



Nonreciprocal responses in superconductors: Finite-q pairing, parity mixing, and quantum geometry



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Why nonreciprocal responses?

Nonreciprocal response: left-mover \neq right-mover

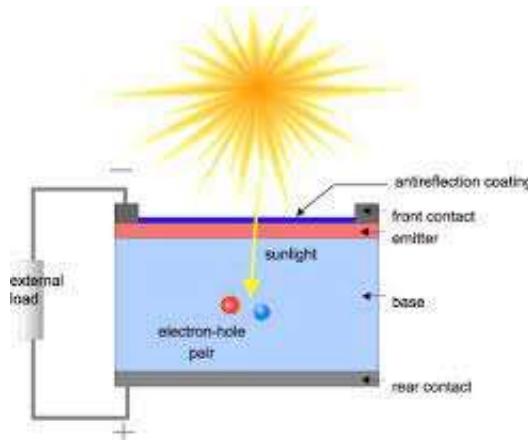


Diode: Building block of modern technology

LED = light-emitting diode



Photocurrent



Solar cell

Light sensor

Second harmonic generation

Green laser



Why nonreciprocal responses?

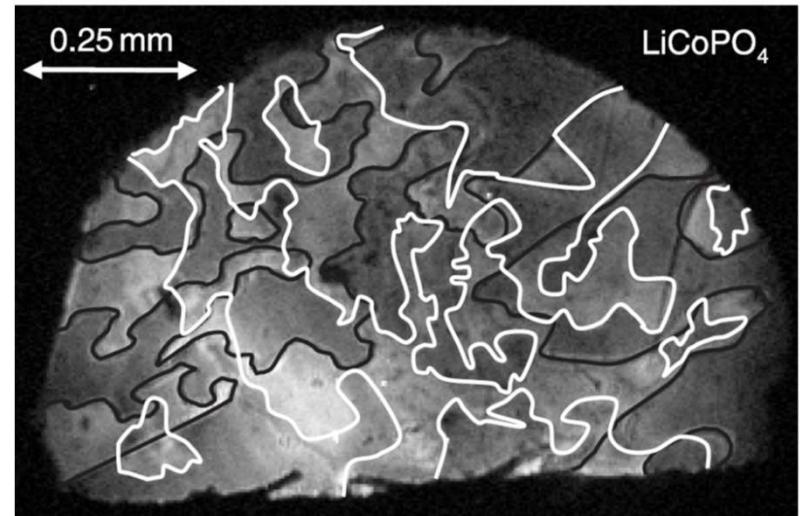
Sensitive to Inversion symmetry breaking, since it is needed.

Probing the quantum materials

Visualization of ferro toroidal domains by SHG

N. A. Spaldin, M. Fiebig, and M. Mostovoy,
[J. Phys. Condens. Matter 20, 434203 \(2008\)](#).

Application to many parity-breaking magnet



Question: probing parity-breaking superconducting phases possible?

Order parameter, symmetry, topology, criticality

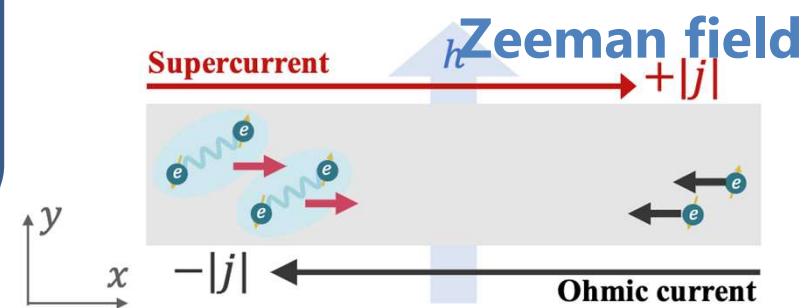
This talk: finite-q pairing, parity mixing in Cooper pairs, quantum geometry

Studied for a long time, but clarification has been awaited.

Contents

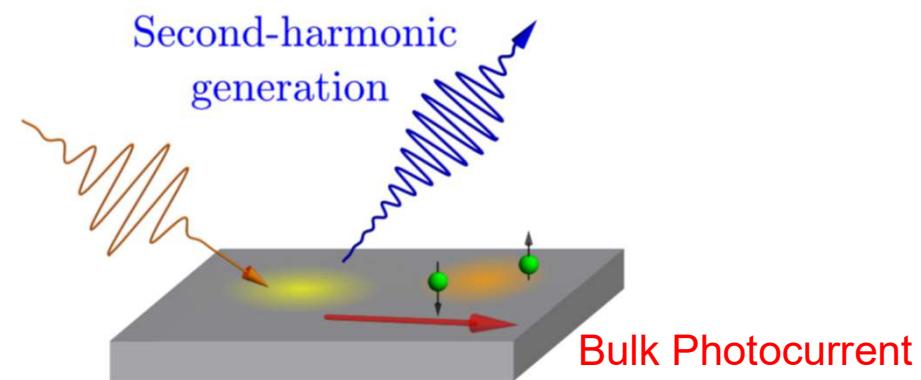
1. Superconducting diode effect (SDE)

Finite- q pairing in the SC state
(FFLO, helical, anapole)



2. Nonreciprocal optics

Quantum geometry of Bloch electrons
Parity mixing in Cooper pairs



Contents

1. Superconducting diode effect (SDE)

Finite- q pairing in the SC state
(FFLO, helical, anapole)



Akito Daido
(Kyoto)

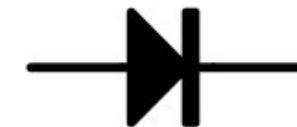
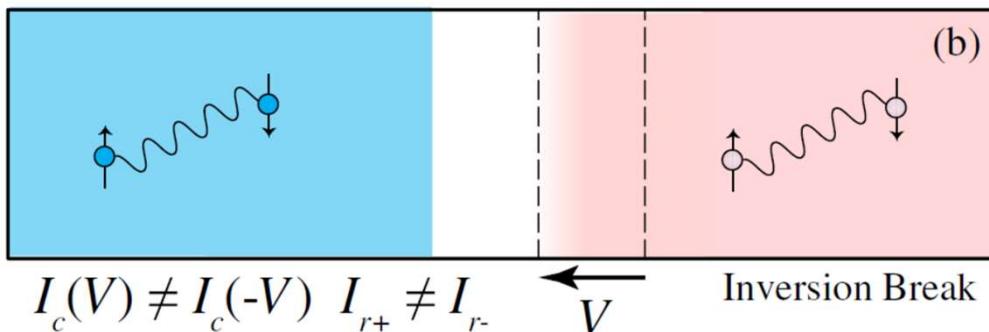


Yuhei Ikeda
(Kyoto)

2. Nonreciprocal optics

Quantum geometry of Bloch electrons
Parity mixing in Cooper pairs

Josephson diode



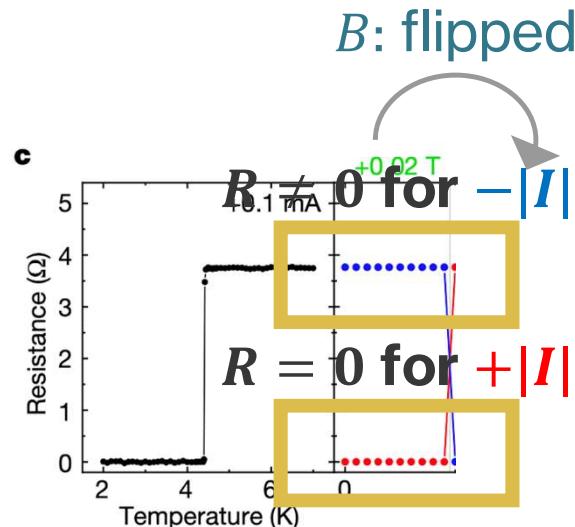
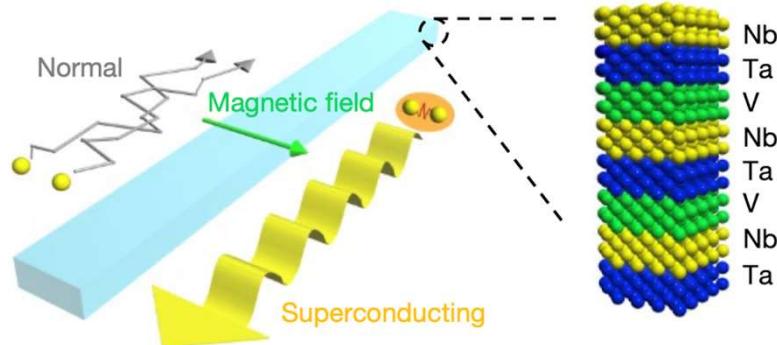
Y. Zhang et al. Phys. Rev. X 12, 041013 (2022)

$$J_T = J_0 \sin \theta + J_1 \cos 2\theta + \dots$$

- J. Hu, C. Wu, and X. Dai, Phys. Rev. Lett. 99, 067004 (2007).
- A. A. Reynoso, G. Usaj, C. A. Balseiro, D. Feinberg, and M. Avignon, Phys. Rev. Lett. 101, 107001 (2008).
- A. Zazunov, R. Egger, T. Jonckheere, and T. Martin, Phys. Rev. Lett. 103, 147004 (2009).
- I. Margaris, V. Paltoglou, and N. Flytzanis, J. Phys. Condens. Matter 22, 445701 (2010).
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- M. A. Silaev, A. Y. Aladyshkin, M. V. Silaeva, and A. S. Aladyshkina, J. Phys. Condens. Matter 26, 095702 (2014).
- G. Campagnano, P. Lucignano, D. Giuliano, and A. Tagliacozzo, J. Phys. Condens. Matter 27, 205301 (2015).
- F. Dolcini, M. Houzet, and J. S. Meyer, Phys. Rev. B 92, 035428 (2015).
- C.-Z. Chen, J. J. He, M. N. Ali, G.-H. Lee, K. C. Fong, and K. T. Law, Phys. Rev. B 98, 075430 (2018).
- M. Minutillo, D. Giuliano, P. Lucignano, A. Tagliacozzo, and G. Campagnano, Phys. Rev. B 98, 144510 (2018).
- S. Pal and C. Benjamin, EPL 126, 57002 (2019).

Superconducting diode effect

F. Ando, Y. Miyasaka, T. Li, J. Ishizuka, T. Arakawa, Y. Shiota, T. Moriyama, Y. Yanase, and T. Ono Nature **584**, 373 (2020)



Superconducting diode effect $R(-I) > R(I) = 0$

- ✓ 100% rectification
- ✓ Ideal diode



= nonreciprocity in *critical current*.

$$\Delta j_c = j_{c+} - |j_{c-}| \neq 0$$

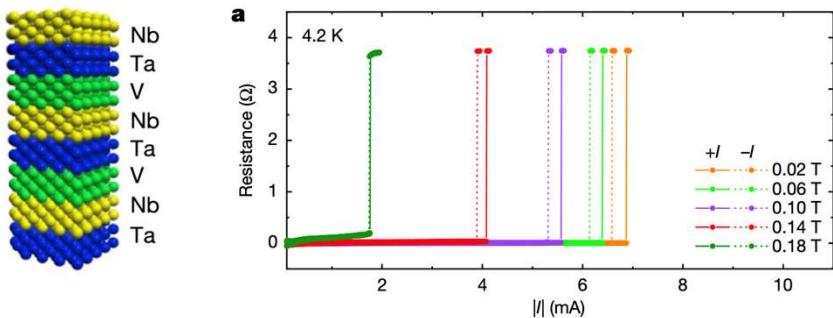
Rikken *et al.*, PRL (2001), (2005);
Magnetochiral anisotropy

$$\text{cf. } R(I) = R_0(1 + \gamma BI)$$

Experiments

Rashba system + in-plane field

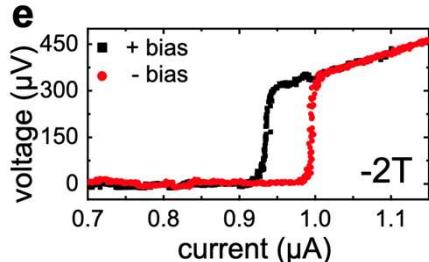
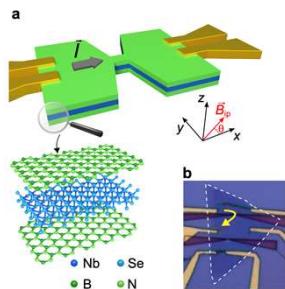
F. Ando *et al.*, Nature **584**, 373 (2020)



