Multicomponent superconductivity and quadrupling condensates

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李政道研究所 Tsung-Dao Lee Institute

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Outline of the course

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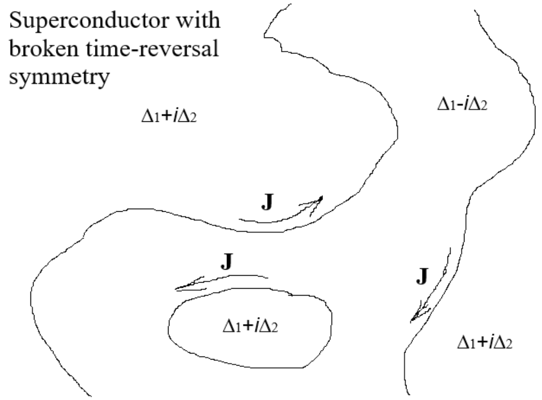
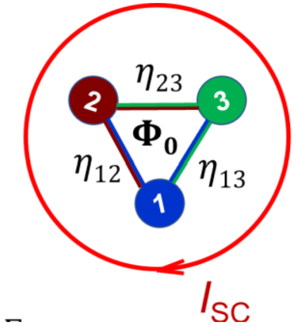
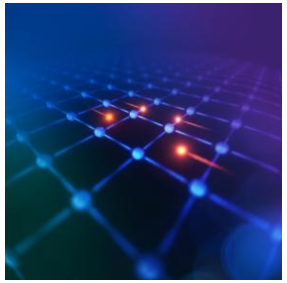
1. Superconductivity that breaks time reversal symmetry.



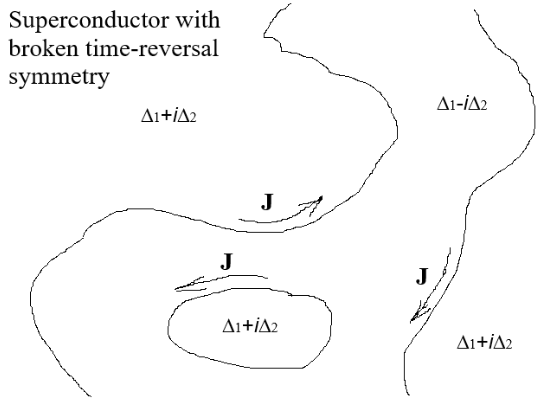
2. Flux quantum fractionalization and unquantized vortices in multicomponent superconductors.



3. Electron-Quadrupling condensates in multicomponent superconductors with strong phase fluctuations.



Lecture I - Superconductivity that breaks time reversal symmetry



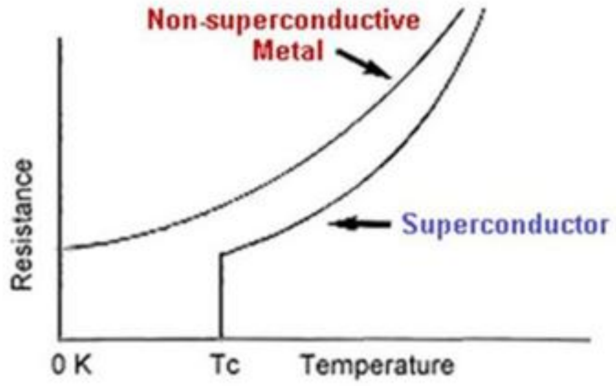
The aim of the first lecture:

1. To introduce the concept of multicomponent superconductivity with broken time reversal symmetry.
2. To introduce experimental techniques that are capable of detecting this type of superconductivity.
3. To discuss the materials in which these states were detected or expected.

Fundamental properties of superconductors

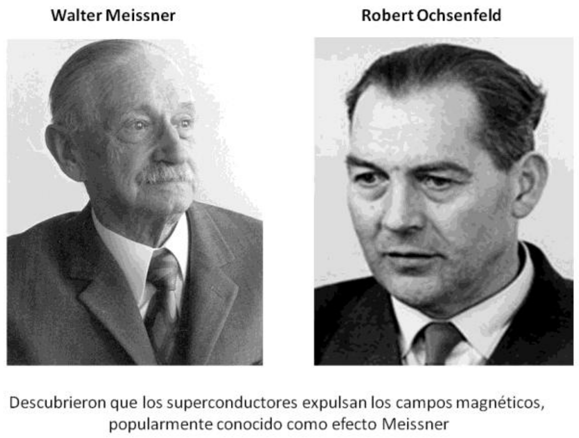
- **Superconductivity is a unique state of matter characterized by zero electrical resistance $\rho_{SC} < 10^{-24}$ Ω·cm (for example, $\rho_{SC} \sim 10^{-9}$ Ω·cm for pure Cupper at T = 4.2 K (liquid He temperature)).**

