Odd-Frequency Pairing in Superconducting Heterostructures

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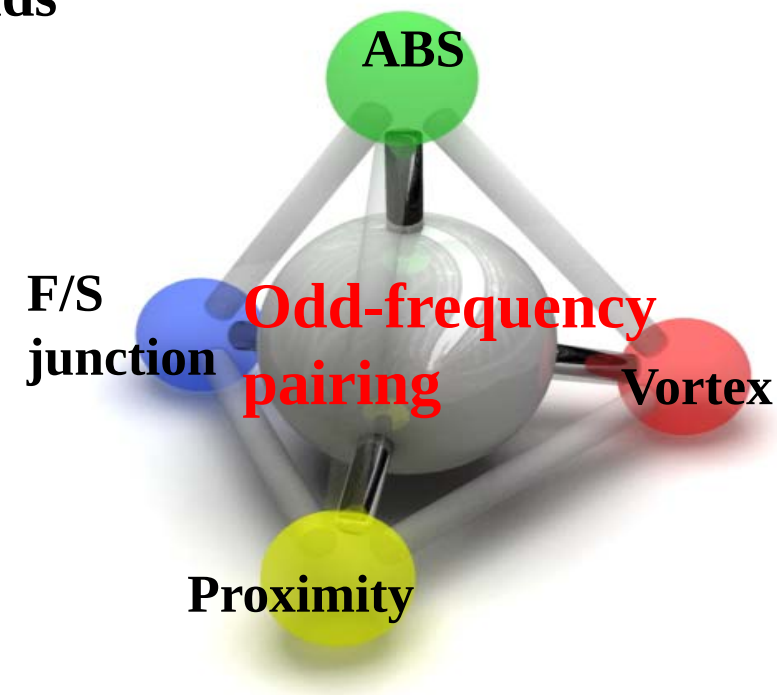

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（1）What is odd-frequency pairing

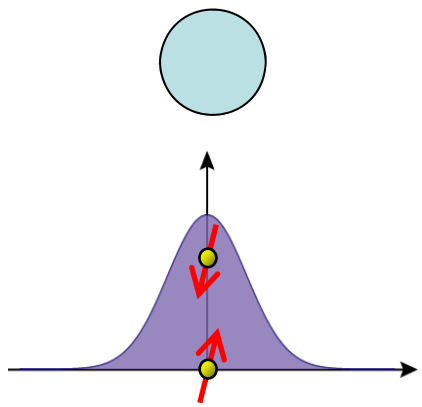
（2）Normal metal / Superconductor junctions

(3) Vortices in p-wave superconductors

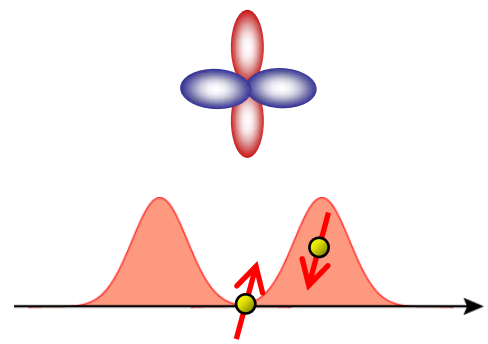
（4）Ferromagnet/Superconductor junctions

Conventional Classification of Symmetry of Cooper pair

Spin-singlet Cooper pair → **Even Parity**

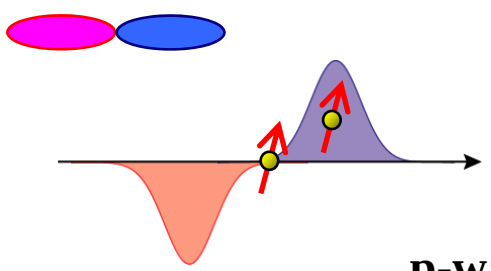


s-wave **BCS**



d-wave **Cuprate**

Spin-triplet Cooper pair → **Odd Parity**



p-wave **^{3}He** **Sr_2RuO_4**

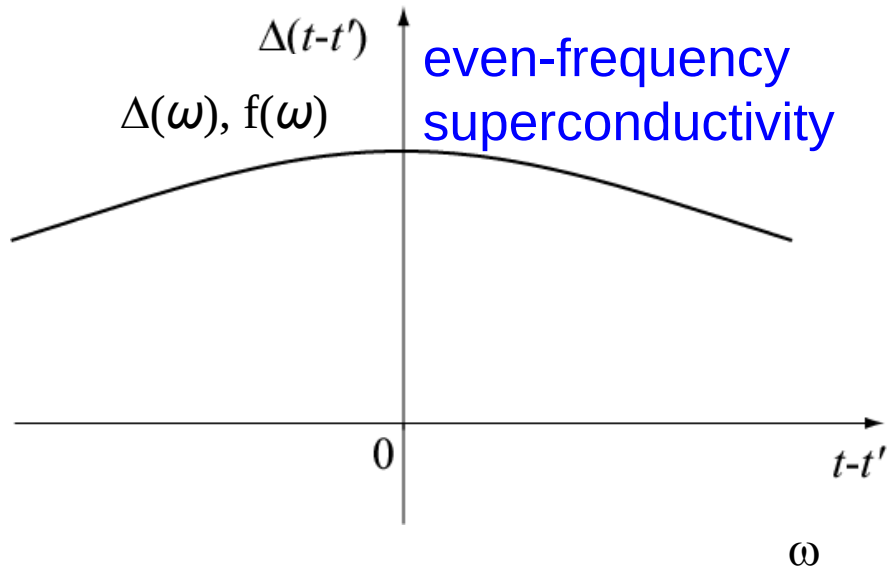
Odd-frequency pairing

Fermi-Dirac statistics

Symmetry of pair wave functions:

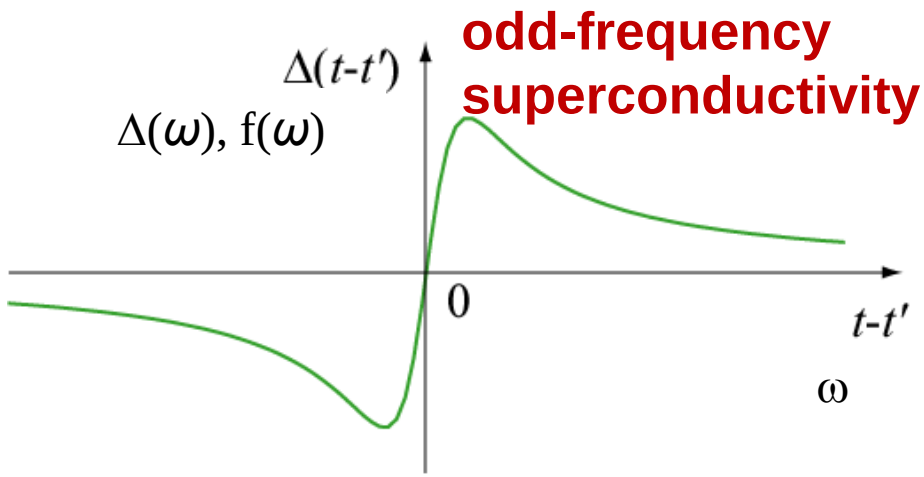
$$\mathbf{k} \otimes \sigma \otimes \omega = \mathrm{odd}$$

Momentum* x *Spin* x *Frequency



Berezinskii
(1974):
Spin-triplet s-wave

Balatsky&Abrahams
(1992):
Spin-singlet p-wave



ω, Matsubara frequency

Pair amplitude

Exchange of time

Even-frequency pairing (conventional pairing)

$$F_{\alpha,\beta}(\boldsymbol{r}_1 t_1, \boldsymbol{r}_2 t_2) = F_{\alpha,\beta}(\boldsymbol{r}_1 t_2, \boldsymbol{r}_2 t_1)$$

Odd-frequency pairing

$$F_{\alpha,\beta}(\boldsymbol{r}_1 t_1, \boldsymbol{r}_2 t_2) = -F_{\alpha,\beta}(\boldsymbol{r}_1 t_2, \boldsymbol{r}_2 t_1)$$

Symmetry of the pair amplitude

+ symmetric, – anti-symmetric

	Frequency (time)	Spin	Orbital	Total	
ESE	+(even)	– (singlet)	+(even)	–	BCS Cuprate
ETO	+(even)	+ (triplet)	–(odd)	–	^{3}He Sr_2RuO_4
OTE	–(odd)	+ (triplet)	+(even)	–	
OSO	–(odd)	– (singlet)	–(odd)	–	

ESE (Even-frequency spin-singlet even-parity)
ETO (Even-frequency spin-triplet odd-parity)
OTE (Odd-frequency spin-triplet even-parity) **Berezinskii**
OSO (Odd-frequency spin-singlet odd-parity) **Balatsky, Abrahams**

- **Odd-frequency pairing state** is possible in inhomogeneous superconductors even for conventional even-frequency paring in the bulk

Broken spin rotation symmetry or spatial invariance symmetry can induce odd-frequency pairing state:

- **ferromagnet/superconducor junctions:**

Bergeret,Volkov&Efetov, 2001

- **non-uniform systems:**

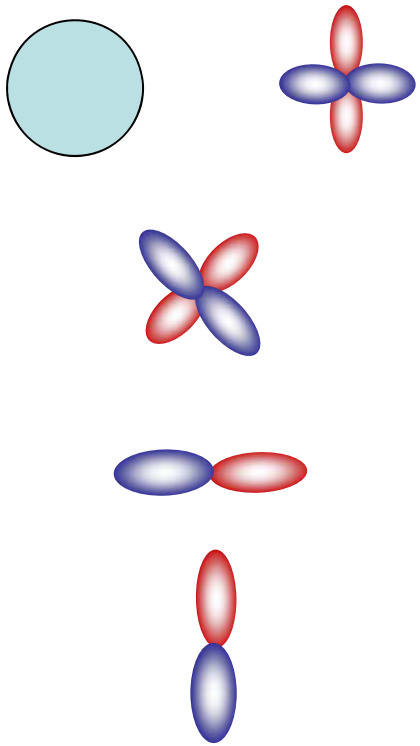
Junctions: Tanaka&Golubov, 2007; Eschrig&Lofwander, 2007

Vortices: Yokoyama *et al.*, 2008; Tanuma *et al.*, 2009)

Cooper pair symmetry near the interface

Normal metal

Even-frequency superconductor



spin-singlet

spin-singlet

spin-triplet

spin-triplet

Eilenberger Equation

$$-i\boldsymbol{v}_F \cdot \nabla \hat{g}^R = \left[\hat{H}_{qc} + \hat{\Sigma}^R, \hat{g}^R\right],$$

$$\hat{H}_{qc} = \begin{pmatrix} \varepsilon & \Delta(\hat{p}, r) \\ -\Delta^*(\hat{p}, r) & -\varepsilon \end{pmatrix}, \qquad \hat{\Sigma}^R = \frac{i}{2\tau_{imp}} < \hat{g}^R >$$

$$\hat{g}^R \hat{g}^R = \hat{1}$$

$$\boxed{\hat{\Sigma}^R = 0 \ \textbf{(ballistic)}}$$

Quasiclassical approximation

（1）Cooper pair is formed by two electrons on the Fermi surface

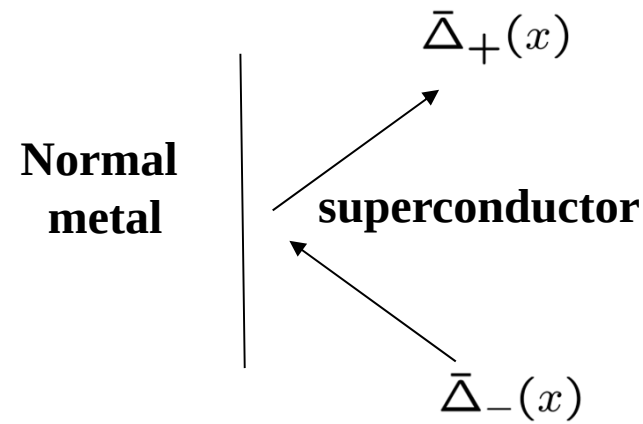
（2）The effective pair potential is determined by the direction of the motion of electrons.

Kopnin Theory of Nonequilibrium Superconductivity", Oxford University Press (2001).

Eilenberger equation

Quasiclassical Green's function (Matsubara representation)

$$\mp i v_{Fx} \partial_x f_{1\pm} = 2\omega_n f_{2\pm} - 2\bar{\Delta}_\pm(x) g_\pm$$
$$\mp i v_{Fx} \partial_x g_\pm = 2\bar{\Delta}_\pm(x) f_{1\pm},$$
$$\mp i v_{Fx} \partial_x f_{2\pm} = -2\omega_n f_{1\pm},$$



Pair potential $\bar{\Delta}_\pm(x)$

Quasiparticle $g_\pm$

Cooper pair which does not exist in bulk $f_{1\pm}$ **Odd-frequency**

Cooper pair in bulk $f_{2\pm}$ **Even-frequency**

Direction of motion $+$

Direction of motion $-$

Condition of the generation of odd-frequency pairing

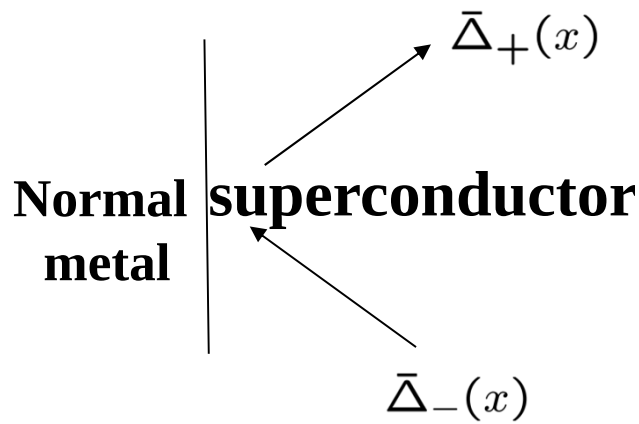
$$\mp i v_{Fx}\partial_x f_{1\pm} = 2\omega_n f_{2\pm} - 2\bar{\Delta}_\pm(x) g_\pm$$
$$\mp i v_{Fx}\partial_x g_\pm = 2\bar{\Delta}_\pm(x) f_{1\pm},$$
$$\mp i v_{Fx}\partial_x f_{2\pm} = -2\omega_n f_{1\pm},$$

$$v_{fx}^2\partial_x^2 f_{1\pm} - 4(\omega_n^2 + \bar{\Delta}_\pm^2(x)) f_{1\pm} \pm 2i \mid v_{Fx} \mid [\partial_x \bar{\Delta}_\pm(x)] g_\pm = 0$$