Ising (or Zeeman)-type SOC + out-of-plane field

NbSe_2 + mag. field L. Bauriedl *et al.*,

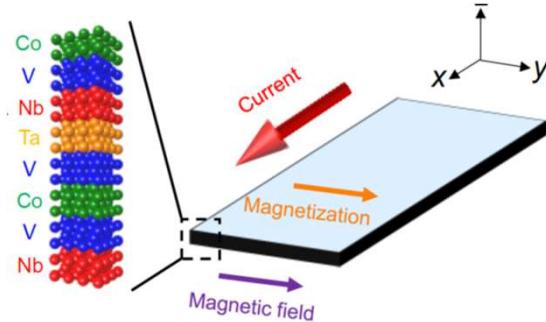
Nat. Commun. **13**, 4266 (2022)



+ Remarkable development in Josephson diode effect

Rashba system/ferromagnet hybrid

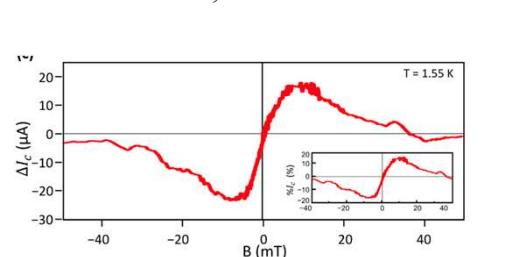
H. Narita *et al.*, Nature Nanotech. **17**, 823 (2022)



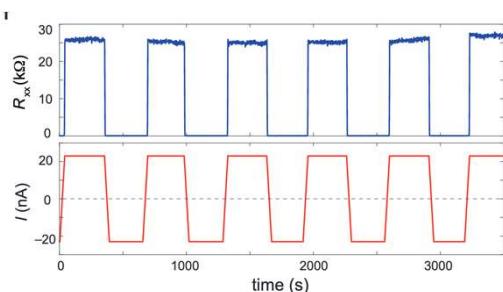
Zero field (triggered by SSB)

Twisted trilayer graphene/WSe₂

J.-X. Lin *et al.*, Nature Phys. **12**, 1221 (2022)



H. Wu *et al.*, Nature **604**, 653 (2022)



Extrinsic SC diode effect

How can SC diode effect occur?

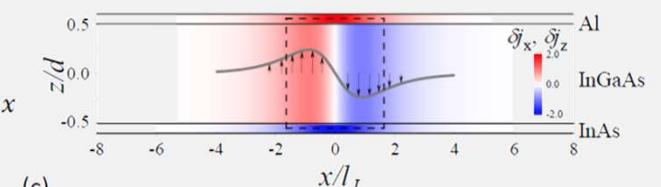
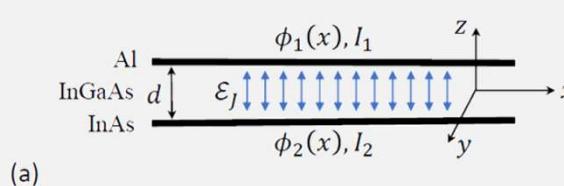
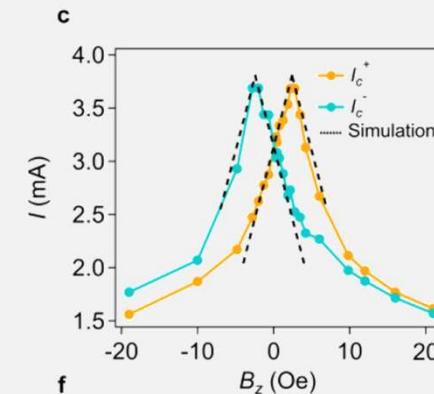
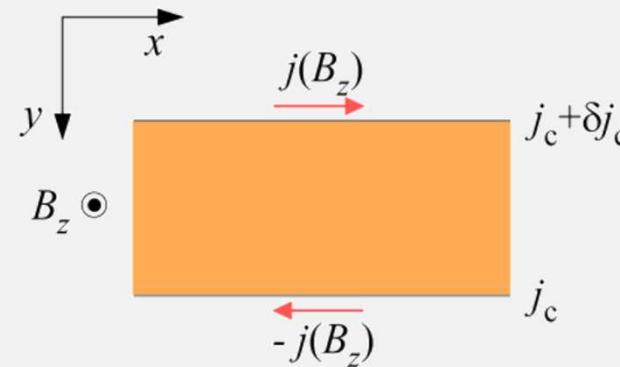
Some experiments might be understood by extrinsic mechanisms

Asymmetric
Meissner screening current

Y. Hou *et al.*, arXiv:2205.09276
M. K. Hope, *et al.*,
PRB **104**, 184512 (2021).

Vortex formation
in asymmetric multilayer SC

A. Sundaresh *et al.*, arXiv:2207.03633



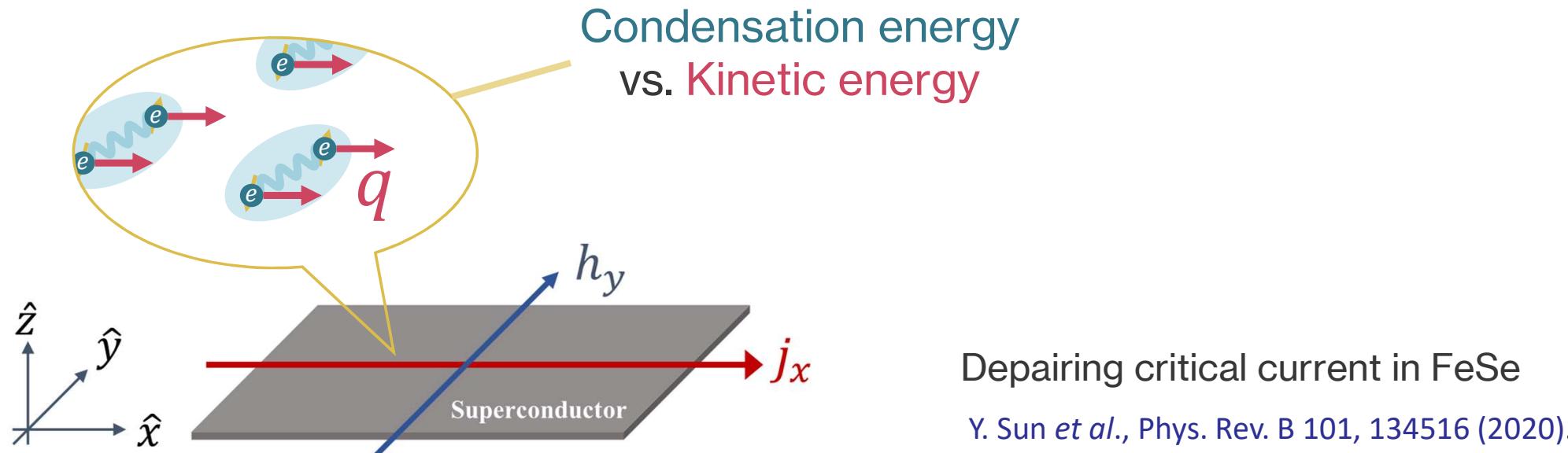
SC diode effect as an *intrinsic material property*?

Our work

The mechanism of SC diode effect is not clarified.

We propose an **intrinsic mechanism**.

Target: *depairing critical current*



Methods

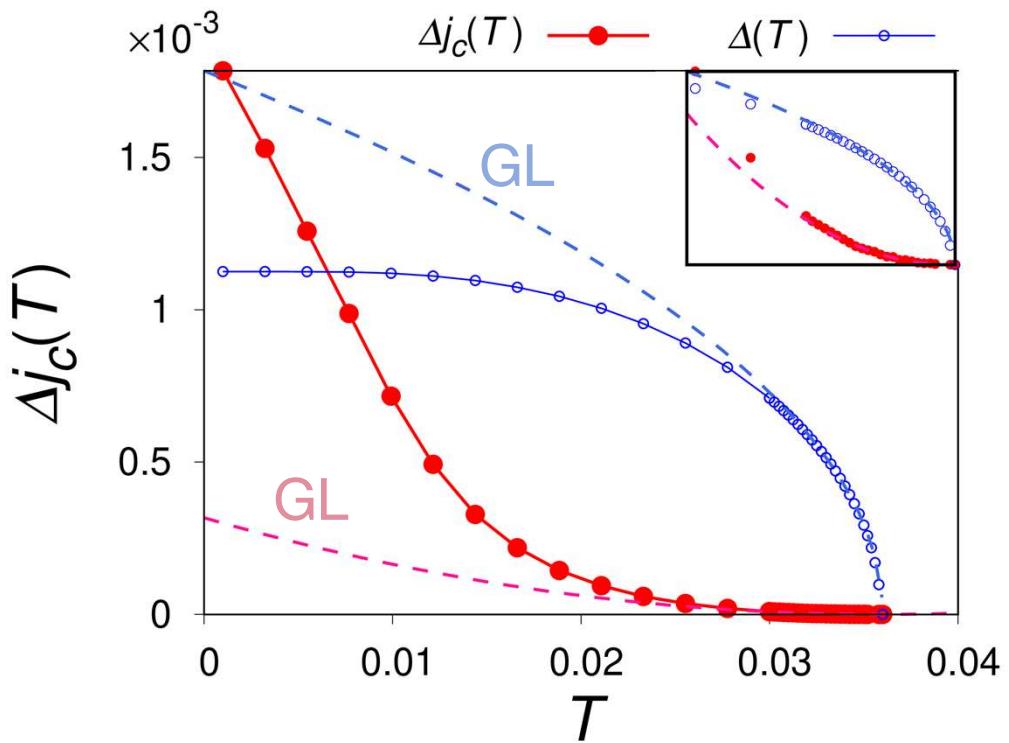
How to evaluate Δj_c ?

No diode effect in the simplest GL theory

Smidman *et al.*, Rep. Prog. Phys. **80**, 036501 (2017).

- 1 **GL with higher-order spatial derivatives**
- 2 **Microscopic analysis: Relation to finite-q helical SC**
[*s*-wave/*d*-wave Rashba-Zeeman model, Mean-field theory]

Δj_c : Temperature dependence



- **Temperature scaling**

$$\text{GL: } \Delta j_c = \left(\frac{16}{27\tilde{\beta}_0\tilde{\alpha}_2} \alpha_3 - \frac{8}{9\tilde{\beta}_0^2} \beta_1 \right) \tilde{\alpha}_0^2,$$

$$\Delta j_c(T) \propto (T_c - T)^2$$

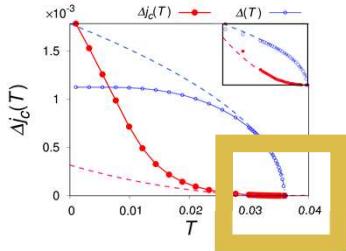
$$\Delta(T) \propto \sqrt{T_c - T}$$

$$q^3 \Delta^2 \quad q \Delta^4$$

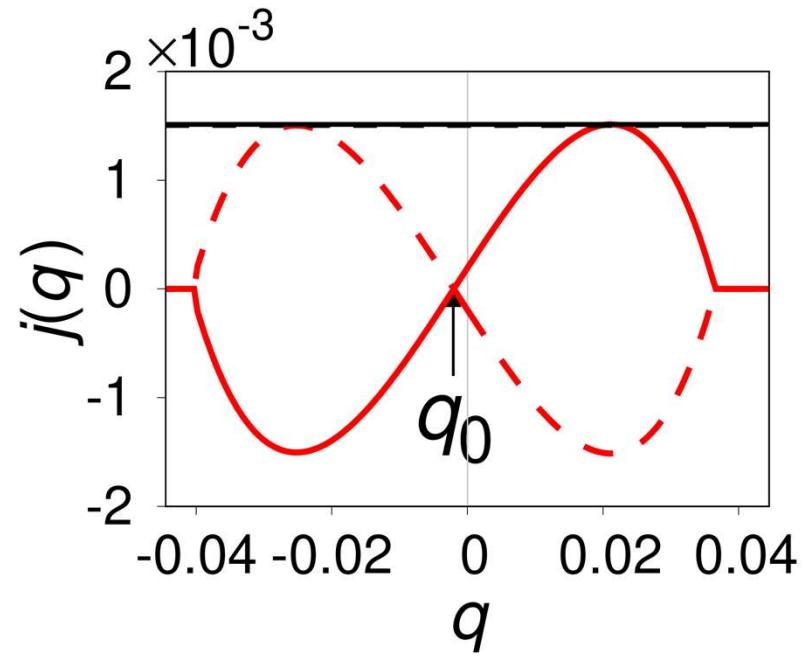
cf.) N. Yuan and L. Fu, PNAS 119, e2119548119 (2022).
J. J. He, Y. Tanaka, and N. Nagaosa, New J. Phys. 24, 053014 (2022).

consistent with microscopic calc.

