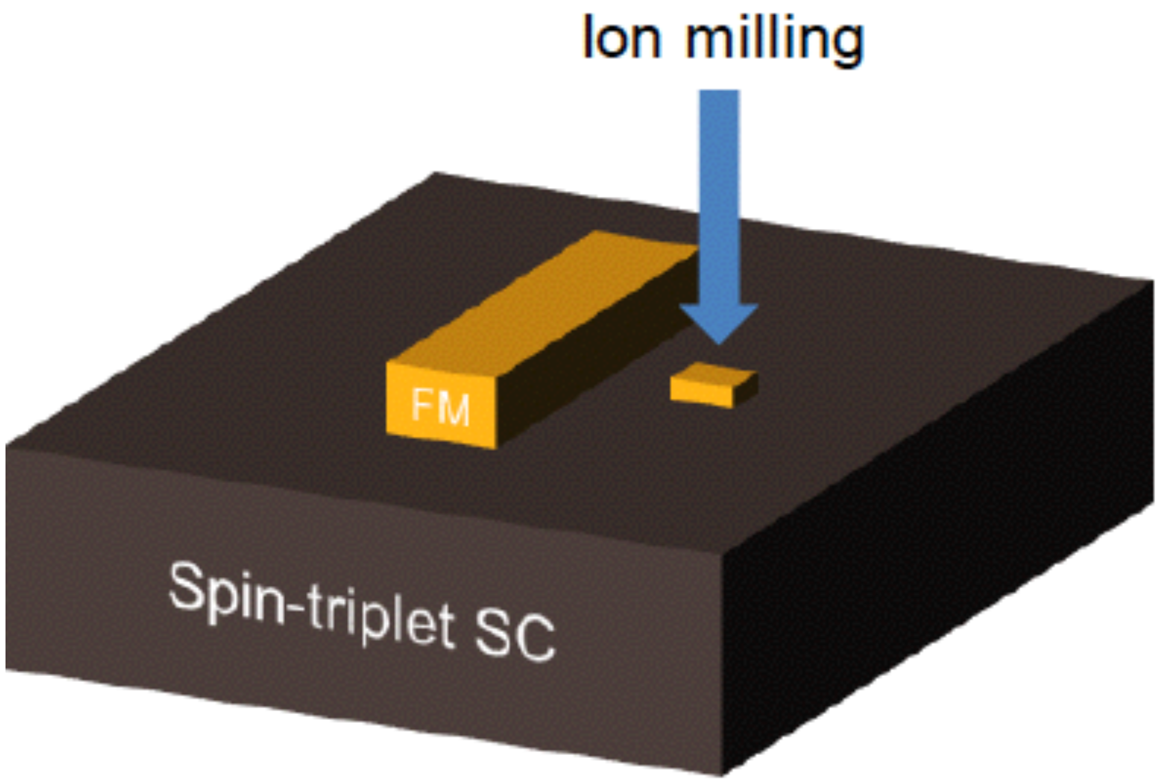
▶ non-local measurement



- Non-local measurement is widely used for detecting giant magnetoresistance in spin valve device
- Non-local measurement can detect pure spin current while local measurement measure both charge and spin current

General device fabrication 1

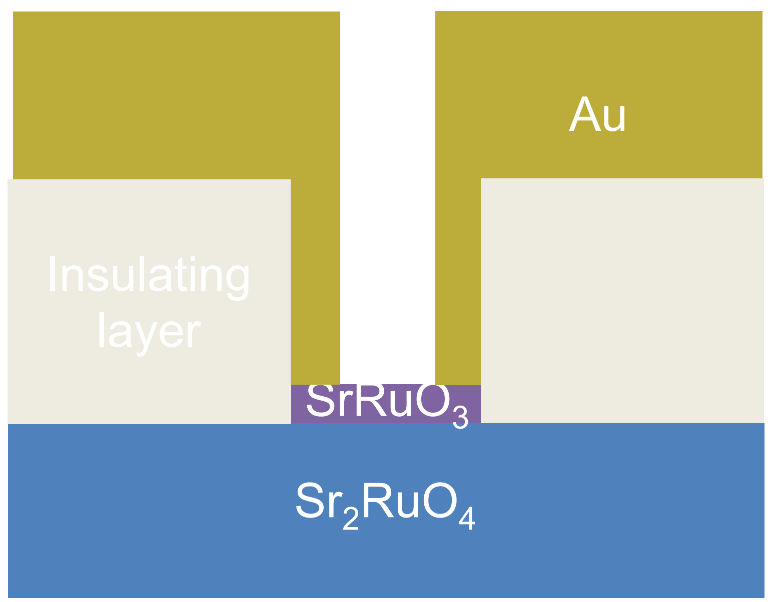
(Ko, B Kim, **SBC**, Noh *et al* in progress)