Spatial change of pair potential

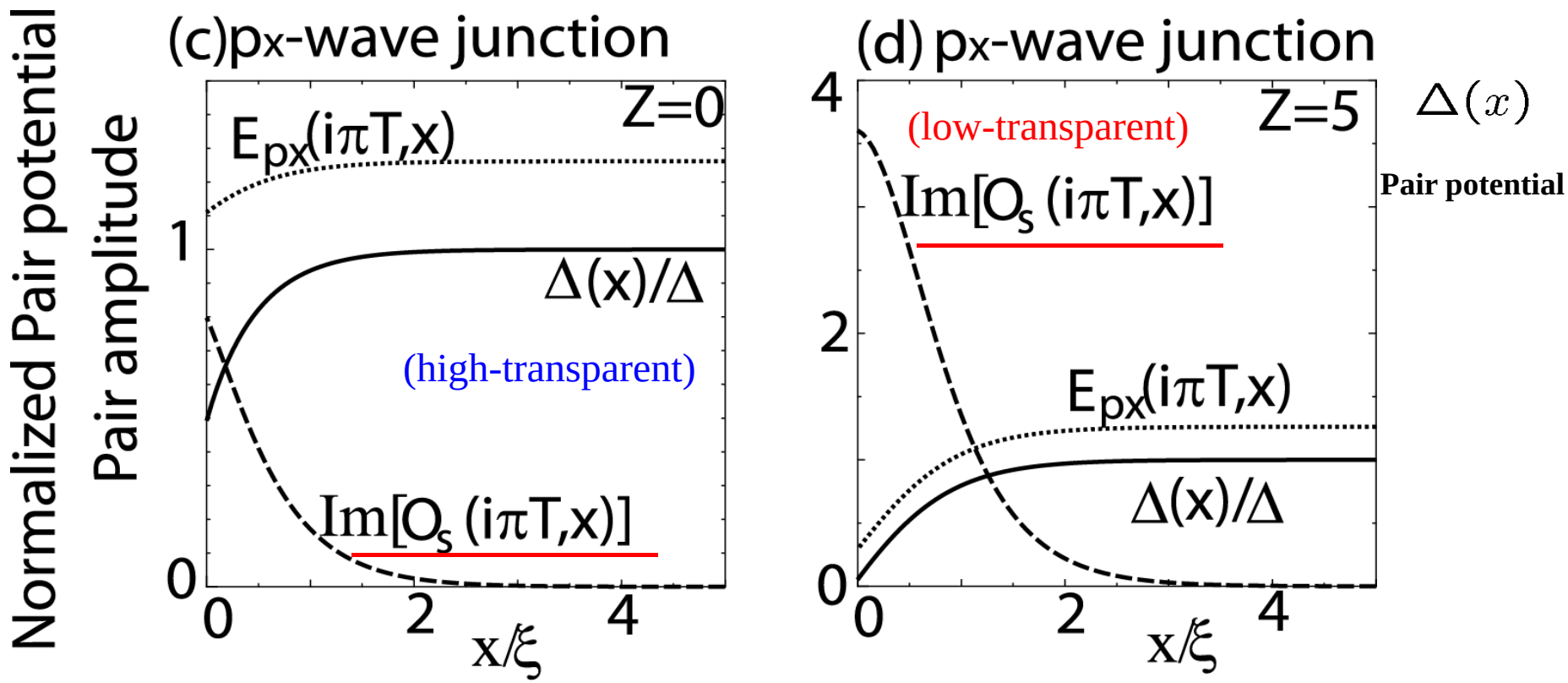
+ nonzero quasiparticle state

Odd-frequency pairing



General theory, Higashitani 2011

Symmetry of the bulk pair potential is ETO



$E_{px}(i\omega_n, x)$ **p_x-wave component of ETO pair amplitude**

$O_s(i\omega_n, x)$ **s-wave component of OTE pair amplitude**

ETO (Even-frequency spin-triplet odd-parity)

OTE (Odd-frequency spin-triplet even-parity) **Y. Tanaka, et al PRL 99 037005 (2007)**

Underlying physics

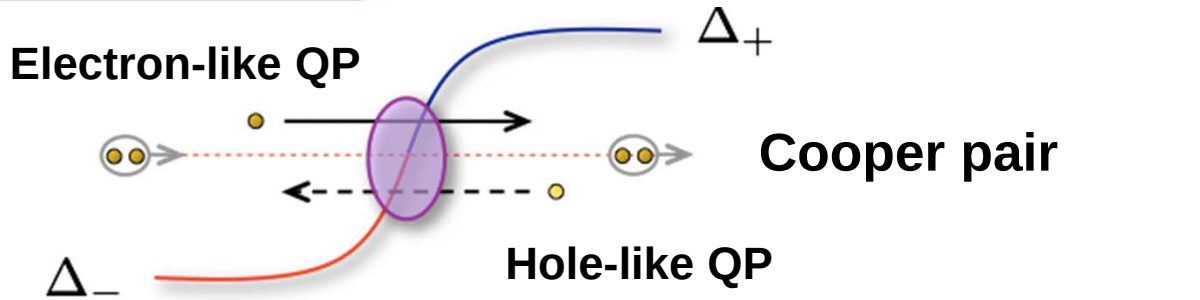
Near the interface, even and odd-parity pairing states (pair amplitude) can mix due to the **breakdown of the translational symmetry**.

⇩ **Fermi-Dirac statistics**

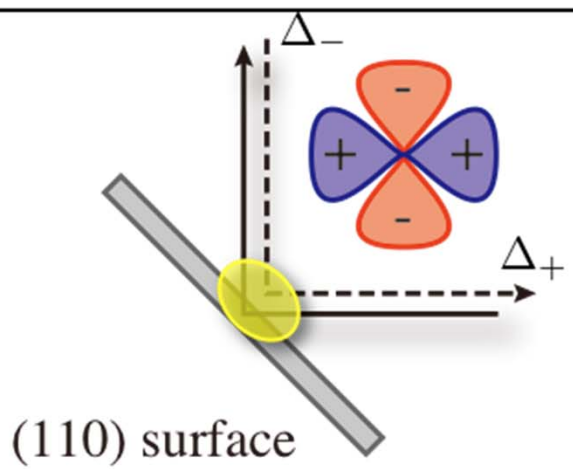
The interface-induced state (pair amplitude) should be odd in frequency where the bulk pair potential has an even -frequency component since there is no spin flip at the interface.

Andreev bound states in inhomogeneous systems are manifestations of odd-frequency pairing amplitude

Andreev bound states

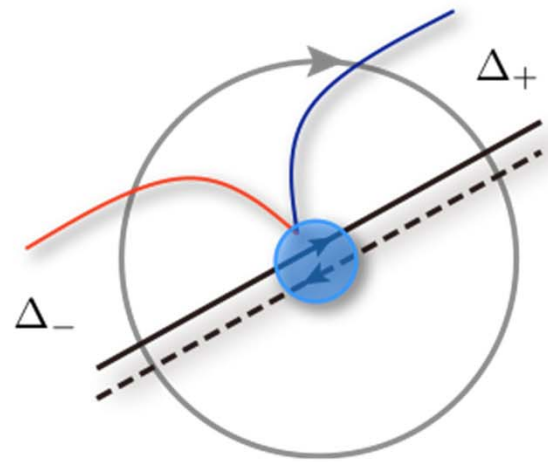


Surface: Tanaka *et al*, 2007



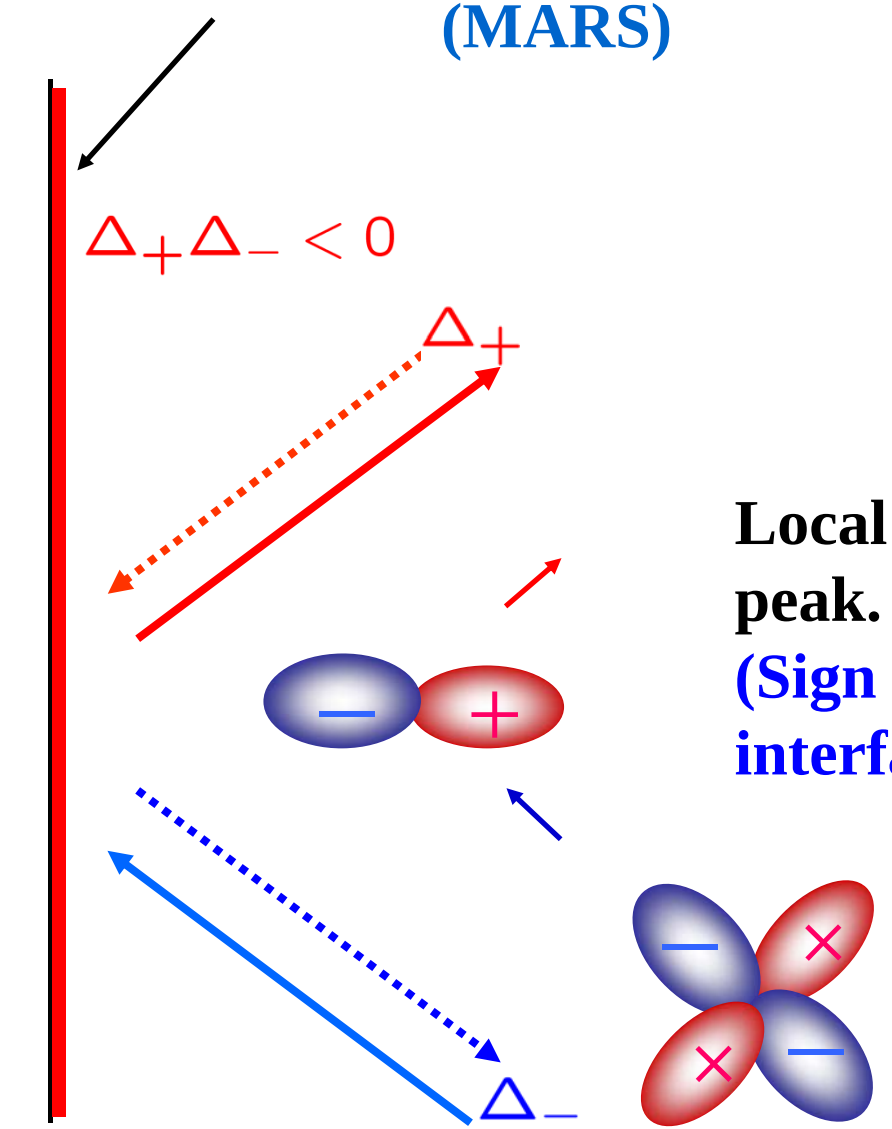
Scattering direction of QP

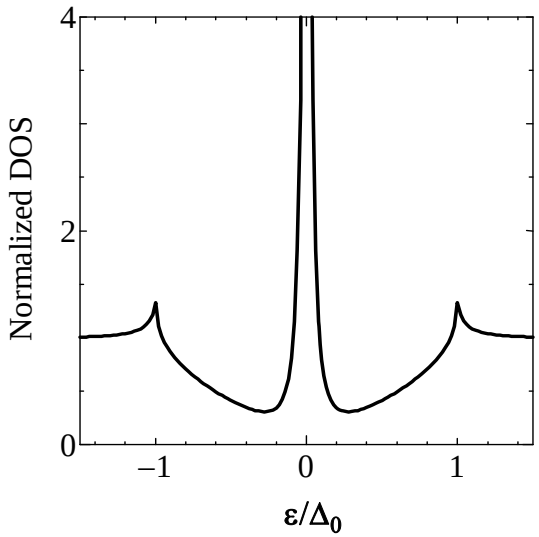
Vortex : Tanuma *et al*, 2009



Phase change due to a vortex

Mid gap Andreev resonant (bound) state (MARS)



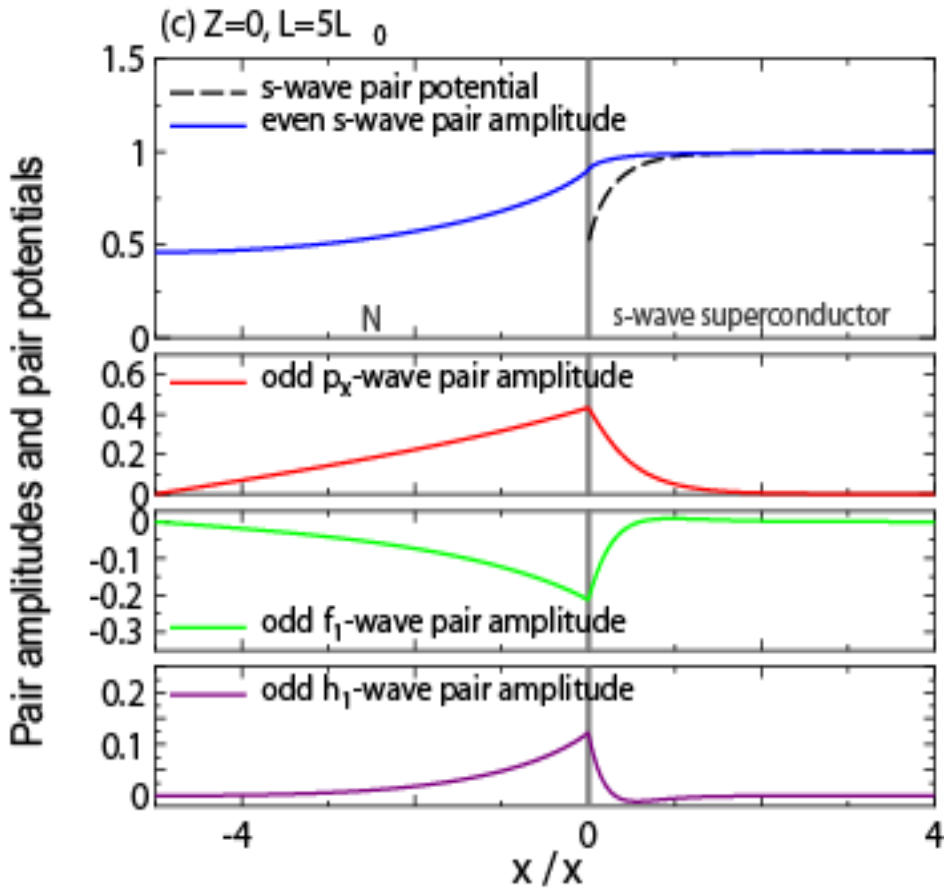


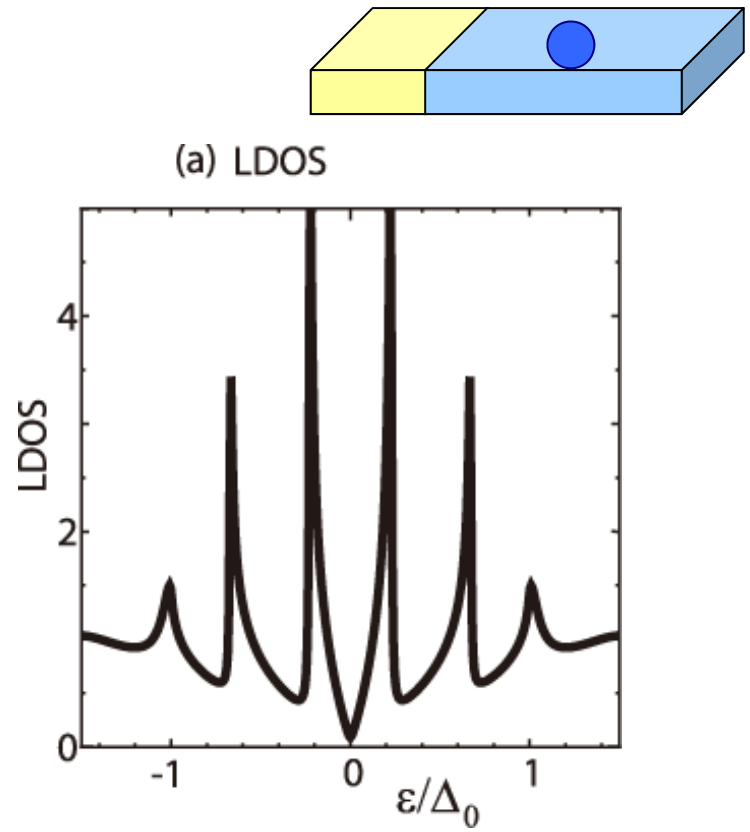
Local density of state has a zero energy peak.