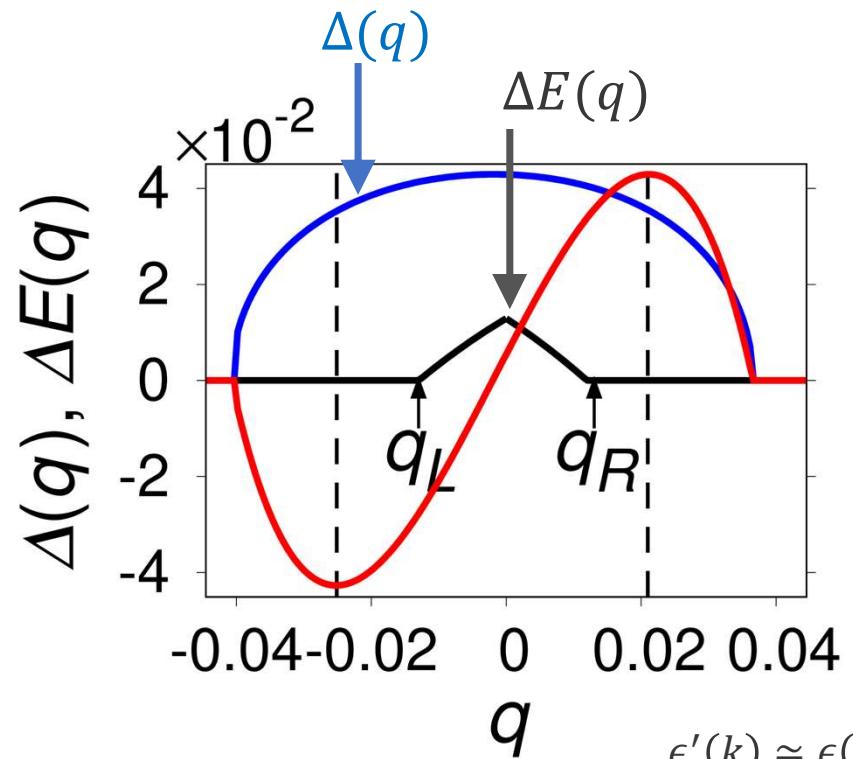
- **Enhancement at low temperature**
Why?



Diode effect at $T \sim T_c$

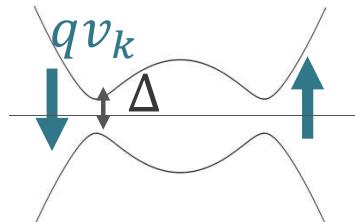


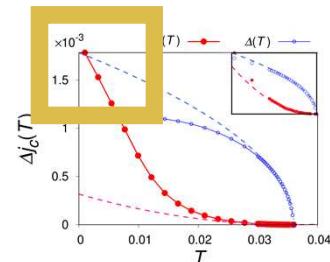
$$j_{c+} \\ |j_{c-}|$$



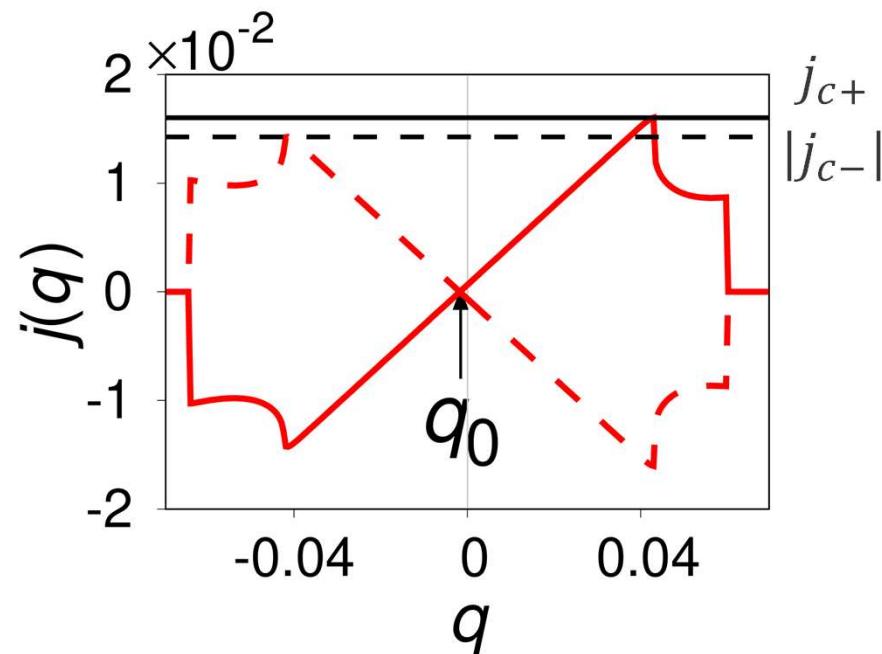
$$\epsilon'(k) \simeq \epsilon(k) + q \cdot v_k$$

- Helical superconductivity $q_0 \neq 0$
- Tiny diode effect by the asymmetry of $j(q)$ (i.e. $f(q)$)
- j_{c+} & j_{c-} achieved in the gapless region

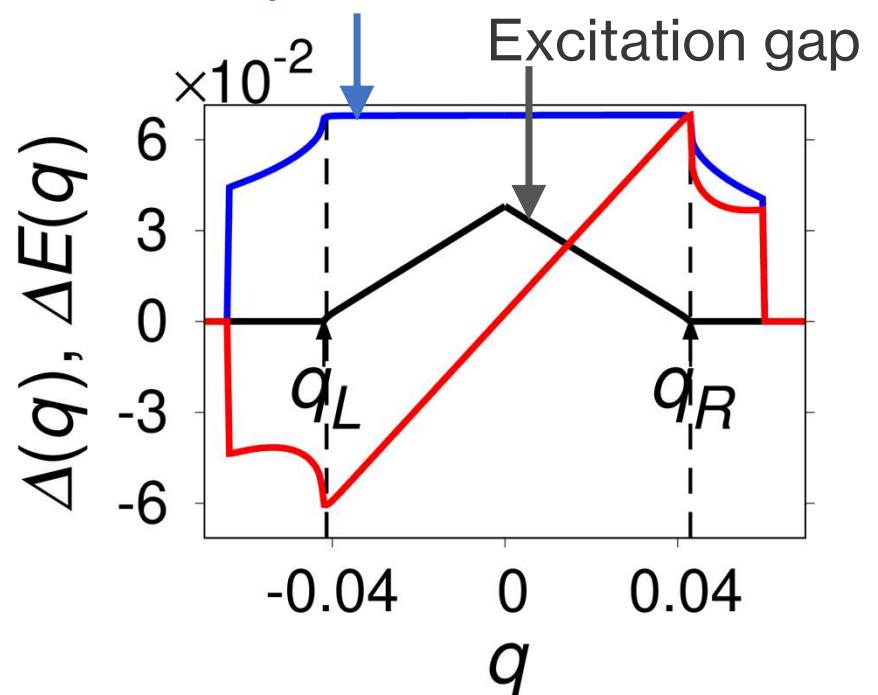




Diode effect at $T \sim 0$



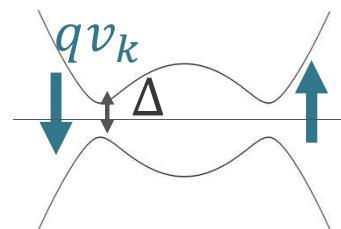
Order parameter



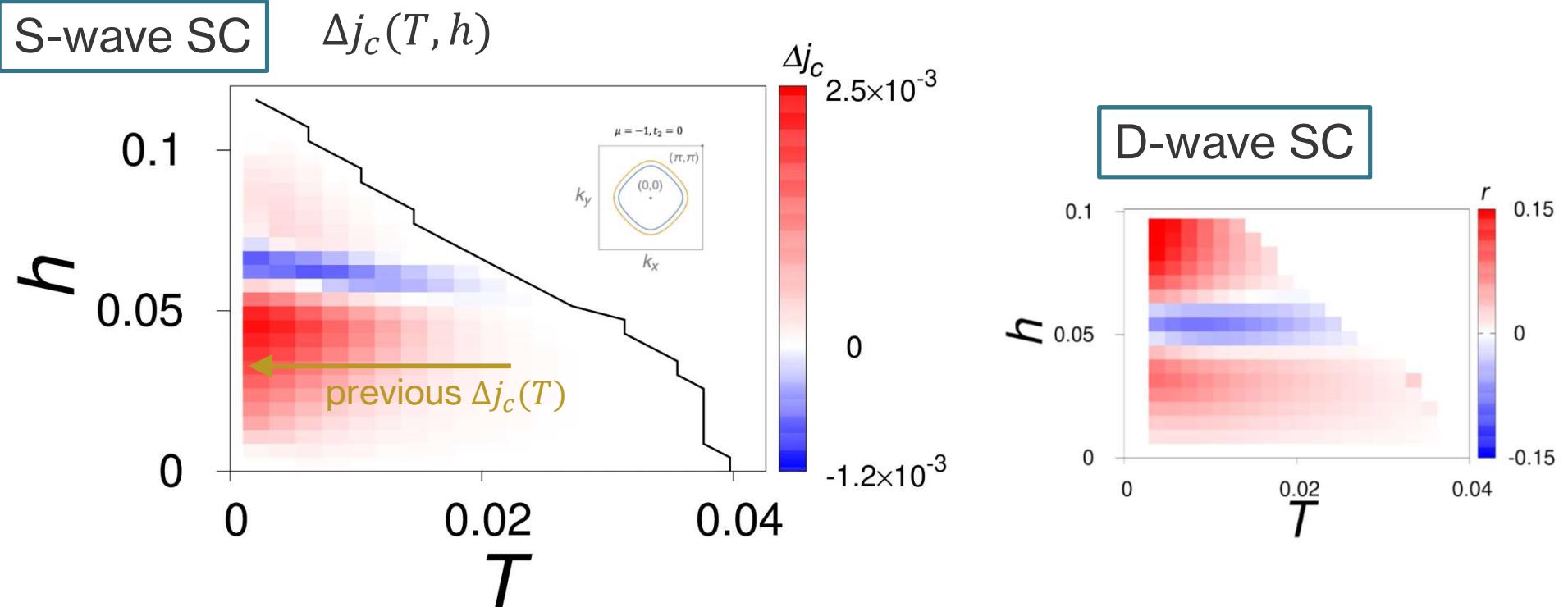
- j_{c+} & j_{c-} achieved at Landau critical momenta

- $j(q_R) = n^s(q_R - q_0)/2 \quad j(q_L) = n^s(q_L - q_0)/2$

$$\Delta j_c = n^s(q_R + q_L - 2q_0)/2$$



Phase diagram for $\Delta j_c(T, h)$

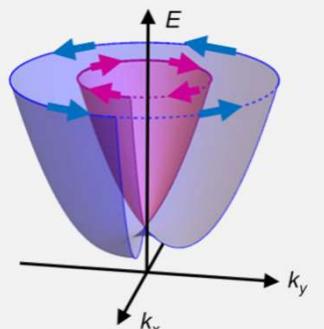


- Large Δj_c at low T and sign reversals by $h \uparrow$
- Typical behavior of noncentrosymmetric SCs

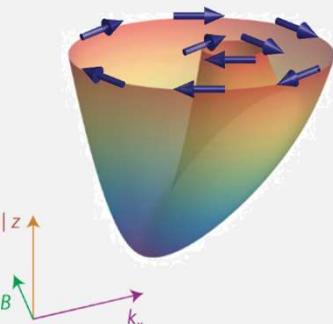
Why?