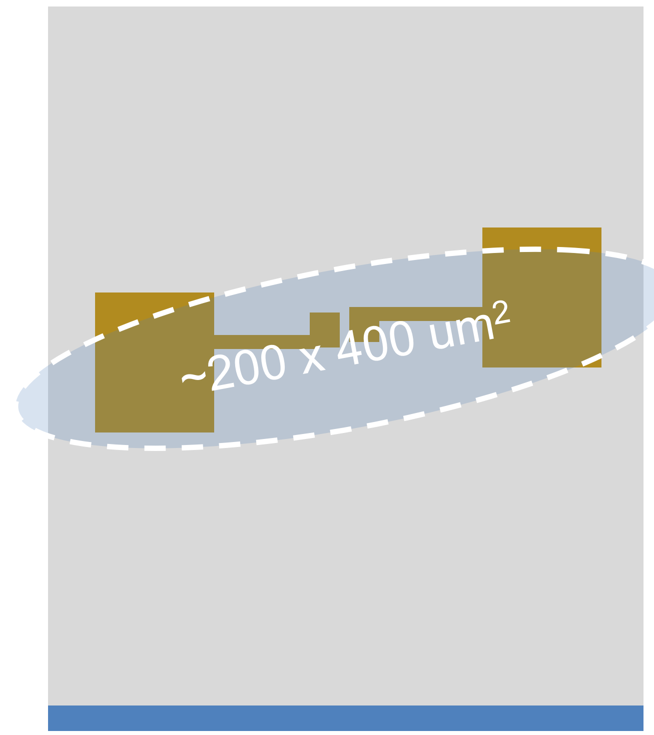


- For a spin valve device, different coercivity required for two ferromagnets
- Different thickness means different coercivity for $SrRuO_3$

General device fabrication 2

(Ko, B Kim, **SBC**, Noh *et al* in progress)