(Sign change of the pair potential at the interface)

Tanaka Kashiwaya PRL 74 3451 (1995),
Rep. Prog. Phys. 63 1641 (2000)
Buchholz(1981) Hara Nagai(1986)
Hu(1994) Matsumoto Shiba(1995)
Ohashi Takada(1995)
Hatsugai and Ryu (2002)

Odd-frequency pairing state in N/S junctions (N finite length)





Bounds state are formed in the normal metal

Y. Tanaka, Y. Tanuma and A.A.Golubov, Phys. Rev. B **76**, 054522 (2007)

Ratio of the pair amplitude at the N/S interface and the bound state level

$\Delta_0 \gg \varepsilon$ **Bound states condition (Z=0)**

(McMillan Thomas Rowell)

$$\varepsilon_n = \frac{\pi v_{Fx}}{2L}(n + 1/2), \quad n = 0, 1, 2, \ldots$$

$$\frac{\left|f_{1+}^{(N)}(\varepsilon,\theta)\right|}{\left|f_{2+}^{(N)}(\varepsilon,\theta)\right|} = |\tan(\pi/2 + \pi n)| = \infty.$$

$f_{1+}^{(N)}(\varepsilon,\theta)$

Odd-frequency pairing

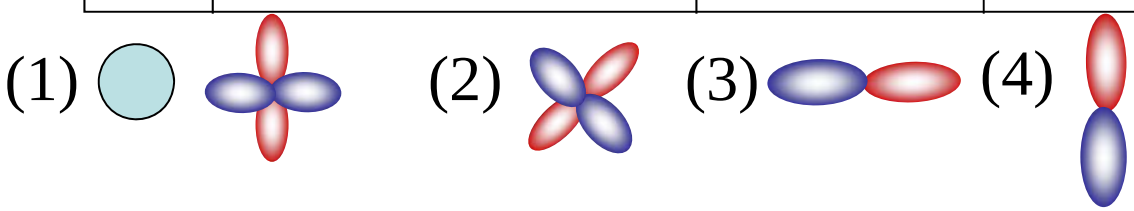
$f_{2+}^{(N)}(\varepsilon,\theta)$

Even-frequency pairing

Bound states are due to the generation of the odd-frequency Cooper pair amplitude

Symmetry of the Cooper pair (No spin flip)

	Bulk state	Sign change **(MARS)**	Interface-induced symmetry (subdominant component)
(1)	ESE (s,d_{x2-y2} -wave)	No	**ESE** + (OSO)
(2)	ESE (d_{xy}-wave)	Yes	**OSO** +(ESE)
(3)	ETO (p_x-wave)	Yes	**OTE** + (ETO)
(4)	ETO (p_y-wave)	No	**ETO** + (OTE)

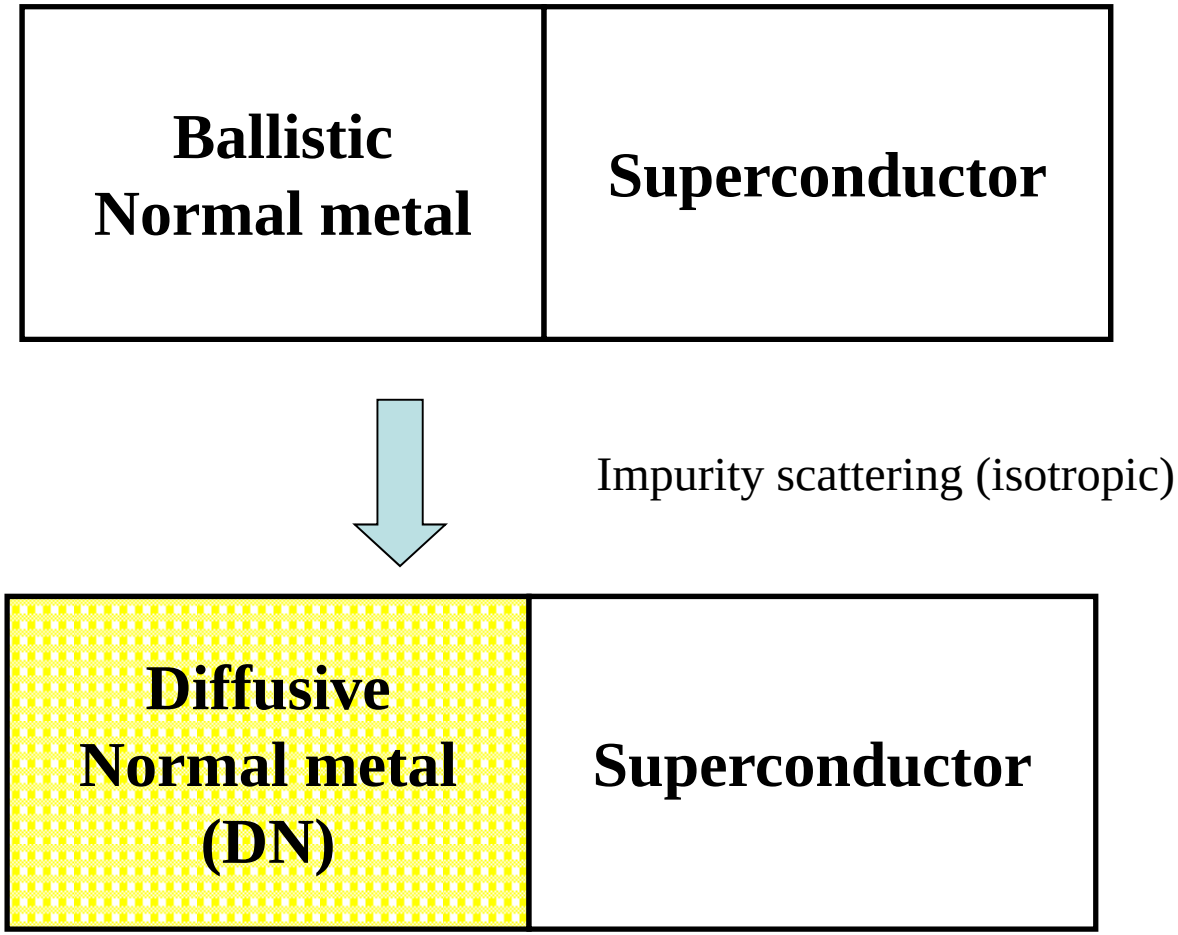


- **ESE (Even-frequency spin-singlet even-parity)**
- **ETO (Even-frequency spin-triplet odd-parity)**
- **OTE (Odd-frequency spin-triplet even-parity)**
- **OSO (Odd-frequency spin-singlet odd-parity)**

Phys. Rev. Lett. 99 037005 (2007)

Impurity scattering effect

Tanaka and Golubov, PRL. 98, 037003 (2007)



Only s-wave pair amplitude exists in DN

(1)ESE
(2)OTE

ESE (Even-frequency spin-singlet even-parity

Usadel equation

Available for diffusive limit $\tau_{imp} T << 1$

$$D\nabla(\hat{g}_0^R \nabla \hat{g}_0^R) + i[\hat{H}_0, \hat{g}_0^R] = 0$$

$$\hat{g}_0^R \Longleftarrow \hat{g}^R$$

Diffusive limit

Angular average

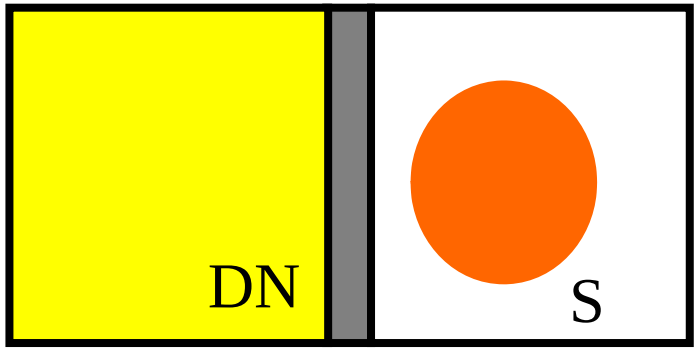
Diffusive normal metal region attached to superconductor

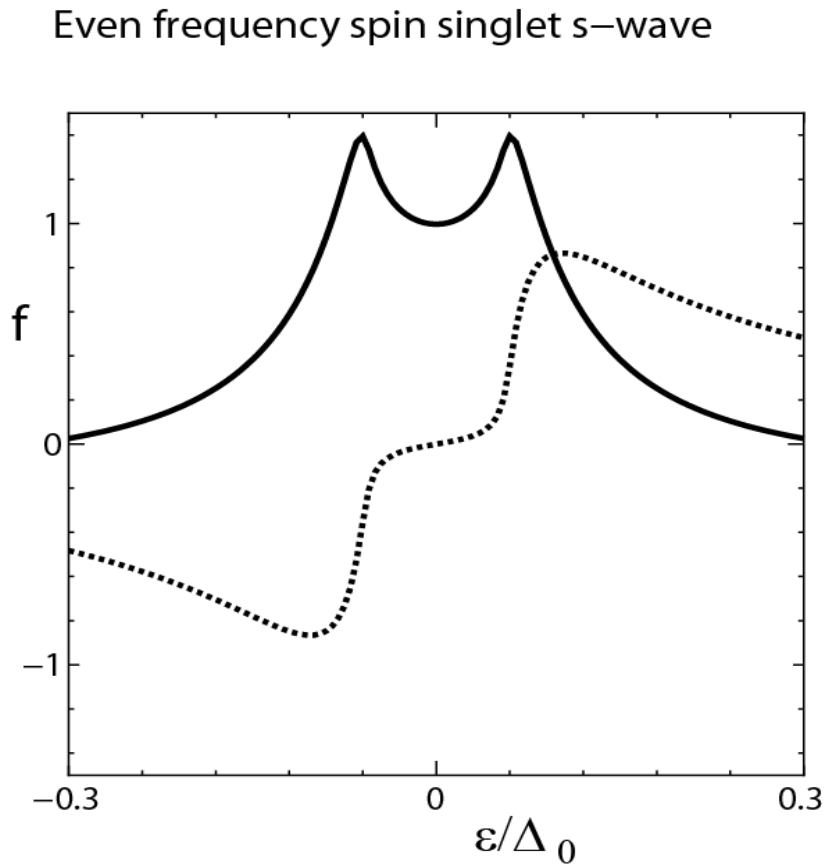
$$\hat{H}_0 = \varepsilon \hat{\tau}_3$$

Boundary condition available for unconventional superconductors

Tanaka et al, PRL 90 167003 (2003), PRB 70 012507 (2004)

Even frequency spin singlet even parity (ESE) pair potential





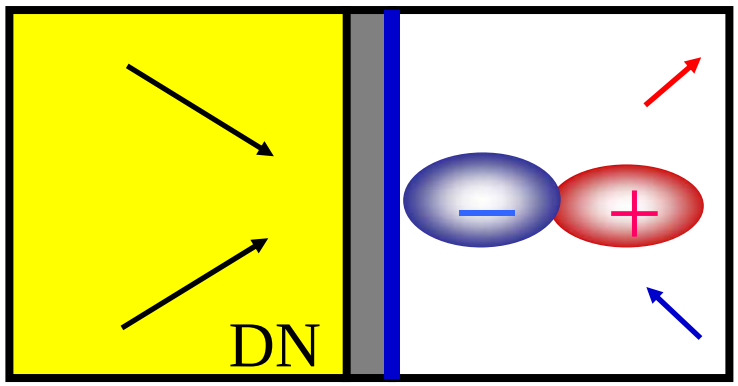
$$\mathrm{Real}[f(\varepsilon)] = \mathrm{Real}[f(-\varepsilon)]$$
$$\mathrm{Imag}[f(\varepsilon)] = -\mathrm{Imag}[f(-\varepsilon)]$$

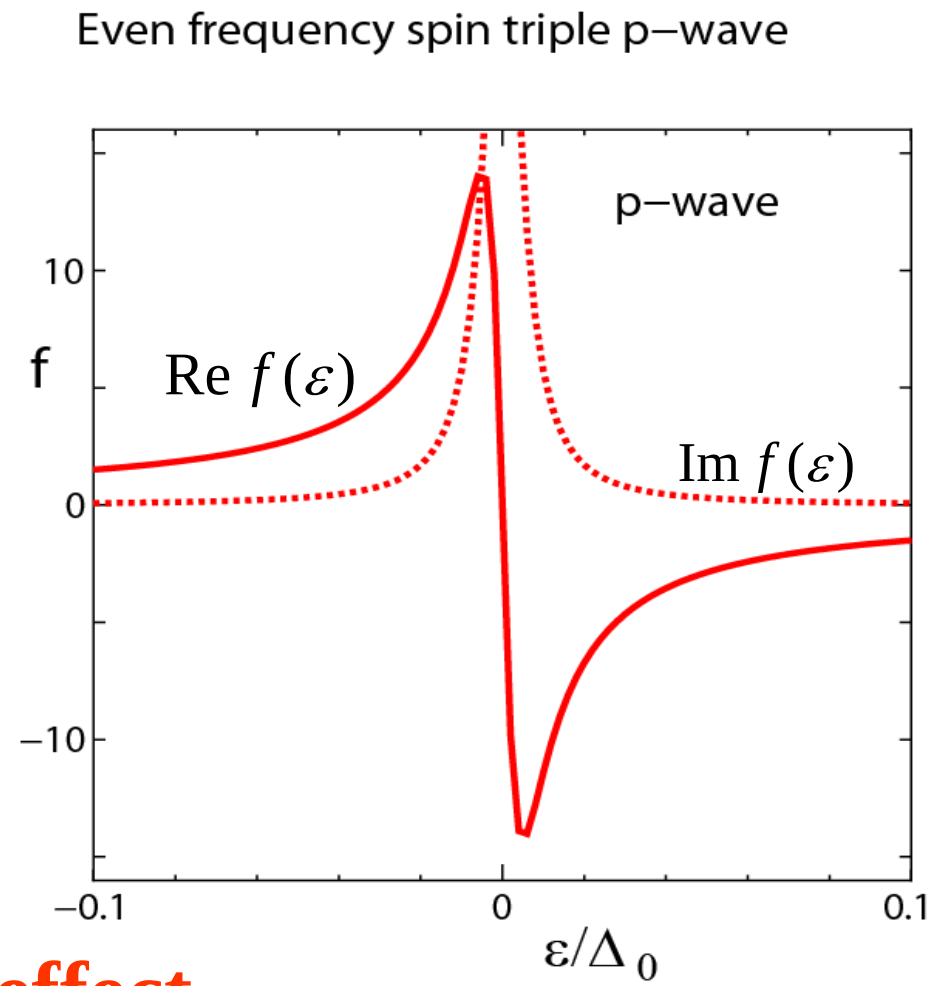
Even frequency spin singlet s-wave (ESE) pair is induced in DN.