Nature of helical SC

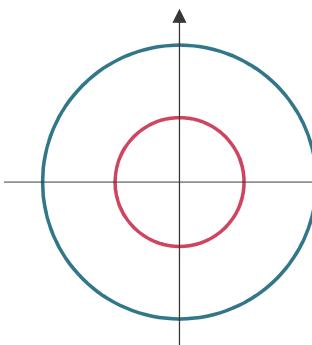
Key: Fermi-surface shift



Shcherbakov *et al.*, Sci. Adv. (2021)

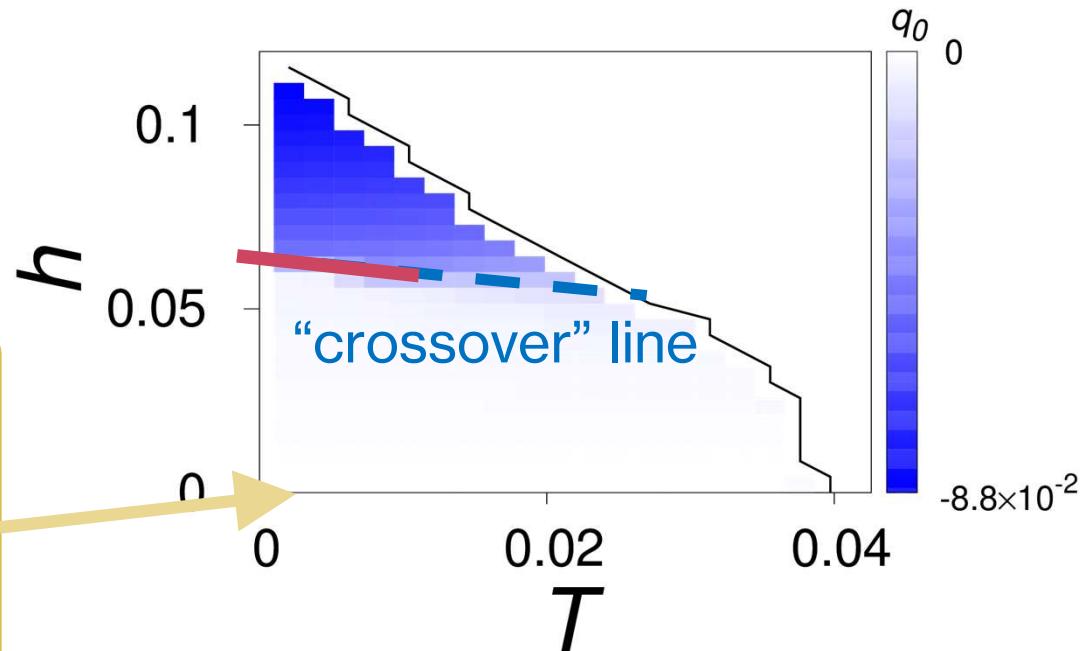


Ideue *et al.*, Nat. Phys. (2017)



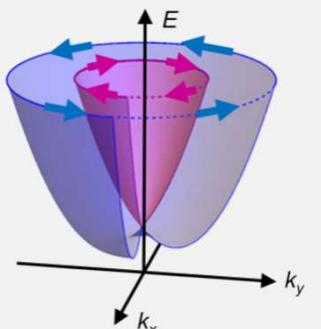
$h = 0$:
Both FS are happy with $q_0 = 0$.

Free-energy minimum at
 $q_0 (h_y, T)$

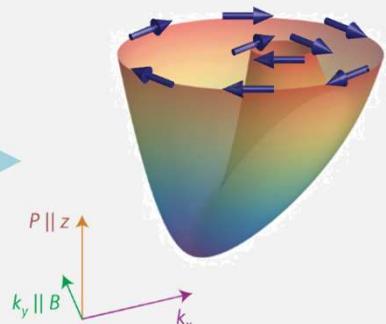


Nature of helical SC

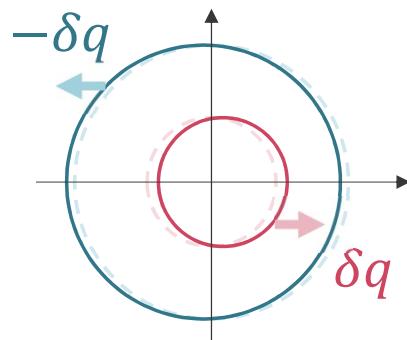
Key: Fermi-surface shift



Shcherbakov *et al.*, Sci. Adv. (2021)



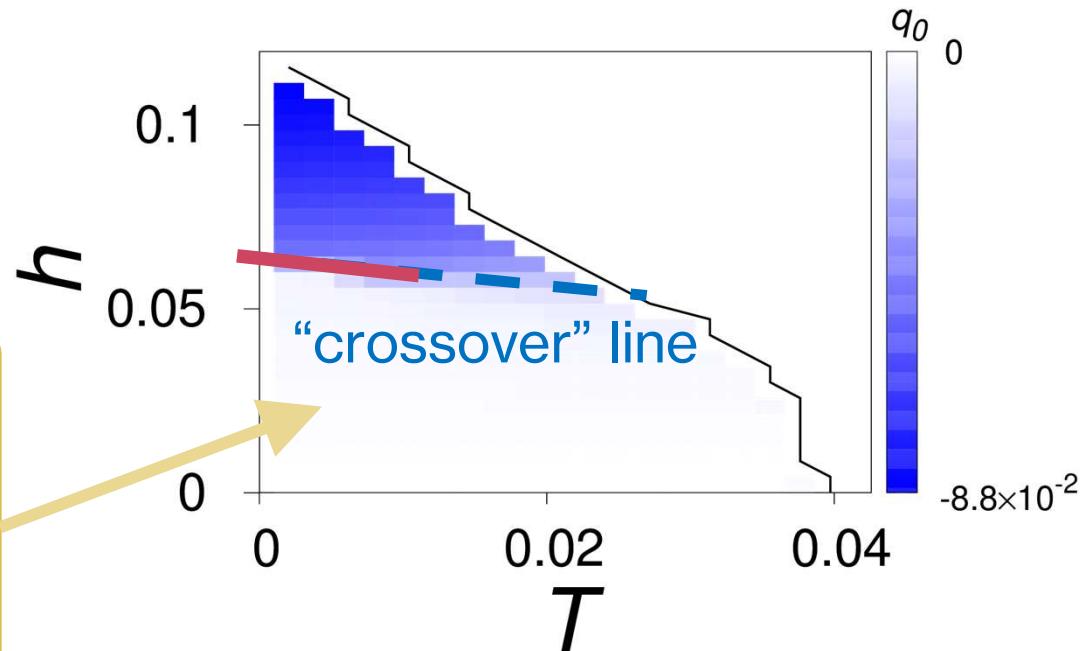
Ideue *et al.*, Nat. Phys. (2017)



$$\underline{\delta q \sim h \ll 1}$$

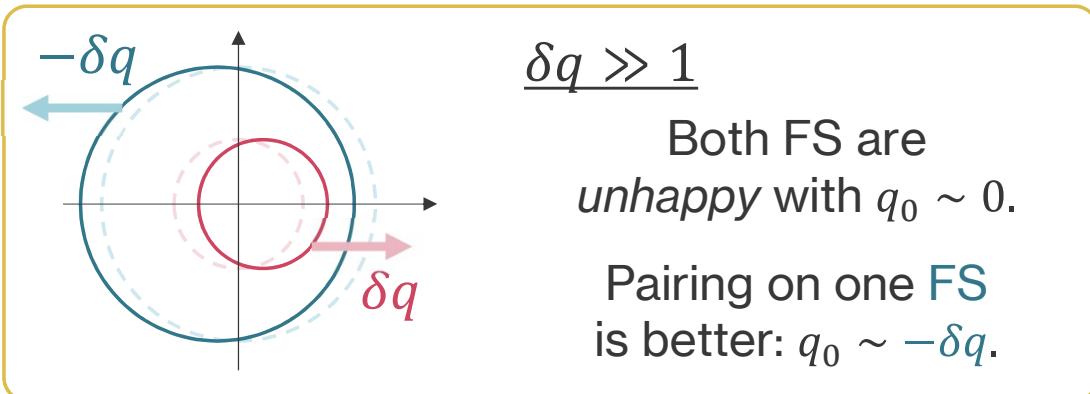
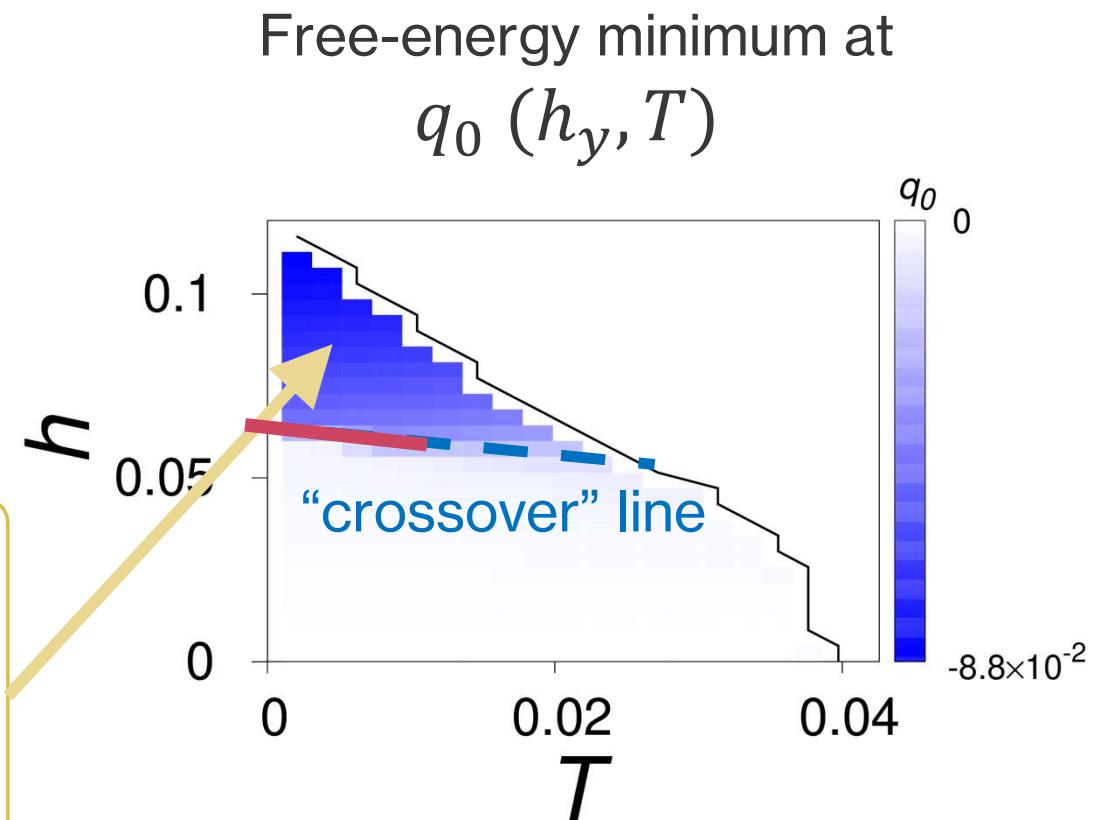
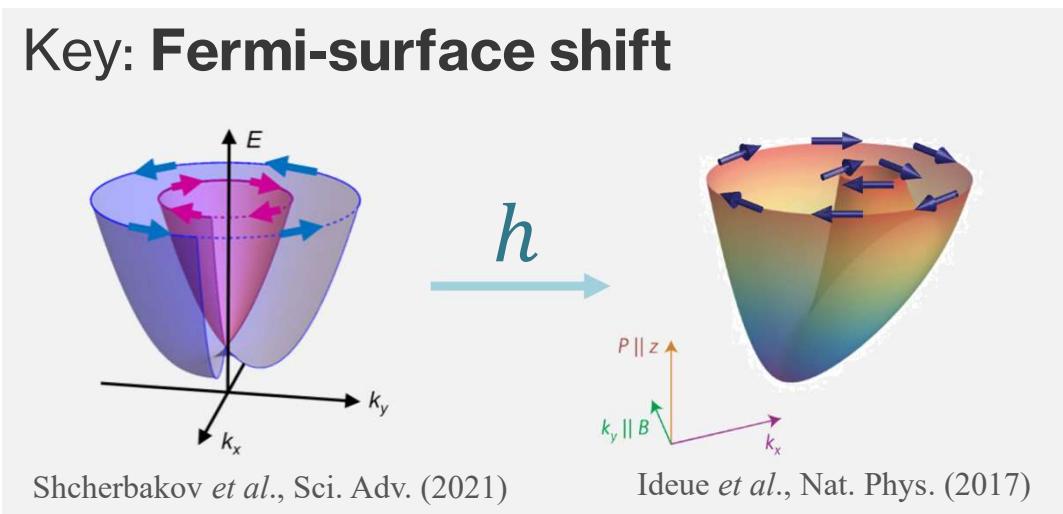
Both FS are
almost happy
with $q_0 \sim 0$.