Heike Kamerlingh Onnes

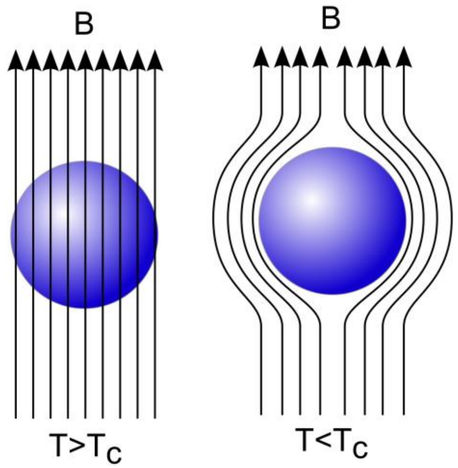


Superconductivity (zero resistivity) was discovered in 1911

- **Superconductors repulse magnetic field – ideal diamagnetic materials (Meissner effect).**



Walther Meißner and his PhD student Robert Ochsenfeld



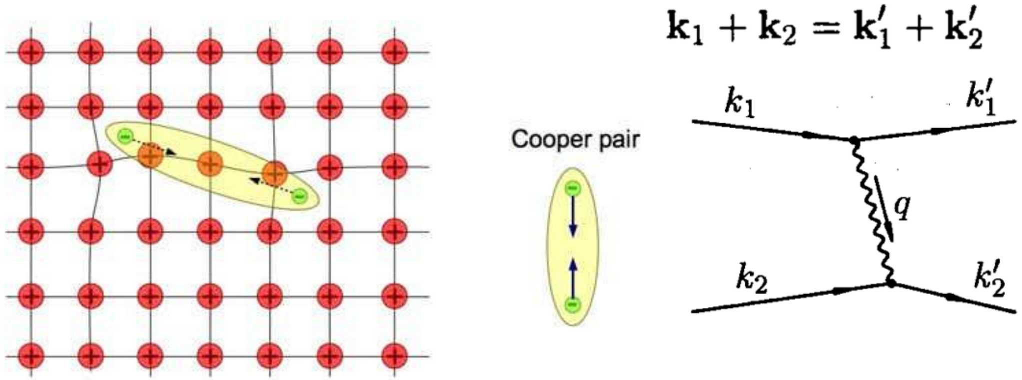
Meissner effect was discovered in 1933

Superconductors are "antagonistic" to magnetism!

https://en.wikipedia.org/wiki/Meissner_effect

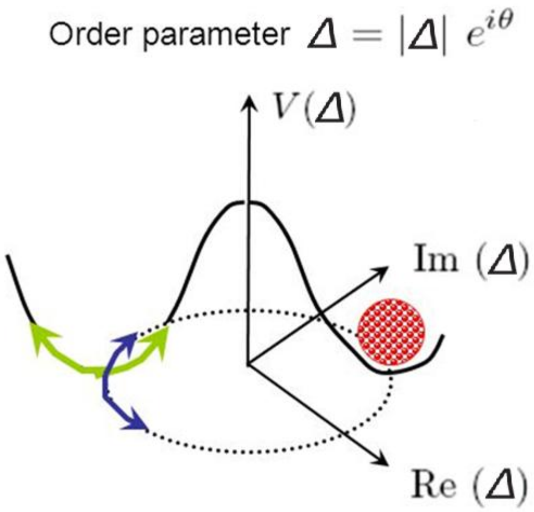
Cooper pairs, superconducting order parameter in Bardeen–Cooper–Schrieffer (BCS) theory

- Pairing of two electrons with opposite spins via exchange of phonons.
- In superconductors, the size of a Cooper pair is large compared to the crystal lattice unite cell.
- Cooper pair is therefore a correlated state of two electrons.



The first was described in 1956 by Leon Cooper.