ESE pair /ESE pair potential

Odd frequency spin triplet s-wave (OTE) pair is induced in DN

P_x-wave case

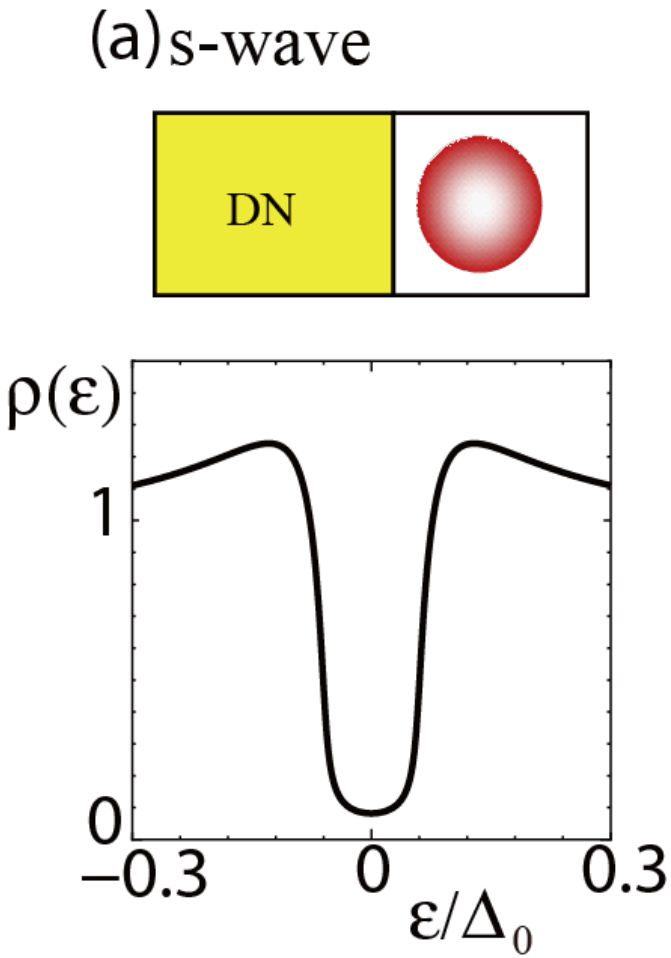


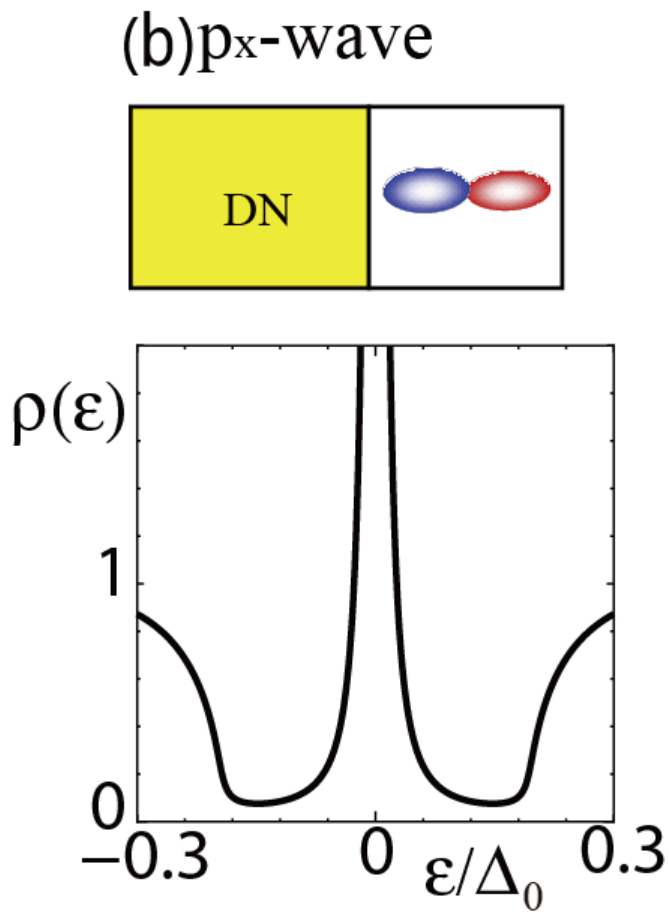


New type of proximity effect

Y.Tanaka, A.A.Golubov, Phys.Rev.Lett. **98**, 037003 (2007)

Density of states in DN





Conventional proximity effect with Even-frequency Cooper pair in DN

Unconventional proximity effect with Odd-frequency Cooper pair in DN

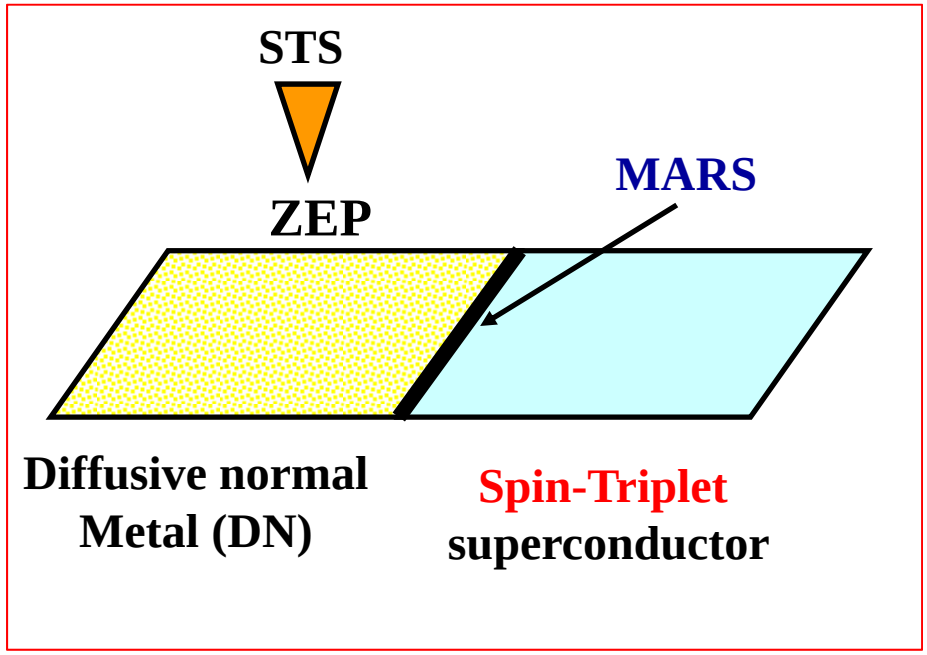
Tanaka&Kashiwaya, 2004

Summary of Proximity effect
(diffusive normal metal, *s-wave* pairing state only)

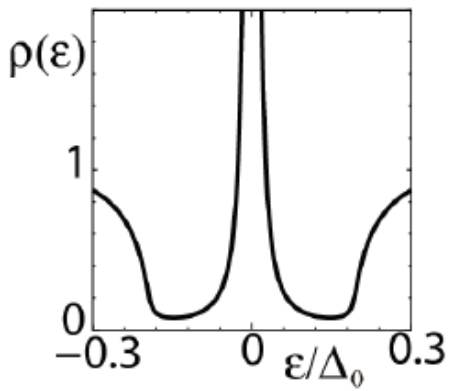
	Symmetry of the pair potential	Induced pair amplitude in DN
(1)	**Even frequency spin singlet even parity (ESE)**	**ESE**
(2)	**Even frequency spin triplet odd parity (ETO)**	**OTE**
(3)	**Odd frequency spin triplet even parity (OTE)**	**OTE**
(4)	**Odd frequency spin singlet odd parity (OSO)**	**ESE**

STS experiments

Proximity effect via odd-frequency pairing

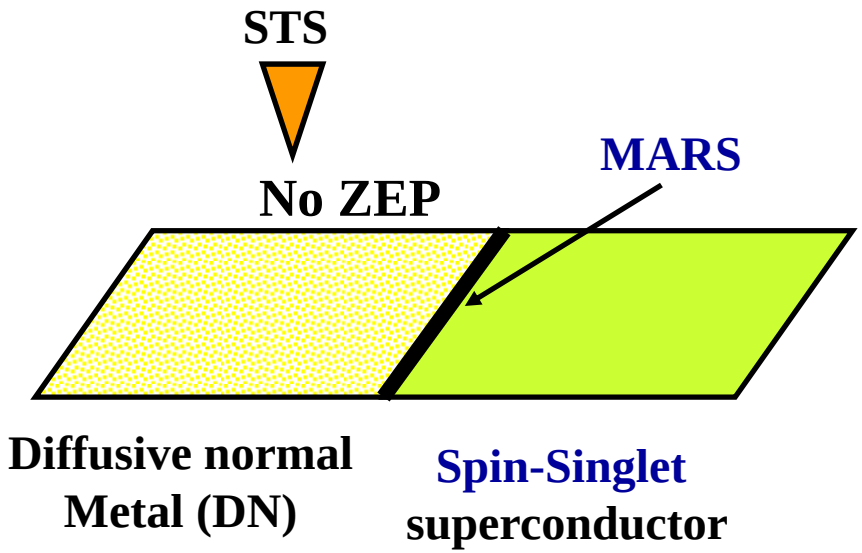


LDOS in DN has a zero energy peak

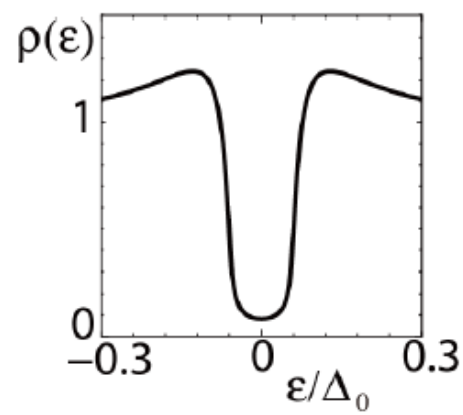


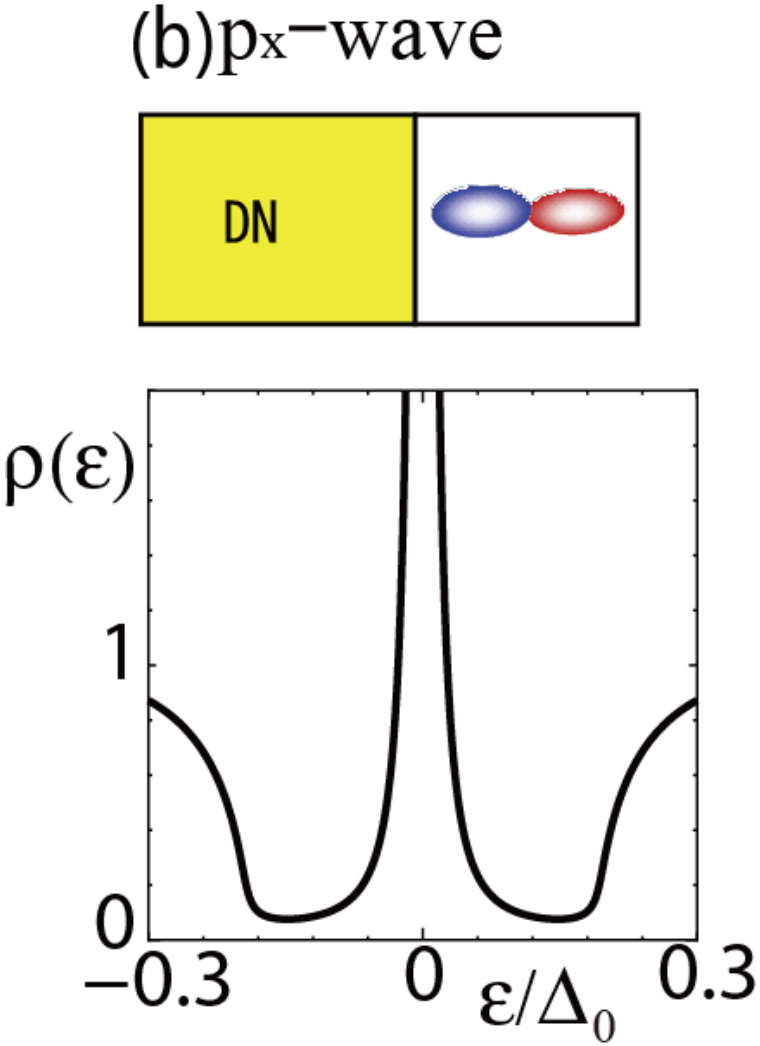
DoS peak is robust again imputiry satterin

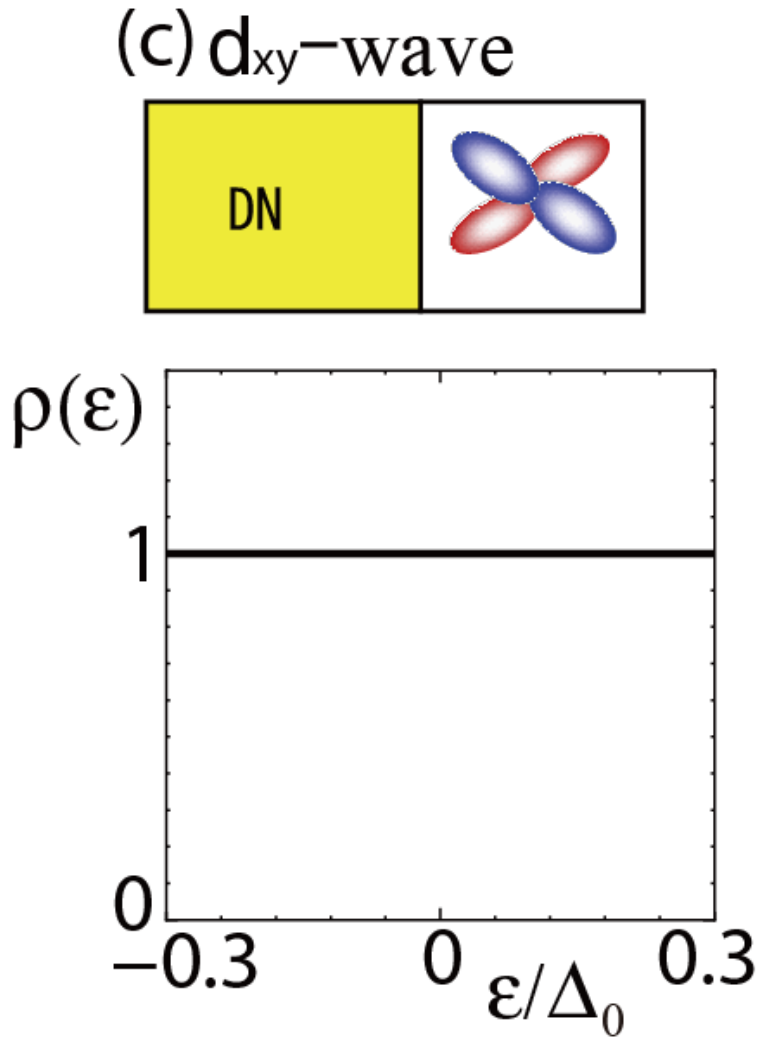
Conventional proximity effect



LDOS in DN has a gap







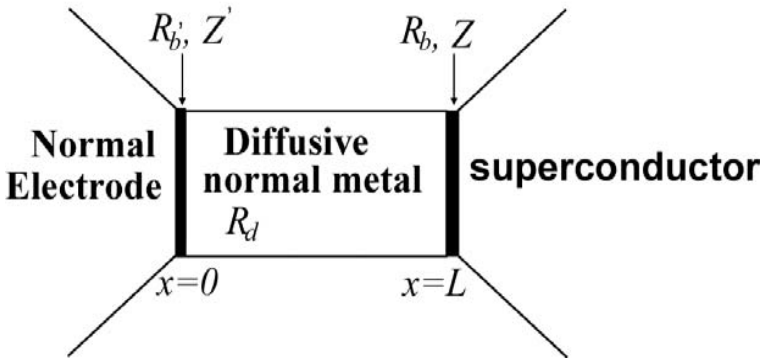
Unconventional proximity effect

No proximity effect

Odd-frequency pairing at the interface includes s-wave component

Odd-frequency pairing at the interface: Odd-parity

Anomalous Meissner effect



$$\hat{R}_N(x) = \sin\theta\hat{\tau}_2 + \cos\theta\hat{\tau}_3$$

$$j(x) = \pi e^2 N(0) D T \sum_{\omega_n} \mathrm{Trace}[\hat{\tau}_3 \hat{R}_N(x)[\hat{\tau}_3, \hat{R}_N(x)]] A(x)$$