Free-energy minimum at
 $q_0 (h_y, T)$

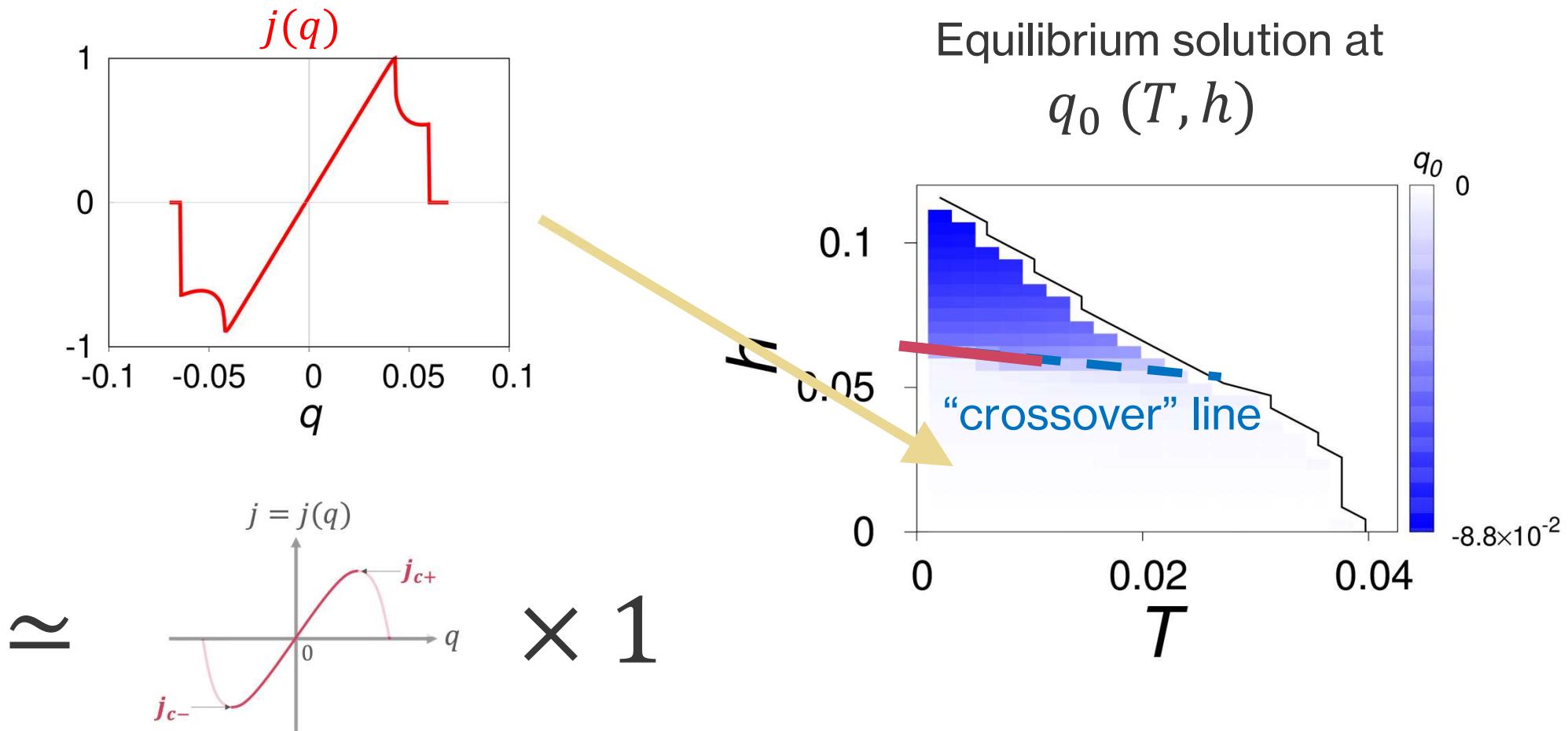


Nature of helical SC

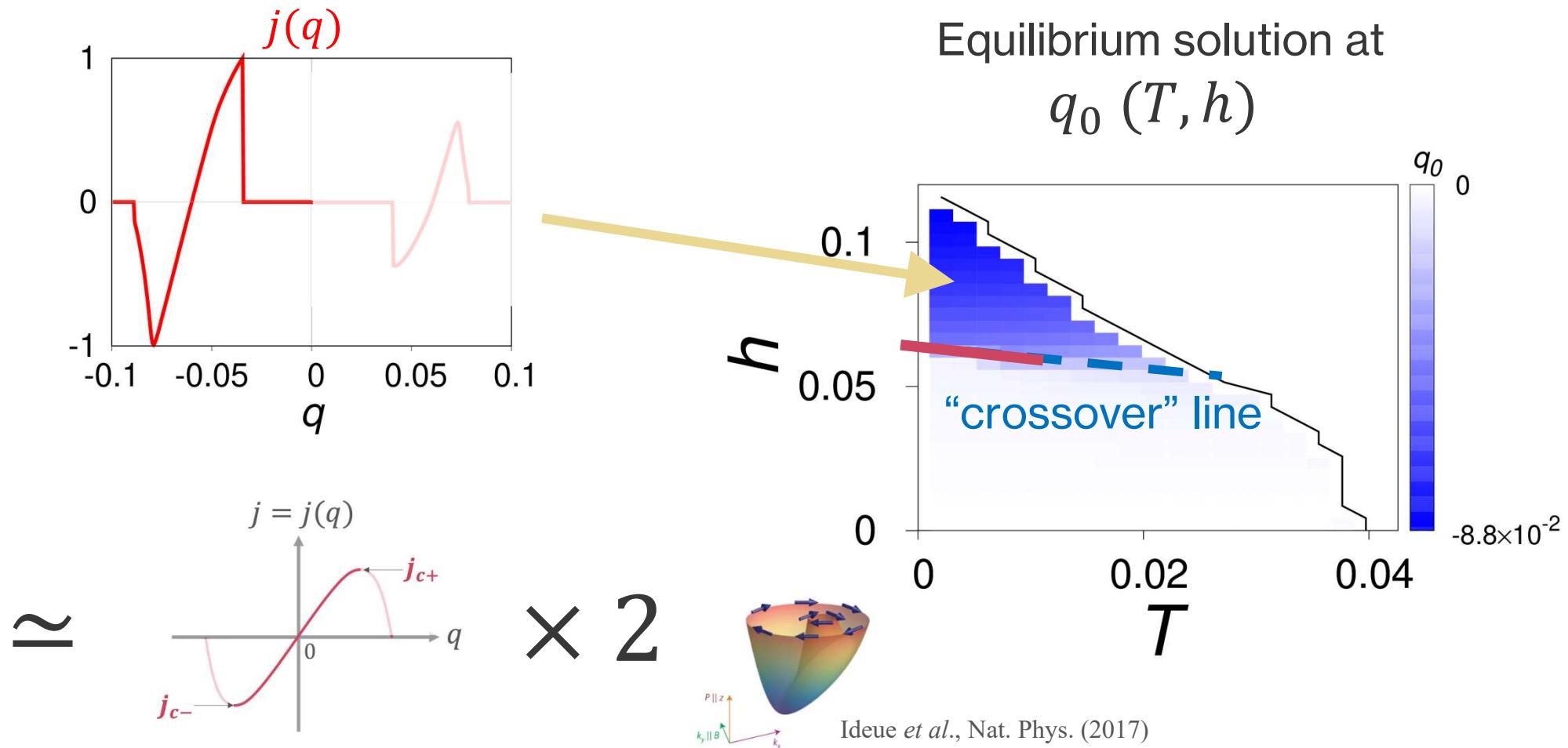
Key: Fermi-surface shift



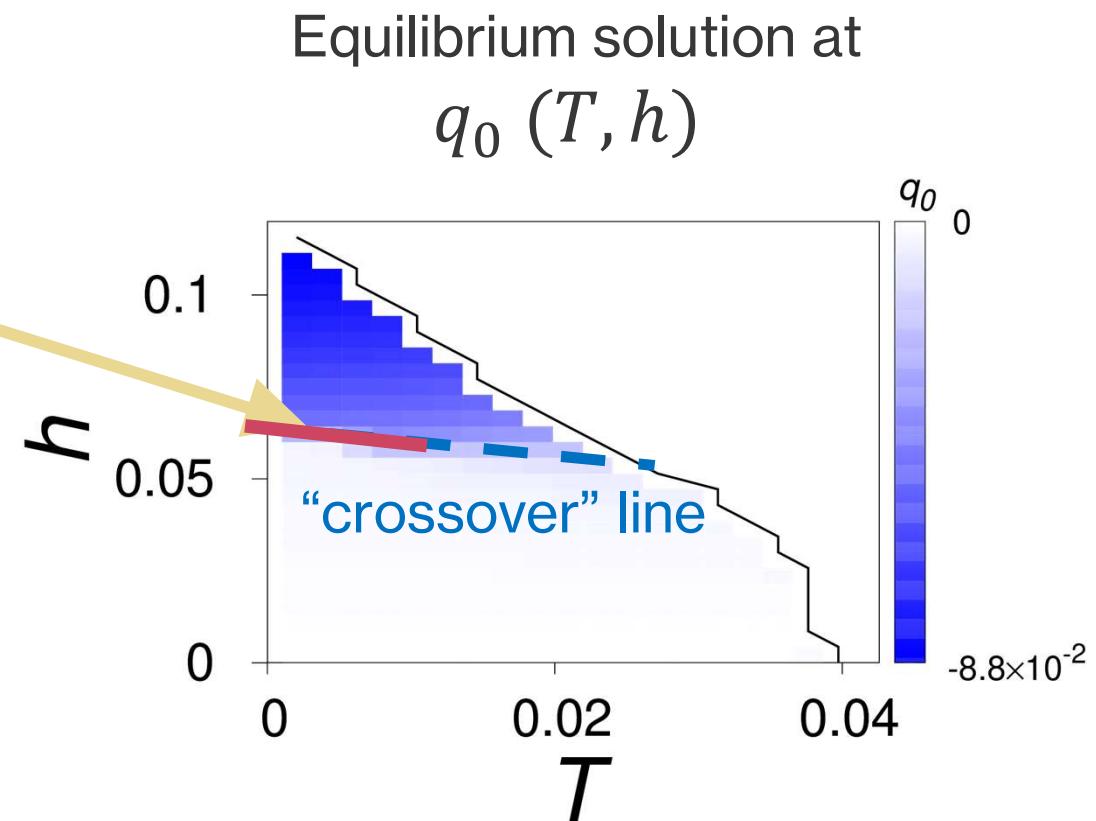
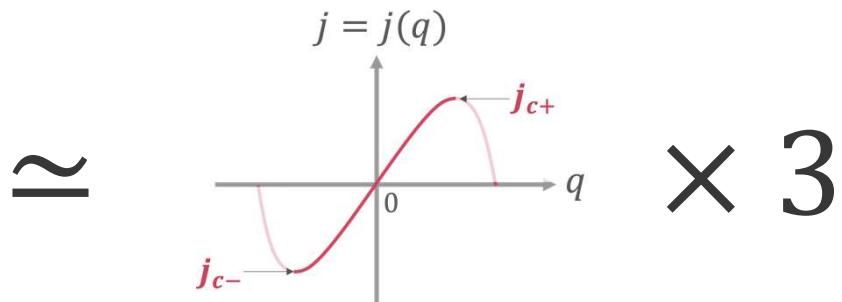
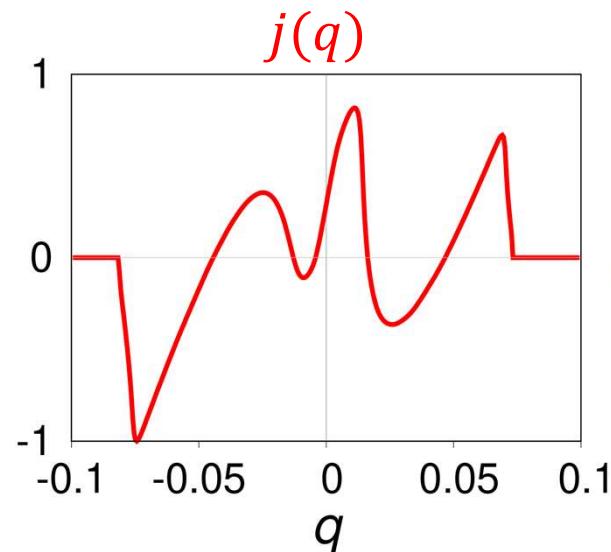
Current density $j(q)$