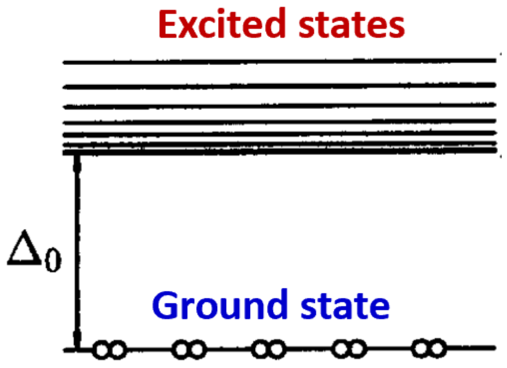
- The formation of electron pairs results in a phase transition at T_c with the order parameter ***Δ = |Δ|e^iφ^, which has amplitude and phase***.
- Below T_c, the state is characterized by an ordered ***φ*** resulting in spontaneous breaking of **U1 gauge symmetry.**
- ***|Δ|** characterizes how strong the symmetry is broken below transition temperature and is the energy characteristic of the state.*



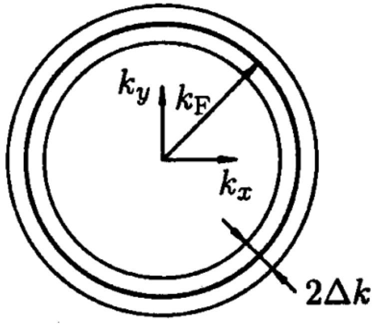
https://slideplayer.com/slide/5363094/

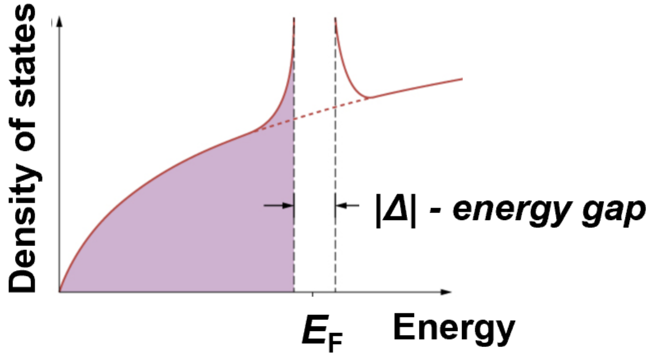
Cooper pairs, superconducting order parameter in Bardeen–Cooper–Schrieffer (BCS) theory

- |Δ| corresponds to the energy gap between the ground state and exited state.
- The electrons interacts only in the vicinity of the Fermi energy.
- For a circular Fermi surface with electron-phonon interaction the gap has an “*s*-wave” symmetry (in analogy for orbitals in atom).

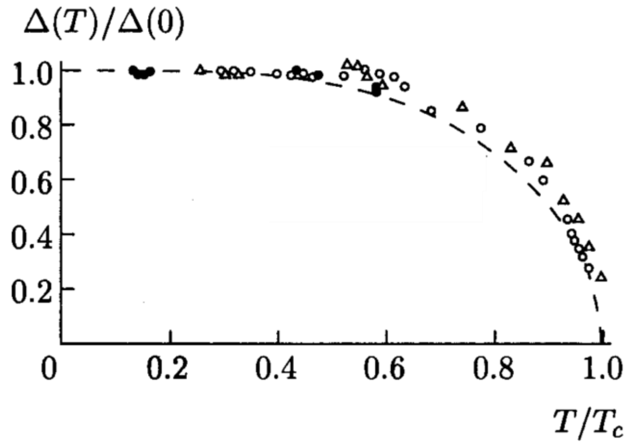


Momentum-independent interaction of the electrons in the vicinity of $E_F = \frac{k_F^2 \hbar^2}{2m}$





- The gap in the density of states can be measured in the experiment and is temperature dependent.

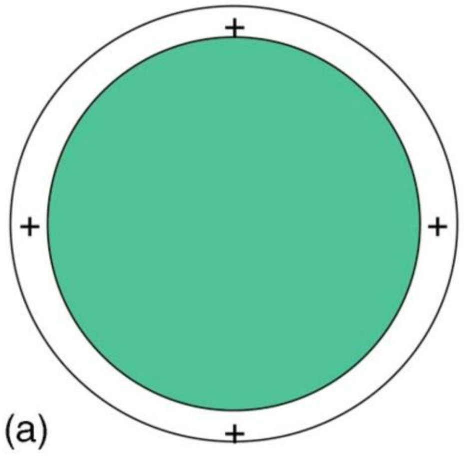


Dots - experimental data, dashed line – BSC theory (1957)