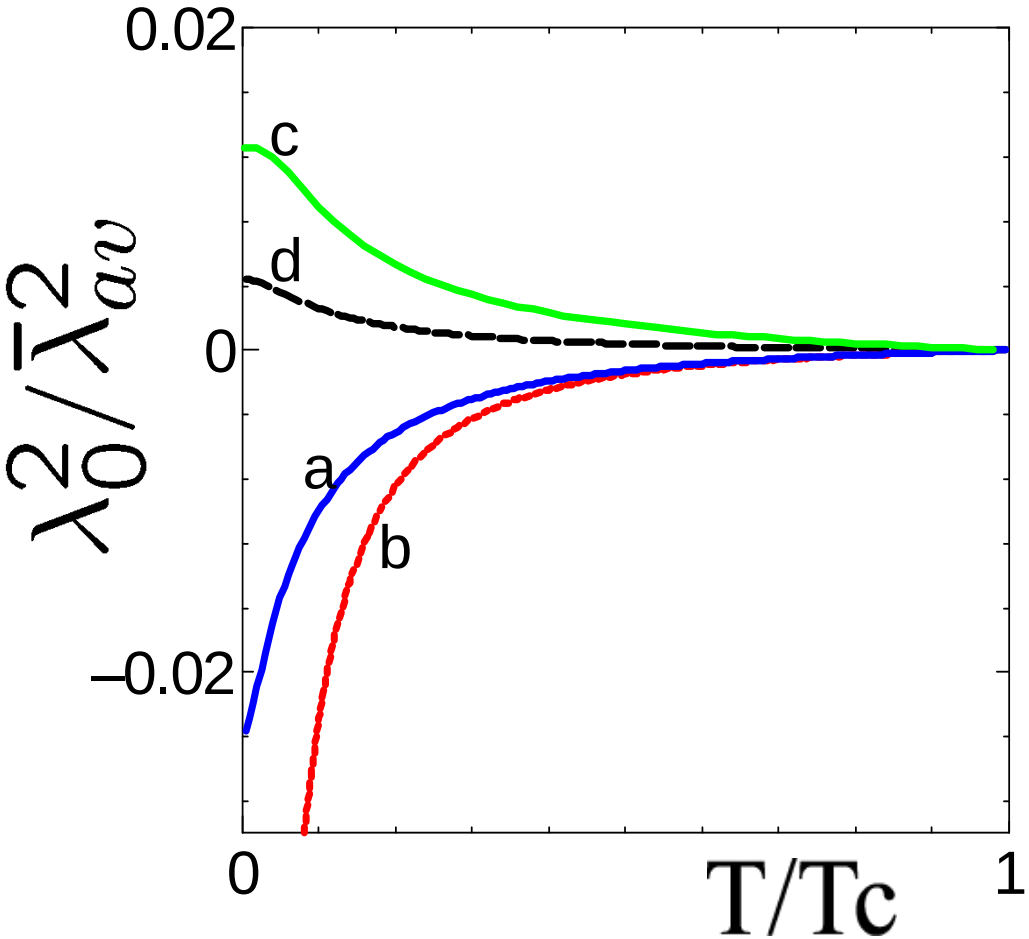
$$H(x) \sim \exp(-x/\lambda(x))$$

$$\frac{1}{\lambda^2(x)} = \frac{T\sum_{\omega_n} \sin^2\theta(\omega_n)}{\lambda_0^2}, \quad \lambda_0^{-2} = 32\pi^2 e^2 N(0) D T_C$$

$$\bar{\lambda}_{av}^2 = L / \int_0^L \frac{dx}{\lambda^2(x)}$$

Narikiyo and Fukuyama, J. Phys. Soc. Jpn. 58, 4557 (1989)
Belzig and Bruder PRB 53 5727 (1996)

Temperature dependence of averaged value of local penetration depth



$\bar{\lambda}_{av}$

a purely imaginary number for spin-triplet junctions

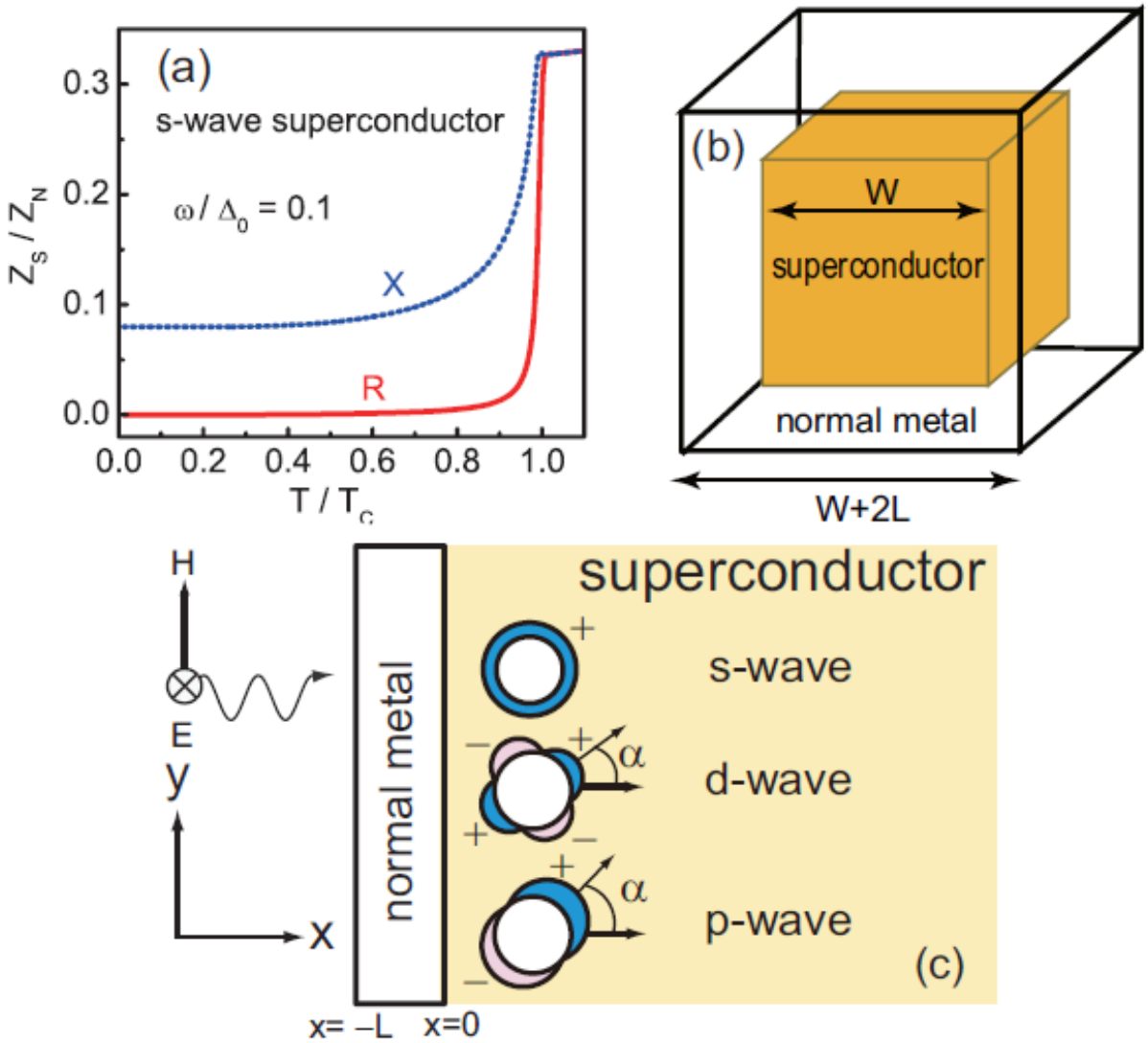
a: $p_x + ip_y$-wave

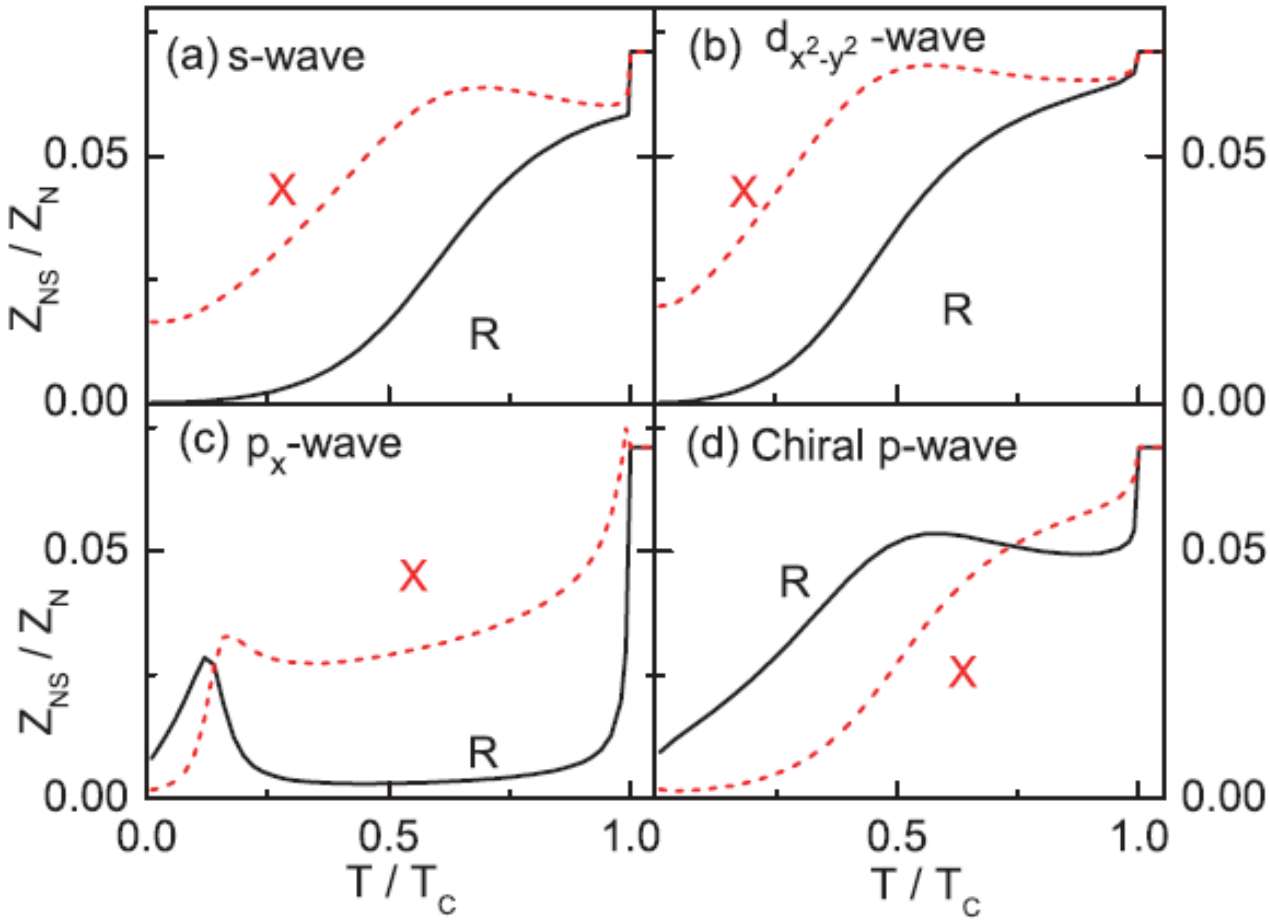
b: p_x-wave

c: s-wave

d: $d_{x^2-y^2} + id_{xy}$-wave

How to detect anomalous Meissner effect?
Surface impedance





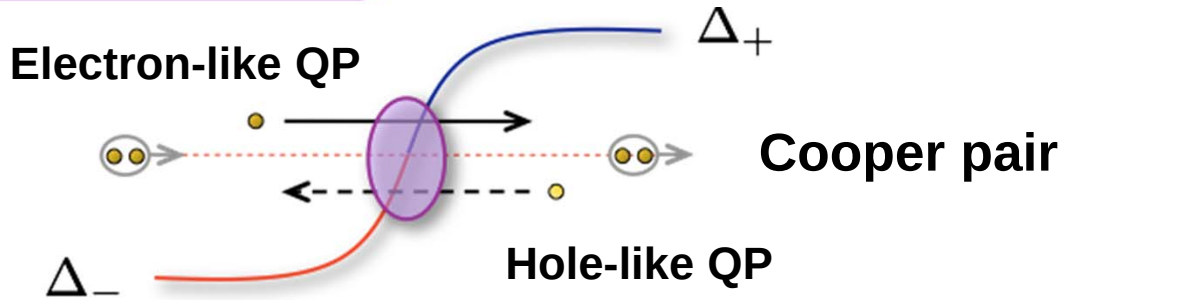
Robust relation R<X holds in an even-frequency state

This relation violates in an odd-frequency state at low T

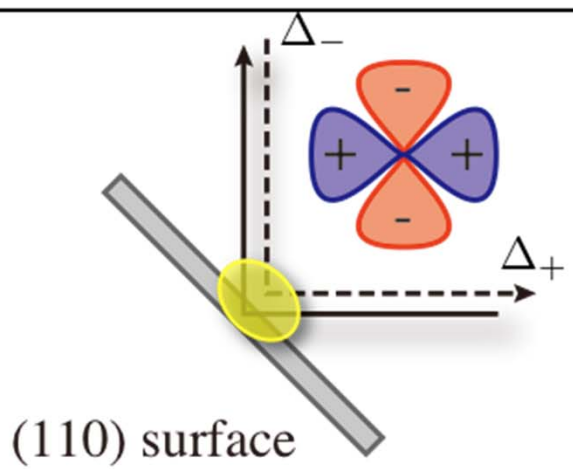
Y.Asano, A.A.Golubov, Ya.V.Fominov, and Y. Tanaka, PRL (2011)