Current density $j(q)$

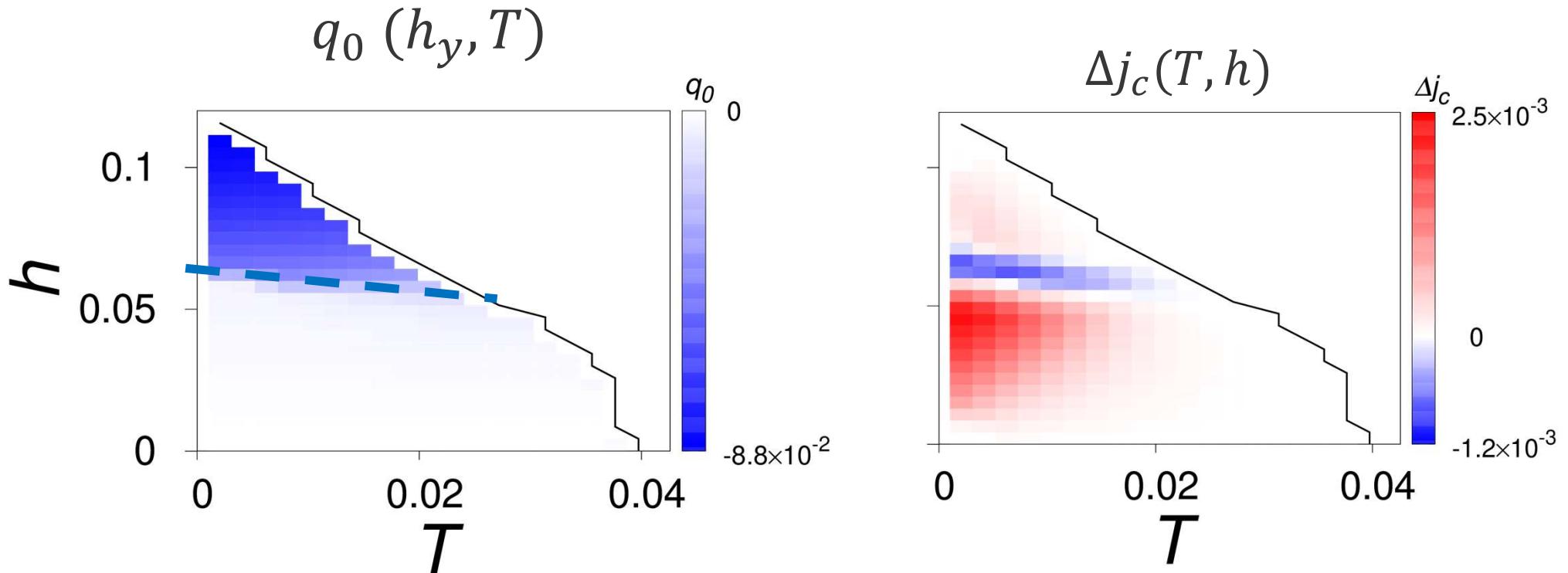


Current density $j(q)$



Phase diagram: origin of sign-reversals

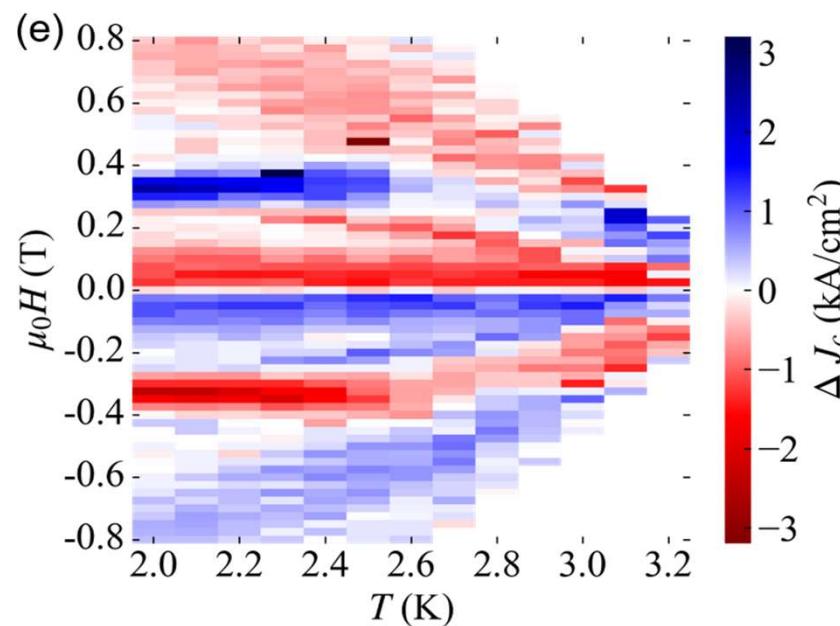
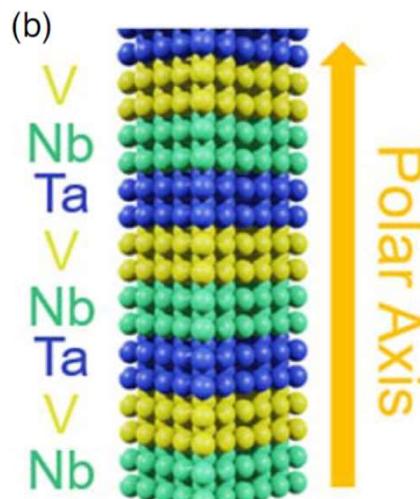
A. Daido, Y. Ikeda, and YY, PRL 128, 037001 (2022)



- Helical superconductivity is realized
- Sign reversal of SDE \leftrightarrow Change in the nature of helical SC
- SC diode effect: promising probe of finite-q helical SC!

Experiment: Polarity oscillation of SDE in H

R. Kawarasaki *et al.* Applied Physics Express 15, 113001 (2022)



- Sign reversal of SDE
- Magnetic field is smaller than theoretical prediction

Future issues

Theory:

- Layered structure
- Vortex state

Experiment:

- More 2D
- Exclusion of extrinsic mechanism (small device)

General theory of intrinsic SDE

General GL model

$$f(\mathbf{q}, \psi) = \alpha(\mathbf{q})\psi^2 + \frac{\beta(\mathbf{q})}{2}\psi^4$$

$$\begin{aligned}\alpha(\mathbf{q}) &= \alpha^{(0)} + \alpha_i^{(1)}q_i + \alpha_{ij}^{(2)}q_iq_j \\ &\quad + \alpha_{ijk}^{(3)}q_iq_jq_k + \alpha_{ijkl}^{(4)}q_iq_jq_kq_l + O(q^5), \\ \beta(\mathbf{q}) &= \beta^{(0)}(1 + \beta_i^{(1)}q_i + \beta_{ij}^{(2)}q_iq_j) + O(q^3).\end{aligned}$$

Effective SOC of Cooper pairs

$$\mathbf{g}_{\text{eff}}(\mathbf{q}) \equiv \frac{2\mathbf{g}_3(\mathbf{q})}{\sum_i q_i^2 / 2m_i} - \mathbf{g}_1(\mathbf{q})$$

$$\begin{aligned}[\alpha_3]_{ijk}\delta q_i\delta q_j\delta q_k &\equiv \mathbf{h} \cdot \mathbf{g}_3(\delta \mathbf{q}), \\ [\beta_1]_{ij}\delta q_i &\equiv \mathbf{h} \cdot \mathbf{g}_1(\delta \mathbf{q}),\end{aligned}$$

Intrinsic SDE in GL region

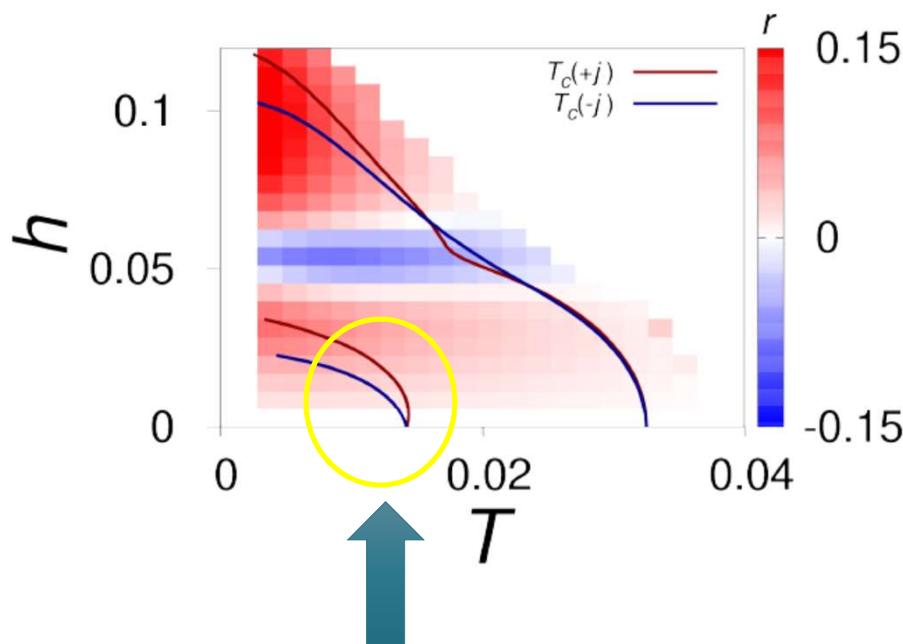
$$\Delta j_c(\hat{n}) = \frac{8a_0^2}{9\beta_0}(T_c - T)^2 \mathbf{g}_{\text{eff}}(\hat{n}) \cdot \mathbf{h}$$

- All the 21 noncentrosymmetric point group allows the SDE.
(even non-gyrotropic)
- Behaviors of SDE can be predicted by the point group.