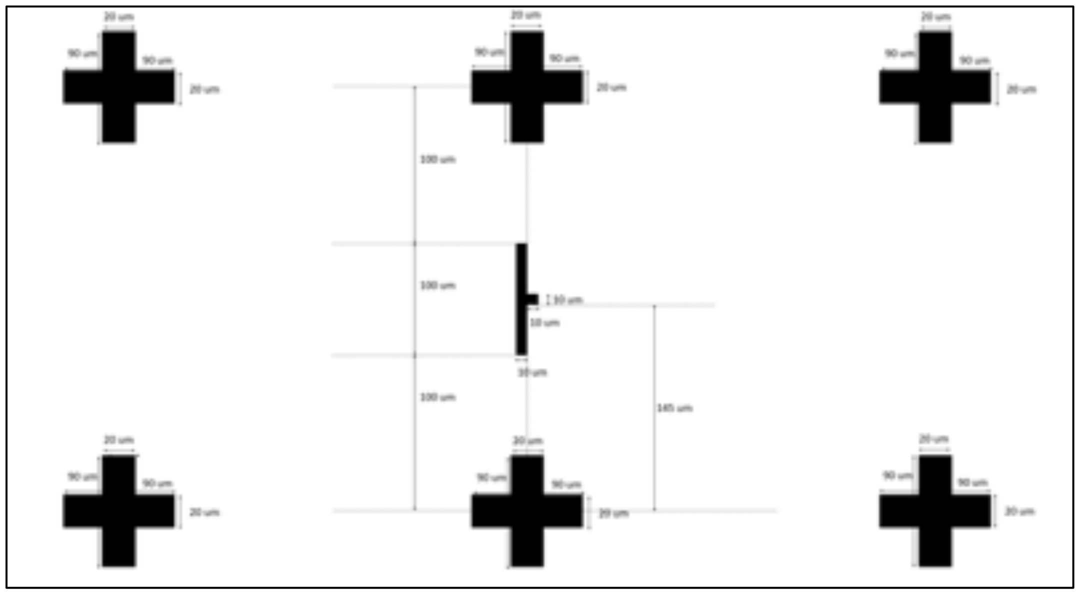


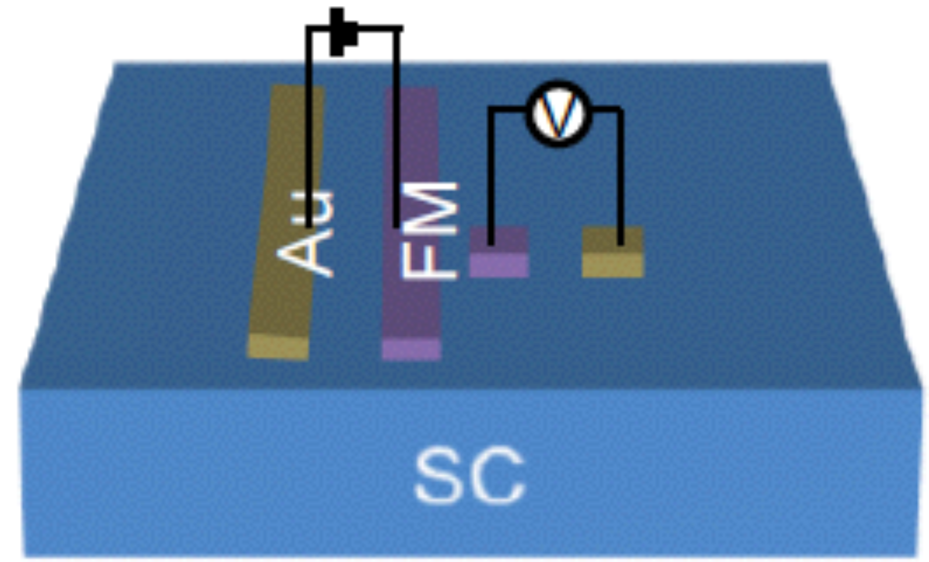


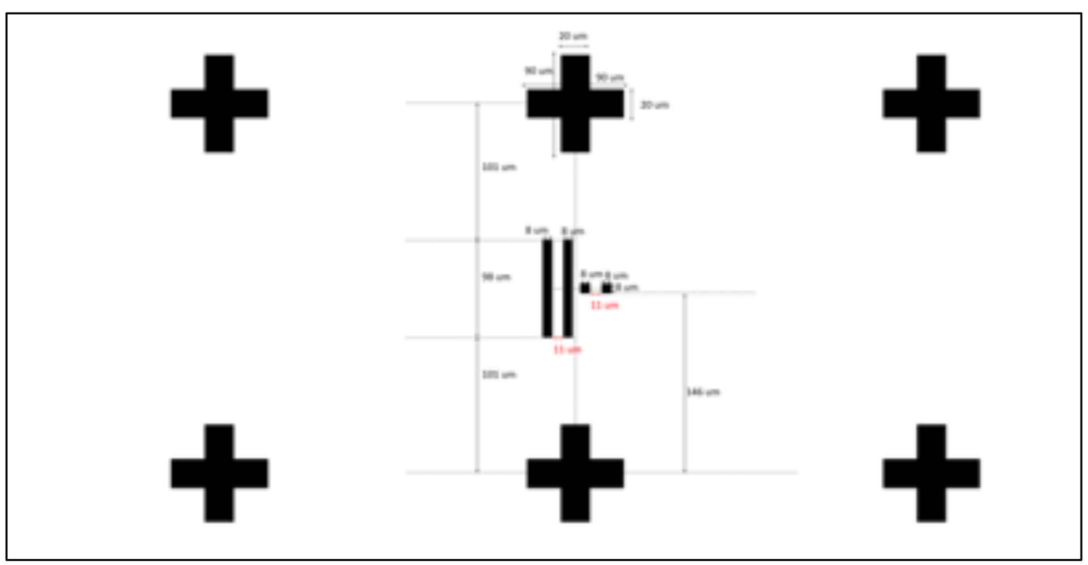
- Small device needed, in order to prevent electrode touching Sr_2RuO_4
- The area of the device should be ≲ 200 x 400 um^2

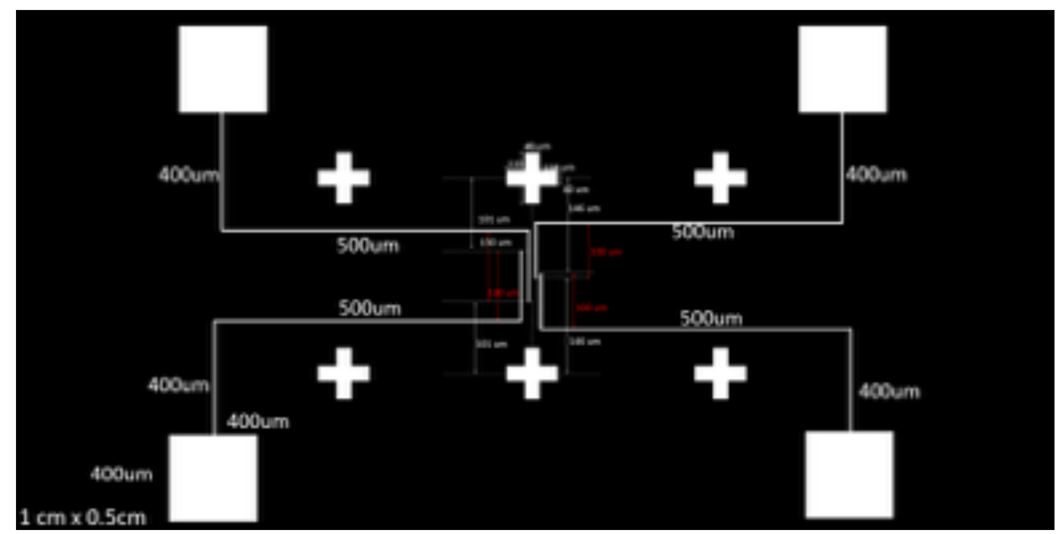
Device design: mask for non-local setup

(Ko, B Kim, **SBC**, Noh *et al* in progress)









- Designing mask for non local measurement is ongoing.

Conclusions

- Spin collective phenomena would be the signature of spin-triplet superconductivity of Sr_2RuO_4.
- Two such phenomena are the spin supercurrent and the order parameter spin collective mode.
- Both can be electrically realized in the $SrRuO_3$ / Sr_2RuO_4 junction.