Andreev bound states in inhomogeneous systems are manifestations of odd-frequency pairing amplitude

Andreev bound states

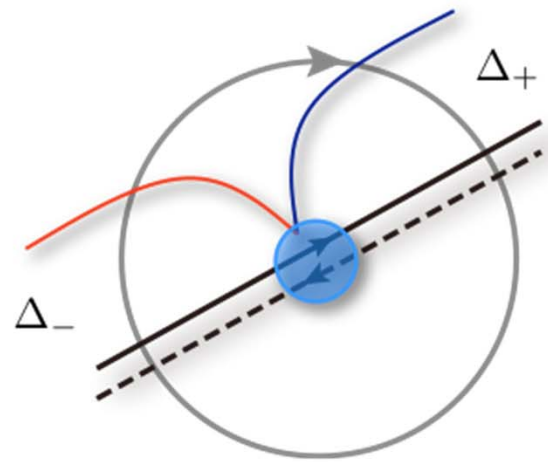


Surface: Tanaka *et al*, 2007



Scattering direction of QP

Vortex : Tanuma *et al*, 2009

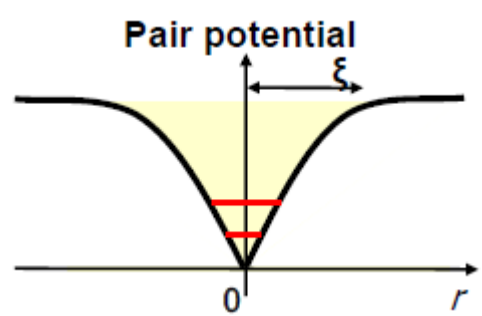


Phase change due to a vortex

Symmetry of the Cooper pair in a vortex core

l; **angular momentum**	*m*; **vorticity**	bulk	Center of the vortex core
Even	Even	ESE (s-wave..)	ESE
Even	Odd	ESE (s-wave..)	**OSO**
Odd	Even	ETO (chiral p-wave)	ETO
Odd	Odd	ETO (chiral p-wave)	**OTE**

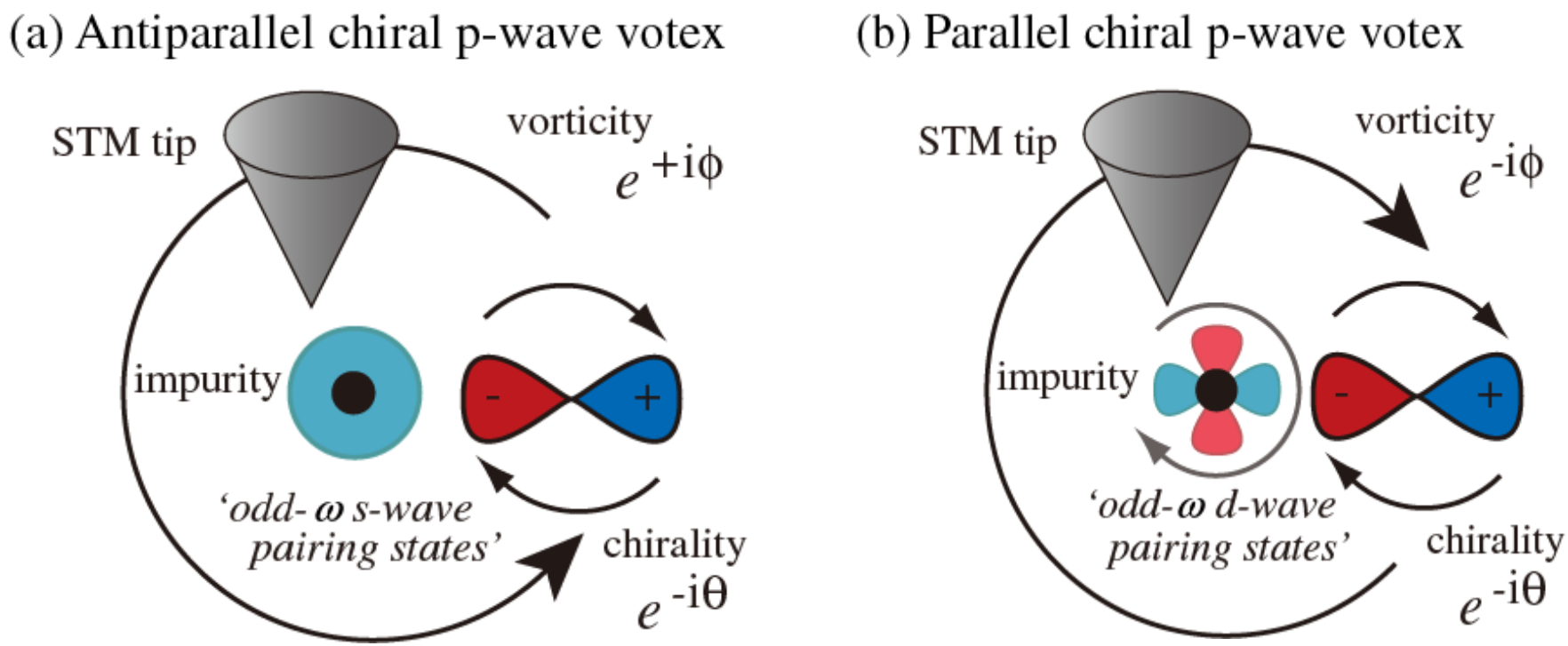
$$\Delta(\mathbf{r}) = \Delta_0 \exp(il\varphi) \tanh\left(\frac{\sqrt{x^2+y^2}}{\xi}\right)\left(\frac{x+iy}{\sqrt{x^2+y^2}}\right)^m$$



ESE (Even-frequency spin-singlet even-parity)
ETO (Even-frequency spin-triplet odd-parity)
OTE (Odd-frequency spin-triplet even-parity)
OSO (Odd-frequency spin-singlet odd-parity

Yokoyama *et al.*, Physical Review B, Vol. 78, 012508, 2008

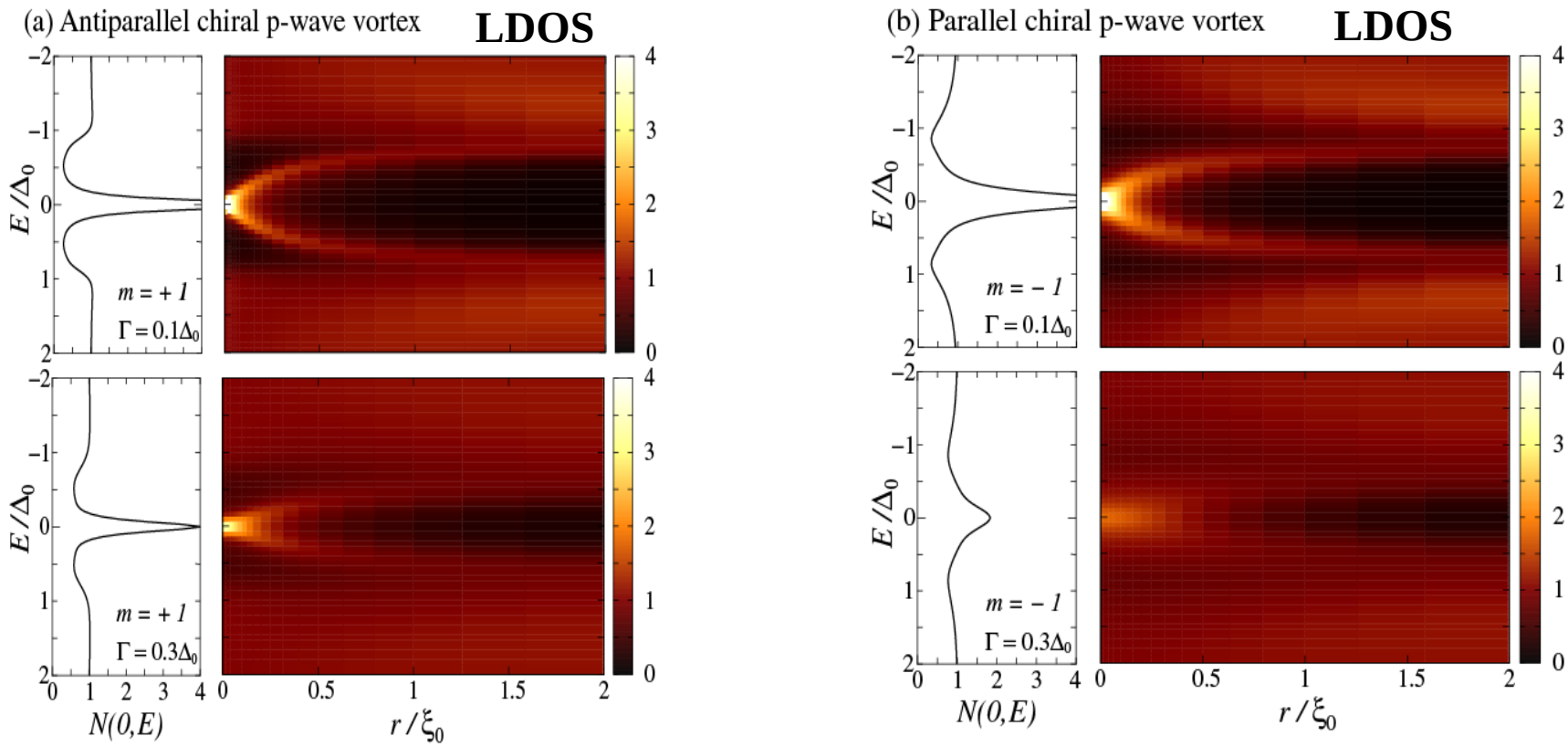
Difference of the angular momentum of the odd-frequency pair at the core center



Angular momentum at the center of core; *l+m*

***l*: angular momentum** ***m*: vorticity**

Impurity effect (Born approximation)



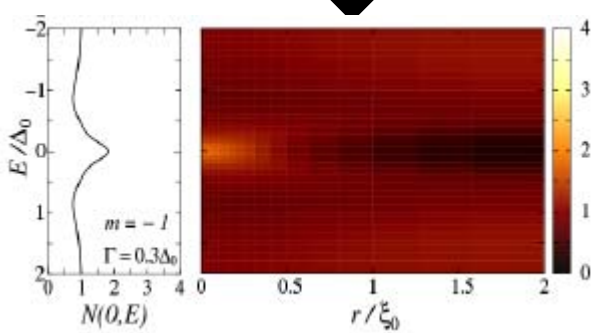
Γ, impurity scattering strength

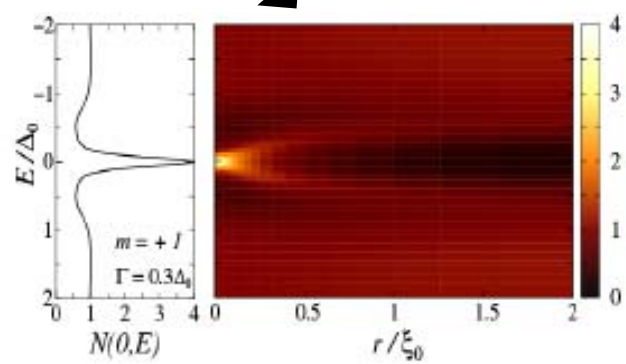
Odd-frequency s-wave **Odd-frequency d-wave**

Odd frequency s-wave state; more robust against the impurity scattering **Tanuma, Hayashi, Tanaka Golubov, PRL 102 117003 (2009).**

Vortex core spectroscopy in chiral domain

Parallel vortex	Antiparallel vortex	Parallel vortex	Antiparallel vortex
Odd d-wave	Odd s-wave	Odd d-wave	Odd s-wave
$p_x + ip_y$	$p_x - ip_y$	$p_x + ip_y$	$p_x - ip_y$
Weak ZEP	Strong ZEP	Weak ZEP	Strong ZEP





Summary (vortices)

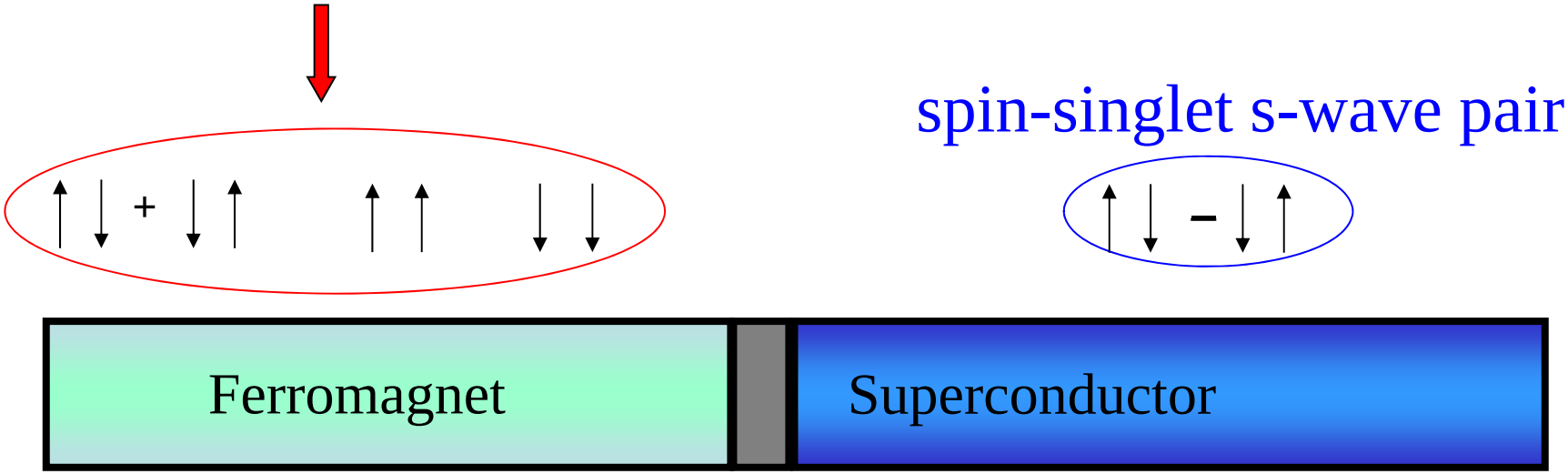
(1)In Abrikosov vortex (m=1), only the odd-frequency Cooper pairing is possible at the center of the vortex core.

(2)Vortex core spectroscopy in chiral p-wave superconductor in the presence of impurity enables one to identify the presence of chirality and the odd-frequency pairing.