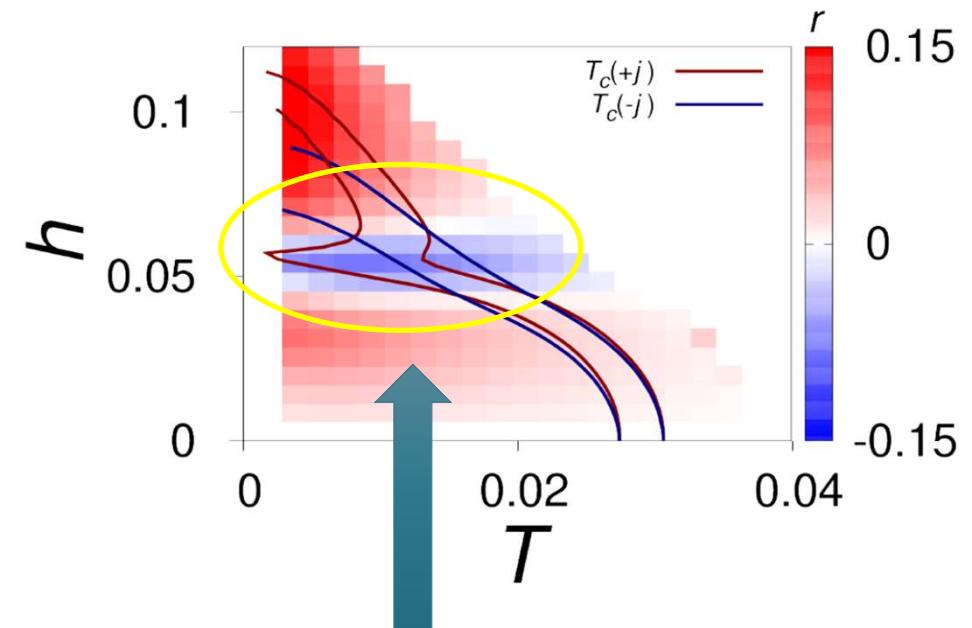
Nonreciprocal phase transition lines

A. Daido and YY, arXiv:2209.03515

Solid lines = phase transition lines under supercurrent



Large current: field-enhanced SC



Moderate current:
nonreciprocal reentrant SC

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(Univ. Tokyo)



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Formulation for nonlinear conductivity (normal state)

■ Response formula for nonlinear conductivity

$$J_{(2)}^\mu(\omega) = \int \frac{d\omega_1 d\omega_2}{(2\pi)^2} \tilde{\sigma}^{\mu;\nu\lambda}(\omega; \omega_1, \omega_2) E^\nu(\omega_1) E^\lambda(\omega_2)$$

(2nd-) nonlinear current

- Dipole-approximation (no photo-magnetic field, not spatially-dispersive)
- Non-interacting electrons + phenomenological scattering

■ Perturbation from photo-electric field

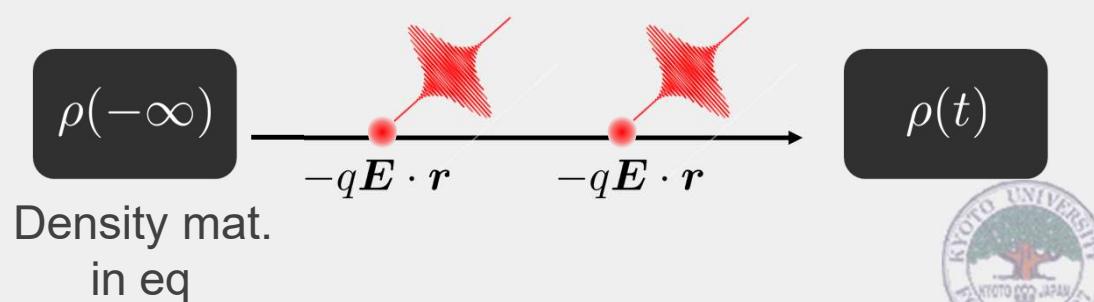
$$i\partial_t \rho(t) = [\mathcal{H}(t), \rho(t)],$$

von Neumann equation

- $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_E(t)$, $\mathcal{H}_E(t) = -q\mathbf{E}(t) \cdot \mathbf{r}$
- $\partial_t \rho \rightarrow \partial_t \rho + i\gamma(\rho - \rho_{\text{eq}})$

J. E. Sipe & A. I. Shkrebtii, Phys. Rev. B 61, 5337 (2000);

G. B. Ventura et al., Phys. Rev. B 96, 035431 (2017)



Formulation for nonlinear conductivity (normal state)

■ Second order term in electric field

Monochromatic light induced photocurrent

$$J(t) = \text{Tr}[qv \rho(t)] \rightarrow J_{\text{dc}}^{\mu} = \sigma^{\mu;\nu\lambda}(\omega = 0; \omega_1 = -\Omega, \omega_2 = \Omega) E^{\nu}(-\Omega) E^{\lambda}(\Omega)$$

■ Polarization analysis of photocurrent conductivity

$$J_{\text{dc}}^{\mu} = \sigma^{\mu;\nu\lambda} E^{\nu}(-\Omega) E^{\lambda}(\Omega) = \sigma^{\mu;\nu\lambda} [E^{\nu}(\Omega)]^* E^{\lambda}(\Omega)$$

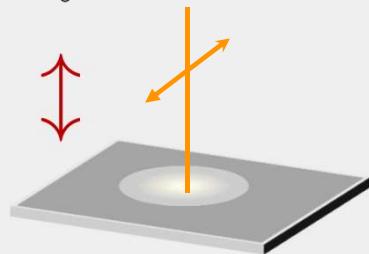
B. I. Sturman & V. M. Fridkin, (CRC Press, 1992).
Photovoltaic and Photo-refractive Effects in Noncentrosymmetric Materials

$$\sigma^{\mu;\nu\lambda} (E^{\nu})^* E^{\lambda} = \frac{1}{2} (\sigma^{\mu;\nu\lambda} + \sigma^{\mu;\lambda\nu}) \text{Re}[(E^{\nu})^* E^{\lambda}] + \frac{i}{2} (\sigma^{\mu;\nu\lambda} - \sigma^{\mu;\lambda\nu}) \text{Im}[(E^{\nu})^* E^{\lambda}],$$

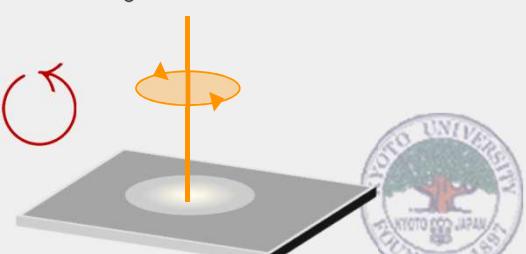
$\text{Re}[(E^{\nu})^* E^{\lambda}]$, (ν, λ) : symmetric

$\text{Im}[(E^{\nu})^* E^{\lambda}]$, (ν, λ) : anti-sym.

Linearly polarized light



Circularly polarized light



Classification of photocurrent [Watanabe and YY, Phys. Rev. X 11, 011001 (2021)]

$P \times T \circ P T \times$

T-symmetric

$P \times T \times P T \circ$

PT-symmetric

Metal

Berry curvature dipole



J. E. Moore & J. Orenstein, Phys. Rev. Lett. 105, 026805 (2010).

Intrinsic Fermi surface term



F. de Juan et al., Phys. Rev. Research 2, 012017 (2020).

Metal
and
insulator

Electric injection current



J. E. Sipe & A. I. Shkrebtii, Phys. Rev. B 61, 5337 (2000).

Shift current



R. von Baltz & W. Kraut, Phys. Rev. B 23, 5590 (1981);
J. E. Sipe & A. I. Shkrebtii, Phys. Rev. B 61, 5337 (2000).

Drude photocurrent



T. Holder et al., Phys. Rev. Research 2, 033100 (2020).

Intrinsic Fermi surface term
This work



Magnetic injection current



Y. Zhang et al., Nat. Commun. 10, 3783 (2019).

Gyration current



This work

Electric/Magnetic multipole \leftrightarrow T/PT symmetry \leftrightarrow Linearly/Circularly polarized light



Classification of photocurrent based on quantum geometry

$$\sigma^{\mu;\nu\lambda} \sim \int \frac{d\mathbf{k}}{(2\pi)^d} \sum_{a \neq b} G_{ab}^{\mu\nu\lambda} [f(\epsilon_a) - f(\epsilon_b)] \delta(\hbar\Omega - \epsilon_b - \epsilon_a), \quad G^{\mu\nu\lambda} = X^\mu T^{\nu\lambda}$$

	Director	Transition amp.
	X^μ	$T^{\nu\lambda}$
Electric injection current	Δ^μ	Group velocity difference $\Omega^{\nu\lambda}$
Shift current	R^μ	Shift vector $g^{\nu\lambda}$
Magnetic injection current	Δ^μ	$g^{\nu\lambda}$
Gyration current	R_\pm^μ	$g^{\nu\lambda}, \Omega^{\nu\lambda}$

$\Omega^{\nu\lambda}$ (band-resolved) Berry curvature

$g^{\nu\lambda}$ (band-resolved) quantum metric



Diverging photocurrent in AFM Dirac/Weyl semimetals

Watanabe and YY, Phys. Rev. X 11, 011001 (2021), J. Ahn *et al.*, Phys. Rev. X 10, 041041 (2020).



Superconductors

H. Watanabe, A. Daido, and YY, Phys. Rev. B 105, 024308 (2022)

H. Watanabe, A. Daido, and YY, Phys. Rev. B 105, L100504 (2022)

H. Tanaka, H. Watanabe, and YY, arXiv:2205.14445

Optical responses unique to superconductors

Variational parameter : λ (= vector potential A)

$$J_{ab}^\alpha(\mathbf{k}) = - \lim_{\lambda \rightarrow 0} \left\langle a_\lambda \left| \frac{\partial H_\lambda}{\partial \lambda_\alpha} \right| b_\lambda \right\rangle \quad H_\lambda(\mathbf{k}) = \begin{pmatrix} H_N(\mathbf{k} - \lambda) & \Delta \\ \Delta^\dagger & -[H_N(-\mathbf{k} - \lambda)]^T \end{pmatrix}$$



λ -derivative and \mathbf{k} -derivative are not equivalent.



Opposite charges of electrons and holes



Divergent response at THz or sub THz regime

Nonreciprocal
superfluid weight

$$\sigma_{\text{NRSF}}^{\alpha;\beta\gamma} = \frac{1}{2\omega_1\omega_2} f^{\alpha\beta\gamma}$$

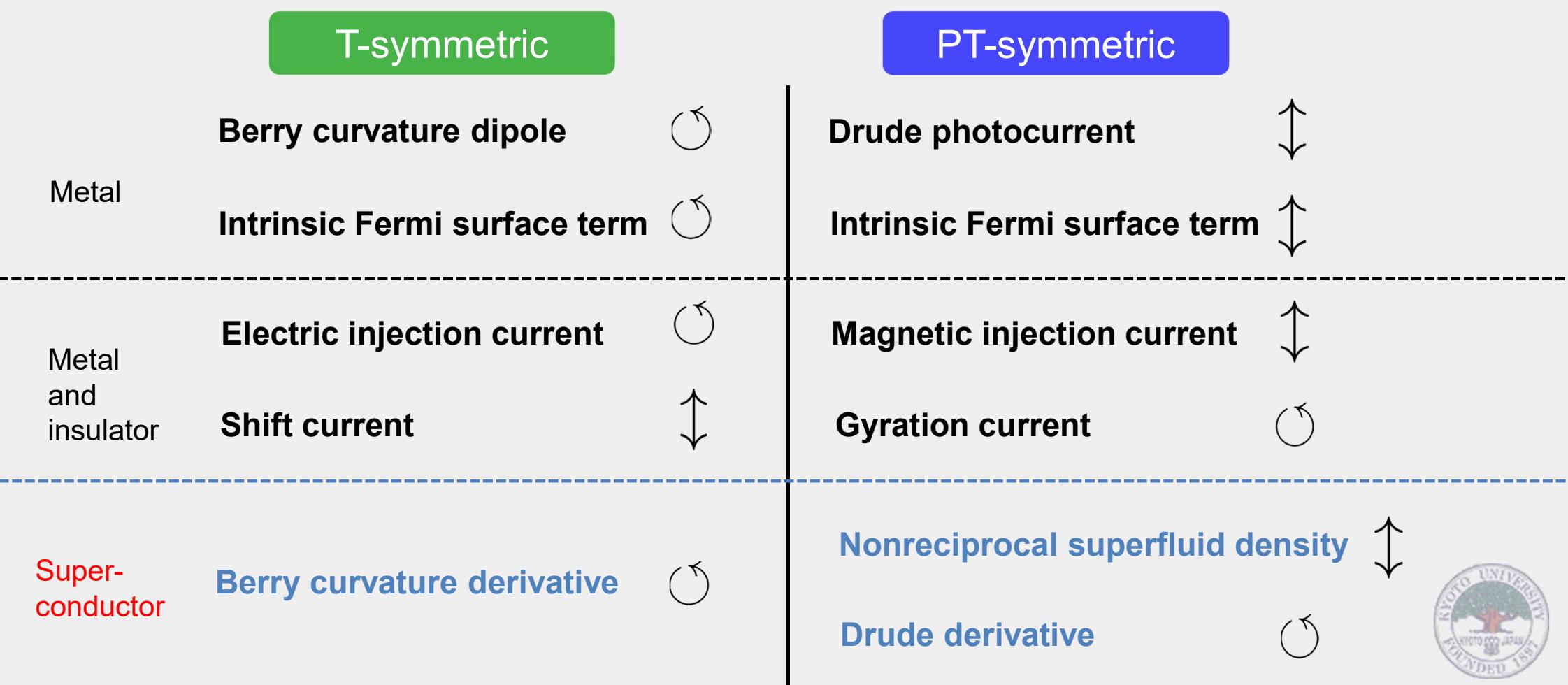
$\sim O(\omega_1^{-1}\omega_2^{-1})$

$\sim O(\omega^{-1})$

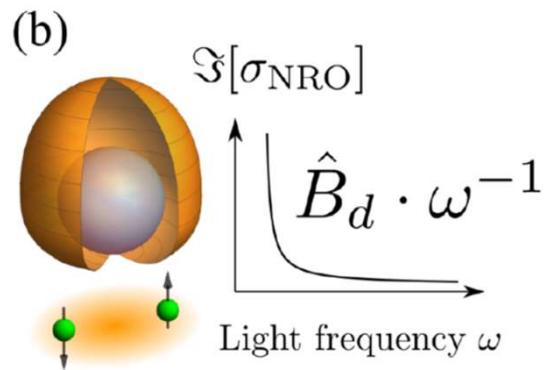
Conductivity
derivative

$$\sigma_{\text{sCD}}^{\alpha;\beta\gamma} = -\frac{i}{4} \left(\frac{1}{\omega_2} \left(D_d^{\beta;\alpha\gamma} + \epsilon_{\alpha\gamma\delta} B_d^{\beta\delta} \right) + \frac{1}{\omega_1} \left(D_d^{\gamma;\alpha\beta} + \epsilon_{\alpha\beta\delta} B_d^{\gamma\delta} \right) \right)$$

Photocurrent in superconductors [Watanabe, Daido, YY, PRB 105, 024308 (2022)]

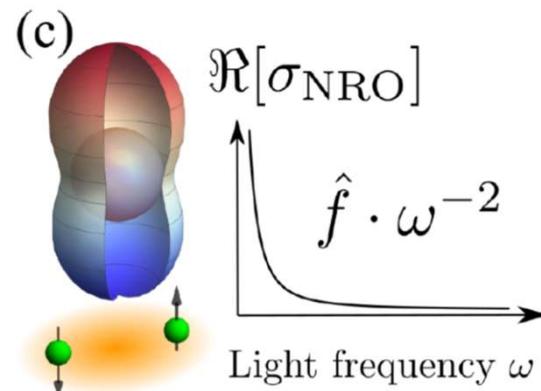


Giant nonreciprocal optical responses in superconductors



$$\sigma_{\text{sCD}}^{\alpha;\beta\gamma} = \lim_{\lambda \rightarrow 0} \frac{i}{4\Omega} \epsilon_{\beta\gamma\delta} \partial_{\lambda_\alpha} \left(\sum_a \Omega_a^{\lambda_\delta} f_a \right) = \hat{B}_d$$

Berry curvature derivative



$$\sigma_{\text{NRSF}}^{\alpha;\beta\gamma} = \lim_{\lambda \rightarrow 0} -\frac{1}{2\Omega^2} \partial_{\lambda_\alpha} \partial_{\lambda_\beta} \partial_{\lambda_\gamma} F_\lambda, = \hat{f}$$

Nonreciprocal superfluid weight

Super-conductor

Berry curvature derivative



Nonreciprocal superfluid density

Drude derivative

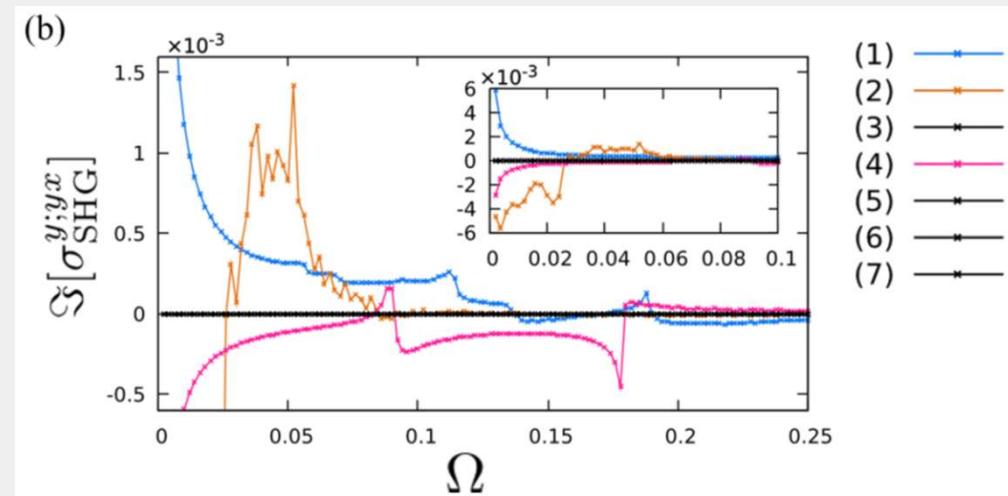


Microscopic conditions [Tanaka, Watanabe, and YY, arXiv:2205.14445]

2D noncentrosymmetric superconductors

$$H_N(\mathbf{k}) = \xi(\mathbf{k}) + \underline{\alpha g(\mathbf{k}) \cdot \sigma} \quad \text{ASOC}$$

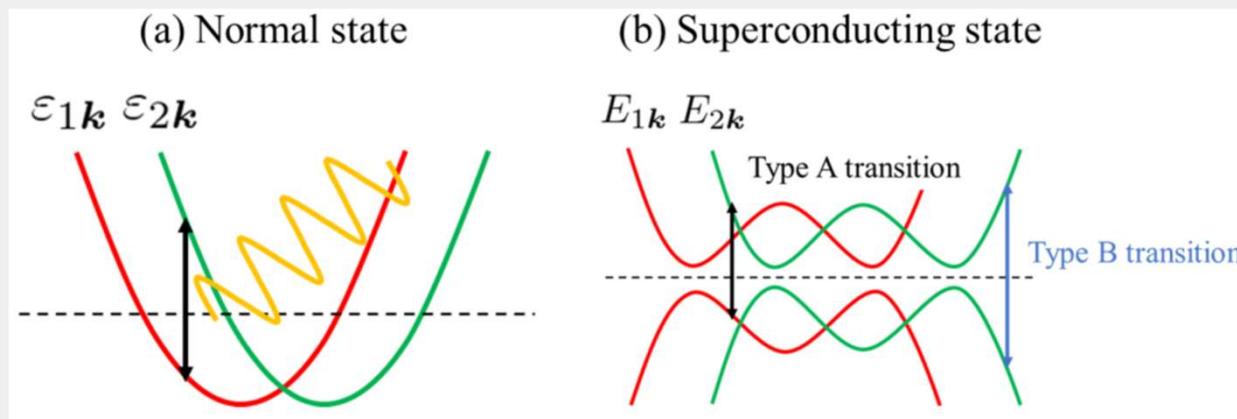
$$\Delta(\mathbf{k}) = [\psi(\mathbf{k}) + d(\mathbf{k}) \cdot \sigma] i\sigma_y \quad \text{Parity mixing}$$



No.	$(\alpha_1, \alpha_2, \alpha_3)$	ψ_0	(d_1, d_2, d_3)	$d_{\mathbf{k}} \cdot g_{\mathbf{k}}$	$d_{\mathbf{k}} \times g_{\mathbf{k}}$	$(\partial_x g_{\mathbf{k}} \times \partial_y g_{\mathbf{k}}) \cdot \hat{d}_{\mathbf{k}}$	Δ_{intra}	Δ_{inter}	response
1	(0.3, 0.8, 0.1)	0.07	(0.04, 0.02, 0.05)	nonzero	nonzero	nonzero	nonzero	nonzero	nonzero
2	(0.3, 0.8, 0.1)	0	(0.04, 0.02, 0.05)	nonzero	nonzero	nonzero	nonzero	nonzero	nonzero
3	(0.2, 0.9, 0.1)	0.01	(0.06, 0.05, 0.03)	nonzero	nonzero	zero	nonzero	nonzero	zero
4	(0.7, 0.2, 0.3)	0.09	(0.03, 0, -0.07)	zero	nonzero	nonzero	nonzero	nonzero	nonzero
5	(0.7, 0.2, 0.3)	0	(0.03, 0, -0.07)	zero	nonzero	nonzero	zero	nonzero	zero
6	(0.4, 0.1, 0.6)	0.09	(0, 0, 0)	zero	zero	undefined	nonzero	zero	zero
7	(0.3, 0.2, 0.6)	0	(0, 0, 0)	zero	zero	undefined	zero	zero	zero



Microscopic conditions [Tanaka, Watanabe, YY, arXiv:2205.14445]



- Type B transition causes the nonlinear optical responses unique to superconductors
- Intraband Cooper pairs and interband Cooper pairs are necessary

1. Finite intraband pairing $\text{Tr} [F_A(\mathbf{k})^\dagger F_A(\mathbf{k})] = 4 [(\alpha \mathbf{g}_k \cdot \mathbf{d}_k)^2 + \psi_k^2 \alpha^2 g_k^2] \neq 0$ Superconducting fitness [Ramires, 2018]
2. Finite interband pairing $\text{Tr} [F_C(\mathbf{k})^\dagger F_C(\mathbf{k})] = 8\alpha^2 |\mathbf{g}_k \times \mathbf{d}_k|^2 \neq 0$

Conclusion:

Spin-triplet Cooper pairs are necessary for SC nonlinear optical responses.



Summary

Intrinsic SC diode effect

A. Daido, Y. Ikeda, and YY

[Phys. Rev. Lett. 128, 037001 \(2022\)](#)

A. Daido and YY, arXiv:2209.03515

- **Nonreciprocal depairing critical current**
 - **Large Δj_c** at low T & moderate h_y
 - **Sign reversals** as $h_y \uparrow$
- **Promising probe of finite-q helical SC**

SC nonreciprocal optics

H. Watanabe, A. Daido, and YY,

[Phys. Rev. B 105, 024308 \(2022\)](#)

[Phys. Rev. B 105, 024308 \(2022\)](#)

Tanaka, Watanabe, and YY, [arXiv:2205.14445](#)

- **Nonlinear optical responses unique to SC**
 - **Diverging photocurrent and SHG at sub THz**
 - **Sensitive to quantum geometry**
- **Promising probe of parity-mixed Cooper pairs**