Odd-frequency pair amplitude (not pair potential) is generated in ferromagnet junctions

Odd frequency spin-triplet s-wave pair

spin-singlet s-wave pair



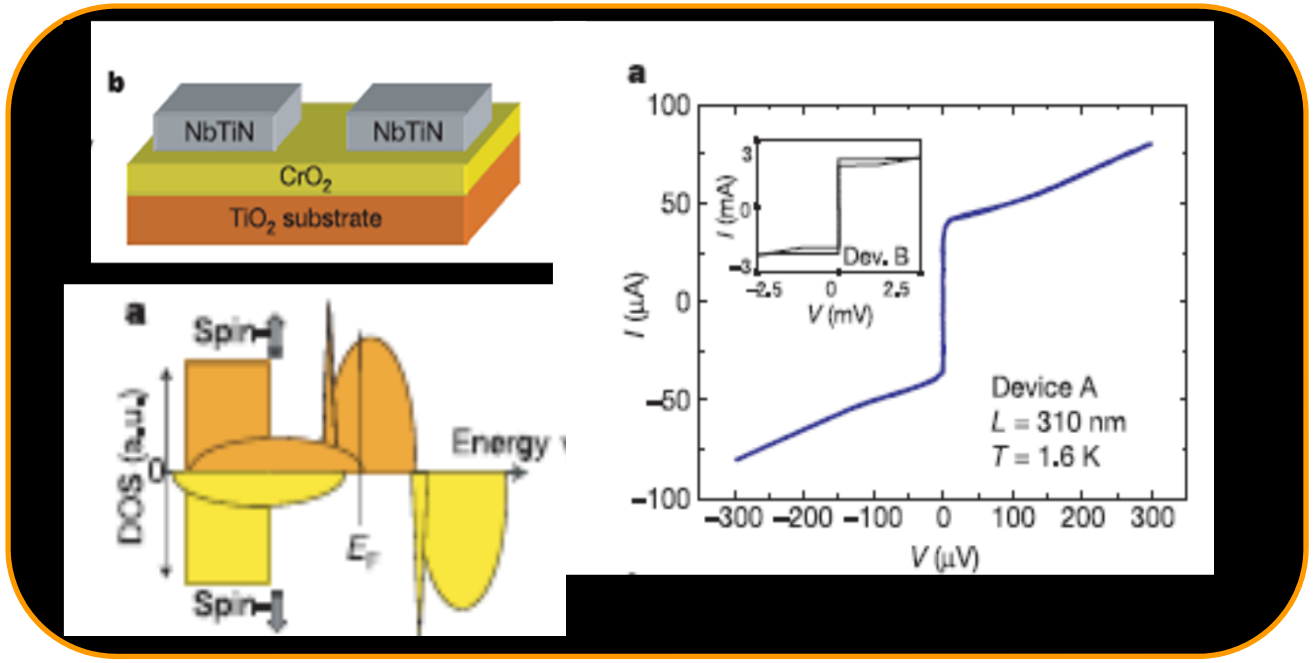
Bergeret, Efetov, Volkov, (2001)

Eschrig, Buzdin, Kadigrobov,Fominov, Radovic

Generation of the odd-frequency pair amplitude in ferromagnet

Josephson current in S/HM/S

Half metal : CrO_2 Keizer *et.al*., Nature (2006)



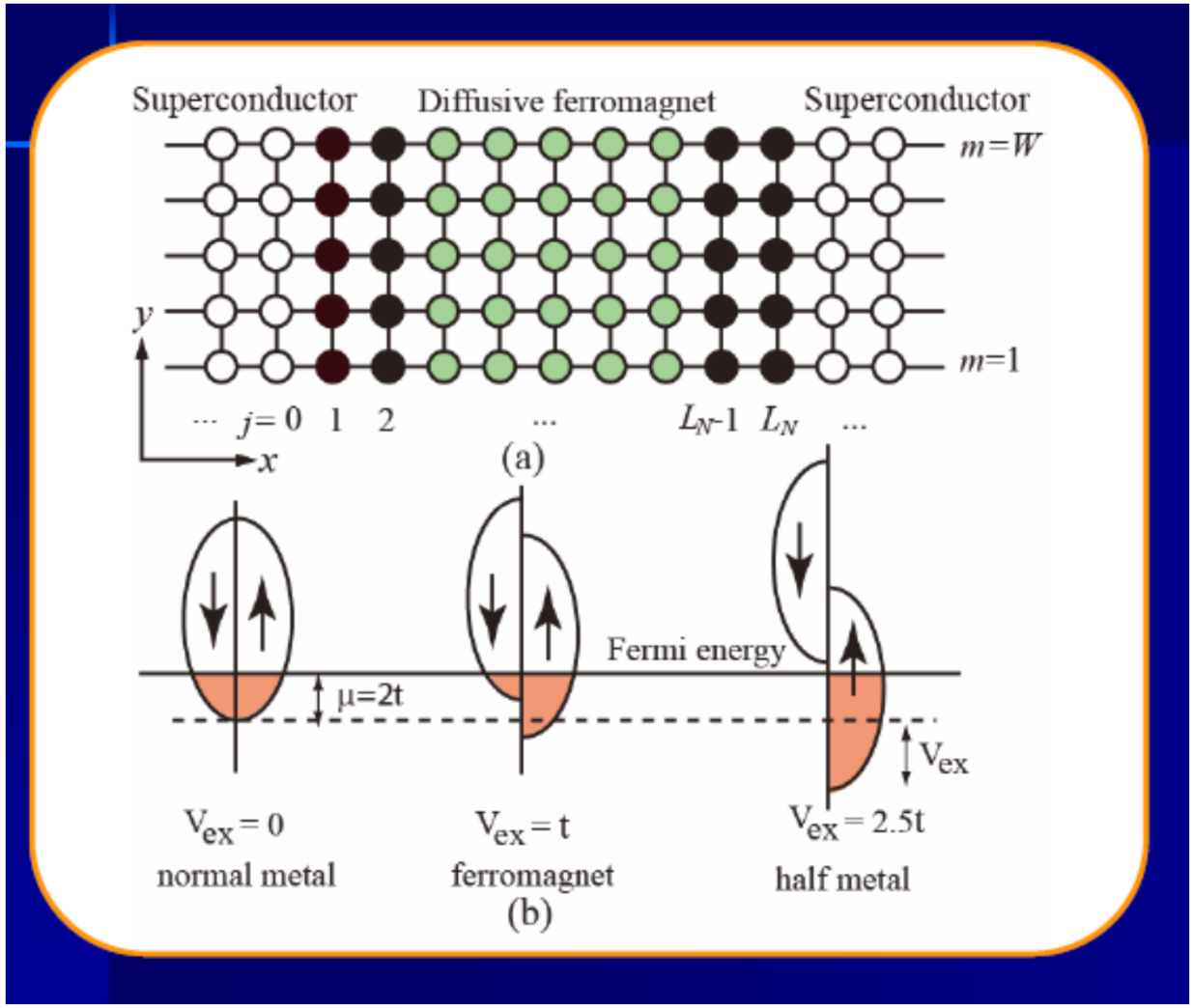
Spin active interface Bergeret et. al., PRL('01), Kadigrobov et. al., Europhys Lett.('01)

Theory in the clean limit Eschrig et. al., PRL('03)

Lofwander and Eschrig, Nature Physics (2008)

Recursive GF

Furusaki, Physica B('92),
Asano, PRB('01)



Advantages

SNS, SFS, S/HM/S

$$\langle J \rangle = \sum_{i=1}^{N_S} J_i$$

$$\delta J = \sqrt{\langle J^2 \rangle - \langle J \rangle^2}$$

Parameters

V_{ex} : exchange

V_S : spin-flip

Y.Asano, Y. Tanaka, A.A.Golubov, Phys.Rev.Lett. **98,** 107002 (2007)

Quasiparticle DOS in Half Metal

SFS $\langle f_0 \rangle$

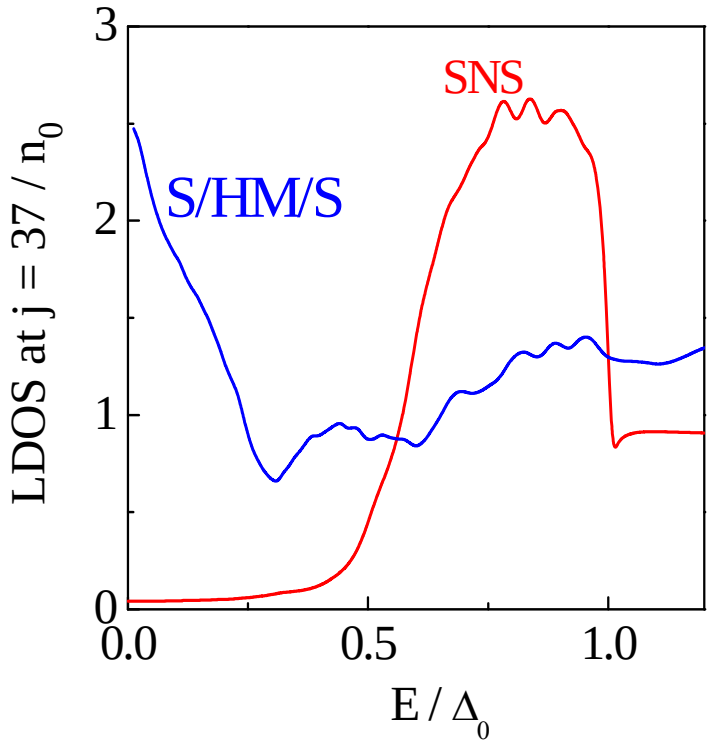
$\langle f_{\uparrow\uparrow} \rangle, \langle f_{\downarrow\downarrow} \rangle, \langle f_3 \rangle$

even-odd mix

S/HM/S

$\langle f_{\uparrow\uparrow} \rangle$ only

pure odd



Zero Energy Peak

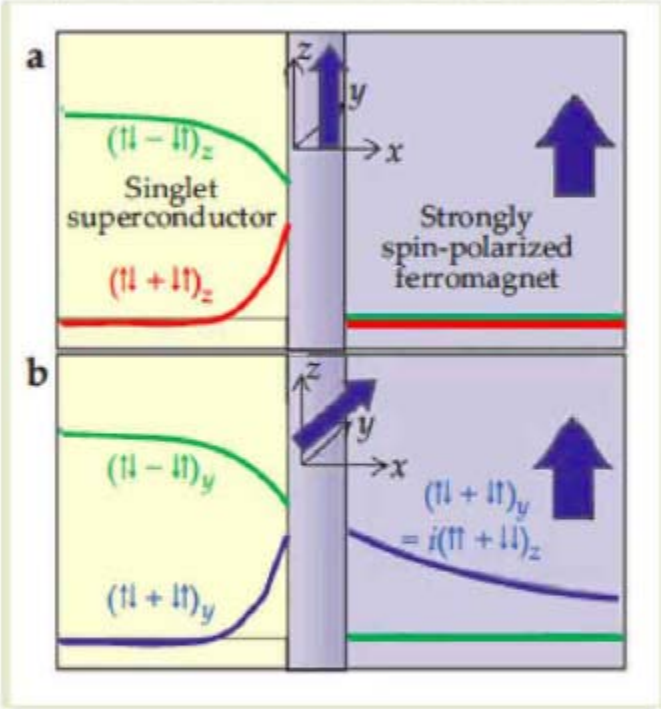
can be detected
by tunneling spectroscopy

Long-range triplet pairing in SF junctions

Bergeret, Volkov & Efetov, PRL 86(2001)4096, RMP 77(2005)1321

Eschrig, Phys. Today. 64 (2011) 43

s-wave superconductor/Ferromagnet



Magnetization

Homogeneous (Colinear)

Inhomogeneous (Noncolinear

$$\frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle) \longrightarrow |\uparrow\uparrow\rangle \quad |\downarrow\downarrow\rangle$$

In nonuniform ferromagnets, the s-wave spin-triplet equal-spin paring can be induced.

Long-range in F

Purpose

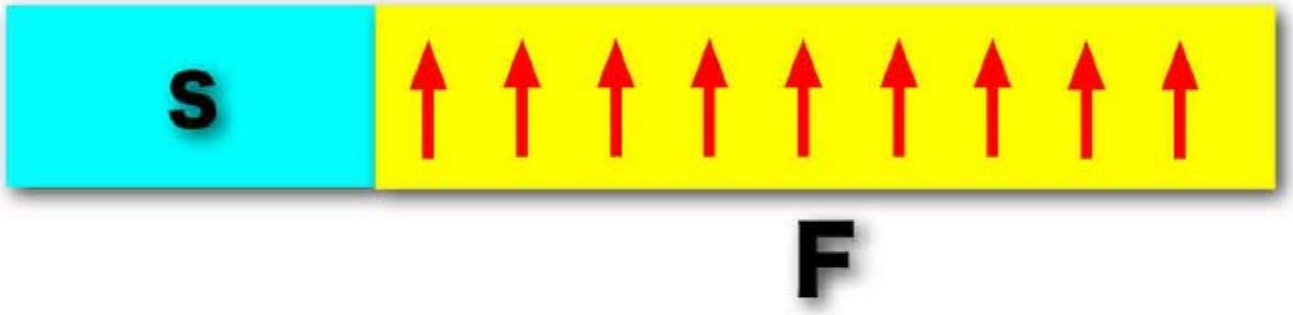
A systematic study of **ZEP** in SF junctions with inhomogeneous magnetization

1. **ROBUSTNESS** of ZEP induced by long-range triplet
2. **ZEP spectroscopy: detection** of the long-range triplet

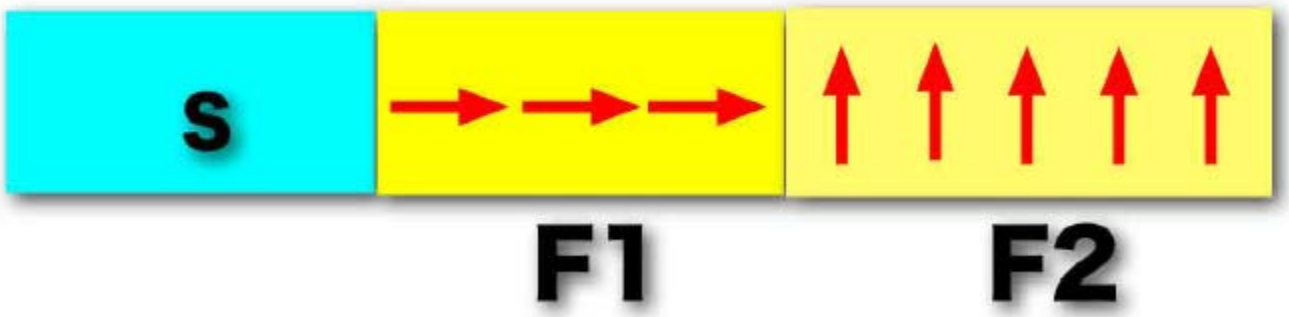
Model

Diffusive SF junction

Homogeneous magnetization



Inhomogeneous NONCOLINEAR magnetization



Quasiclassical theory

Solving the NONLINEAR Usadel equation numerically

$$i\hbar D \frac{d}{dx}\left(\check{g}\frac{d}{dx}\check{g}\right) - \left[\check{H}, \check{g}\right] = 0$$

4×4 matrix

Green function $\check{g}(x,\epsilon) = \begin{bmatrix} \hat{g}(x,\epsilon) & \hat{f}(x,\epsilon) \\ \underset{\sim}{\hat{f}}(x,\epsilon) & \underset{\sim}{\hat{g}}(x,\epsilon) \end{bmatrix}$

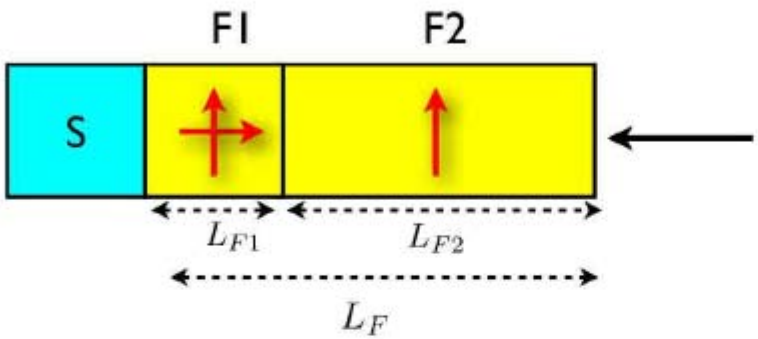
Pair function $\hat{f} = \begin{bmatrix} f_{\uparrow\uparrow} & f_{\uparrow\downarrow} \\ f_{\downarrow\uparrow} & f_{\downarrow\downarrow} \end{bmatrix} = [f_0 + \vec{f}\cdot\vec{\sigma}]\sigma_2 = \begin{bmatrix} if_1 + f_2 & -if_3 - if_0 \\ -if_3 + if_0 & -if_1 + f_2 \end{bmatrix}$ 2×2

f_0	Singlet
f_3	Triplet (Short range)
f_1 f_2	Triplet (Long range)

Kupuryanov-Lukichev boundary condition & Ricatti parametrization

Schopohl, cond-mat/9804064

Parameters



DOS & Pair function@EDGE of F2

$\xi_{Tc} = \sqrt{\frac{D}{2\pi T_c}}$ Coherence length

$E_{\mathrm{Th}} = \frac{\hbar D}{L_F^2}$ Thouless energy

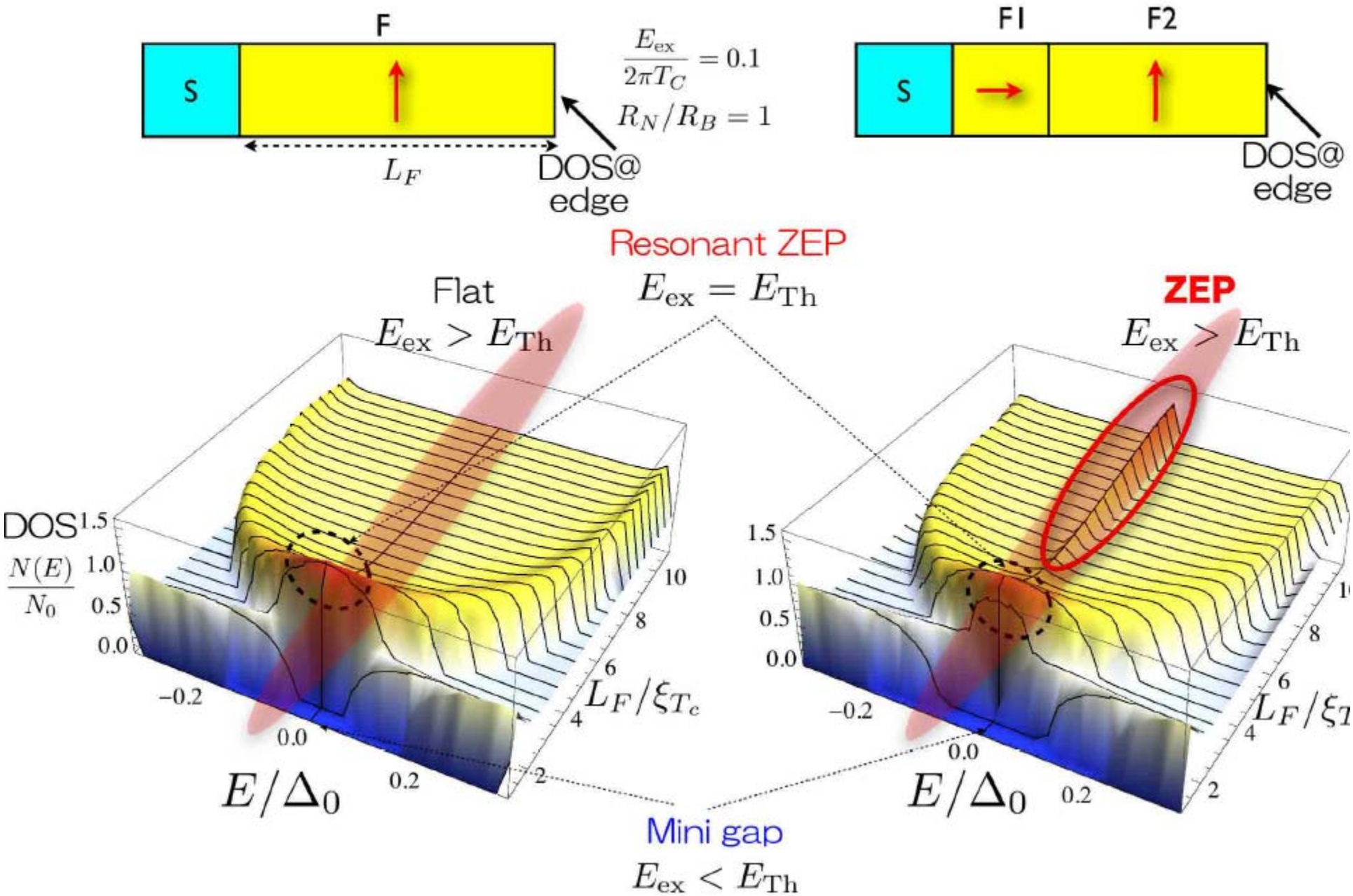
R_N Resistance of F layer

L_F	F layer thickness
E_{ex}	Exchange energy ($\ll\Delta$) weak ferromagnet
R_B	SF barrier resistance

Assumptions $D = D_F = D_S$

$L_{F_1} = 0.5\xi_{T_c}$

DOS


F
S
L_F
DOS@ edge
$\frac{E_{ex}}{2\pi T_C} = 0.1$
$R_N/R_B = 1$
F1
F2
S
DOS@ edge
Resonant ZEP
$E_{ex} = E_{Th}$
Flat
$E_{ex} > E_{Th}$
ZEP
$E_{ex} > E_{Th}$
DOS
$\frac{N(E)}{N_0}$
1.5
1.0
0.5
0.0
−0.2
0.0
0.2
E/Δ_0
L_F/ξ_{T_c}
2
4
6
8
10
Mini gap
$E_{ex} < E_{Th}$

Phase diagram

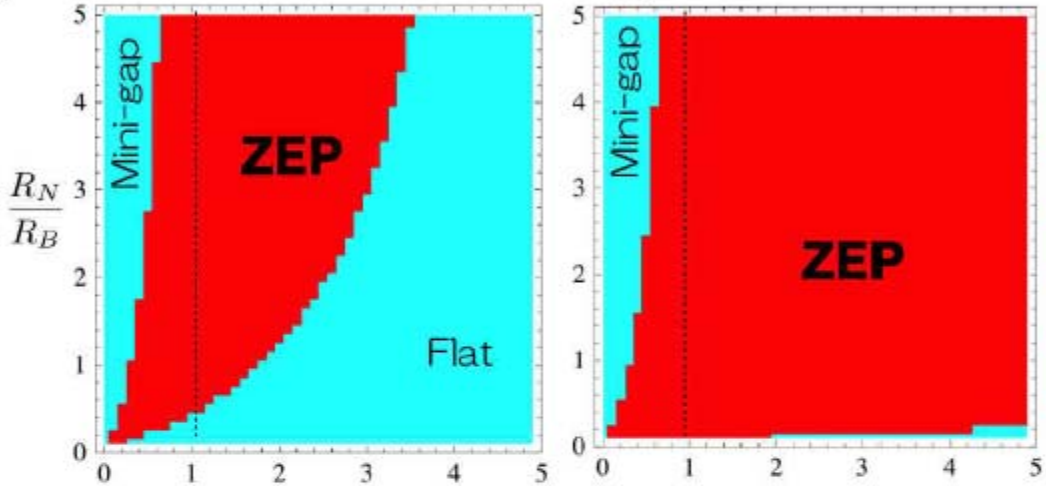
DOS,f@edge

Proximity effect

Strong

Weak

Zero energy DOS

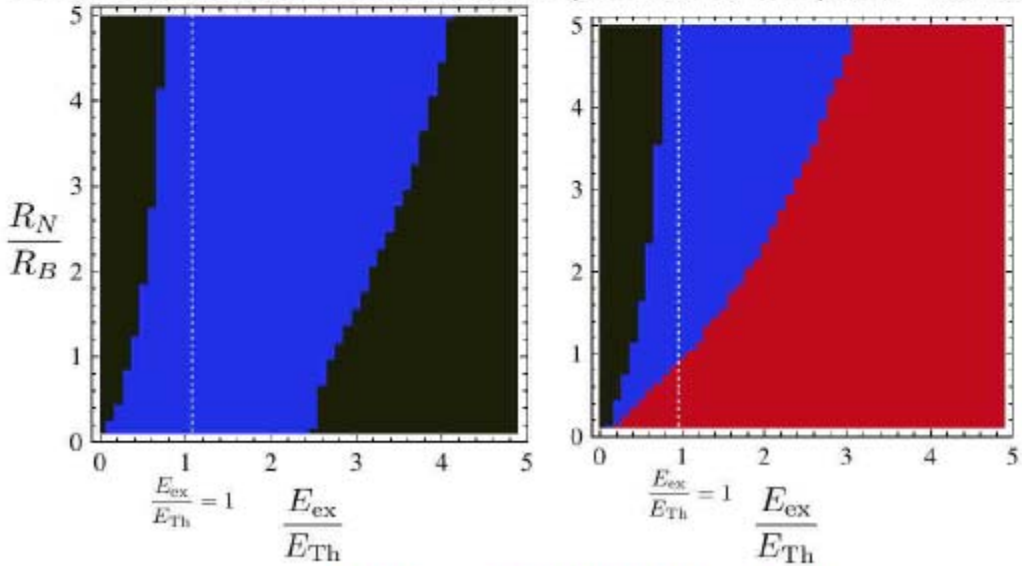


ZEP $\frac{N(E=0)}{N_0} > 1.02$

Flat $\frac{N(E=0)}{N_0} \approx 1$

Mini-gap $\frac{N(E=0)}{N_0} \ll 1$

The most dominant component of pair functions

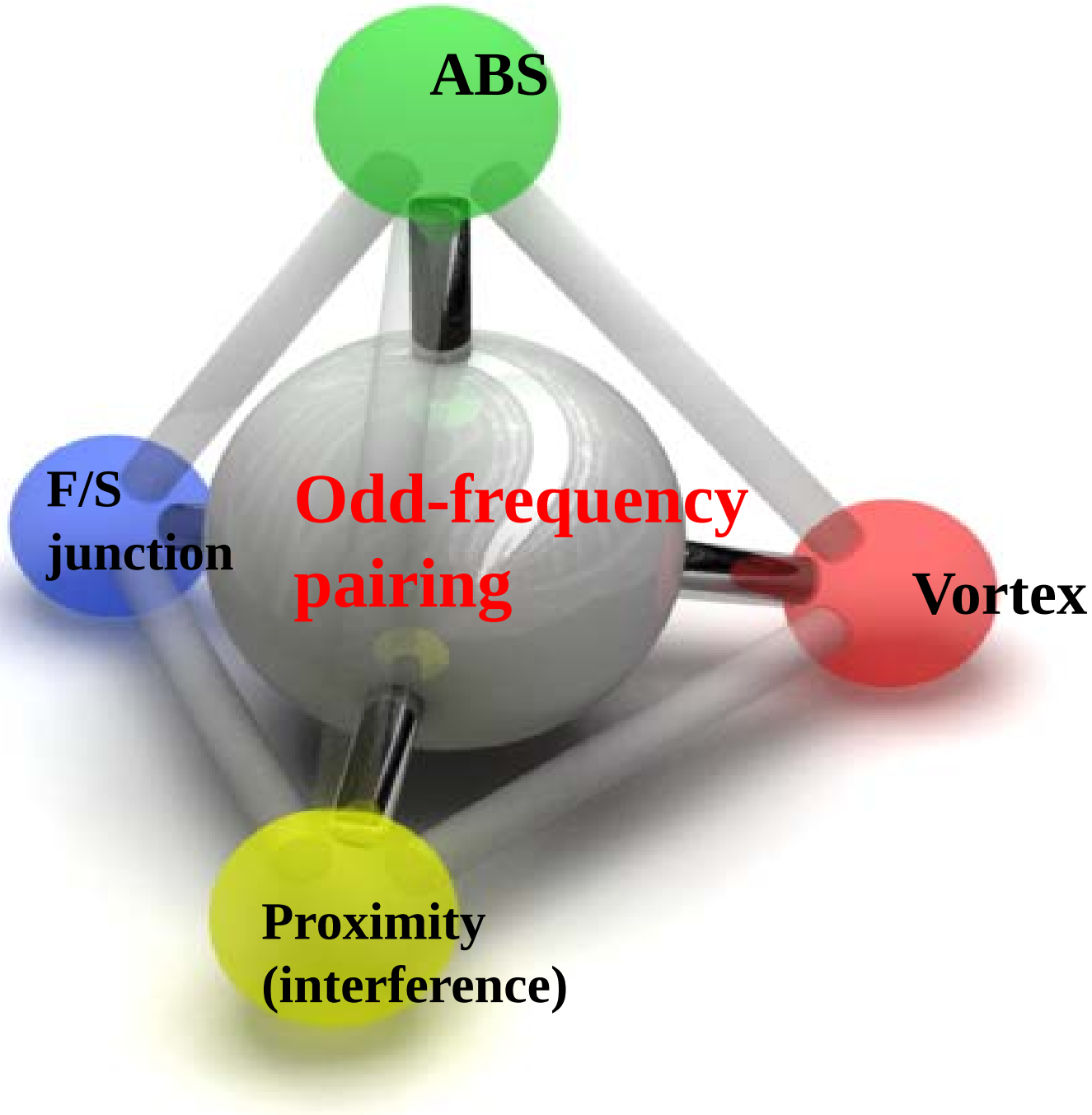


Singlet

Triplet (Short-range)

Triplet (Long-range

Formation of ZEP is ROBUST in inhomogeneous cases


ABS
F/S
junction
Odd-frequency
pairing
Vortex
Proximity
(interference)

Summary

(1) Ubiquitous presence of the **odd-frequency pairs** in **inhomogeneous** systems.

(2) Low energy Andreev bound states can be expressed in terms of odd-frequency pairing (**proximity effect** and **vortices**).

(3) The origin of the **anomalous proximity effect in DN/spin-triplet p-wave junction** is the generation of the odd-frequency pairing state.

(4) Odd-frequency triplet pairs are generated in **S/F** junctions.