V. V. Sshmidt, “Physics of superconductors” (1997)

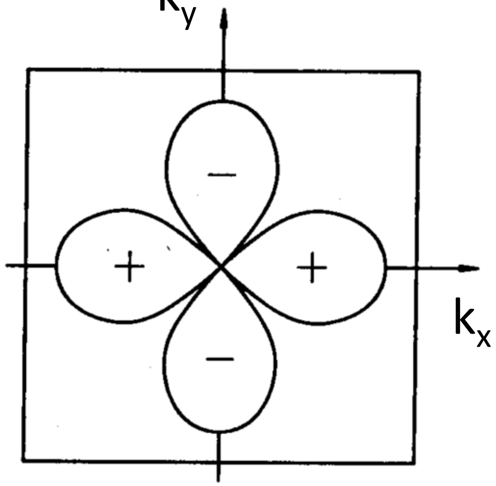
Superconducting order parameter symmetry

Dependence of the order parameter on the direction of the Cooper pair momentum on the Fermi surface



An *s*- wave order parameter: independent on momentum

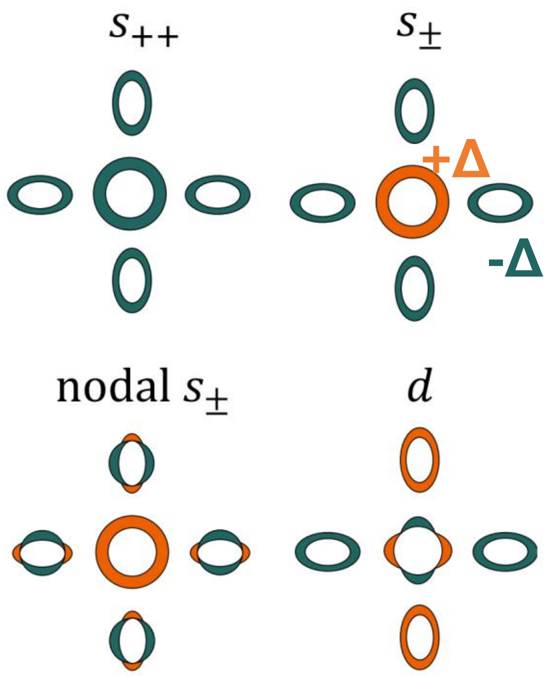
Rev. Mod. Phys., 75, (2003)



$$\Delta_{\mathbf{k}} = \Delta_0 \cos 2\theta_{\mathbf{k}}$$

An *d*- wave order parameter: has 4 nodes

Complex Fermi surface



Rep. Prog. Phys. **74** (2011) 124508

- **The order parameter symmetry is named after the symmetry of the atomic orbitals: *s*, *p*, *d*, etc;**
- **An s-wave superconductivity is more common in nature- conventional superconductivity;**
- **Other than an *s*-wave type of the order parameter imply unconventional superconductivity.**

Ginzburg-Landau (GL) theory the first phenomenological theory of superconductivity

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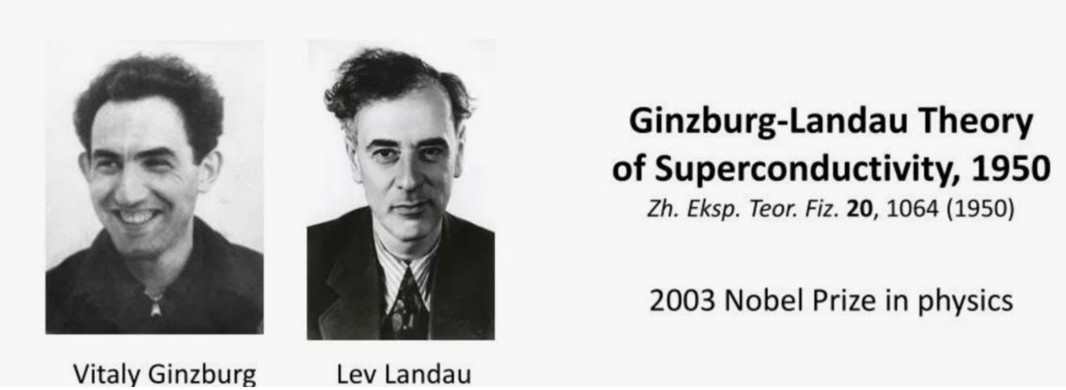
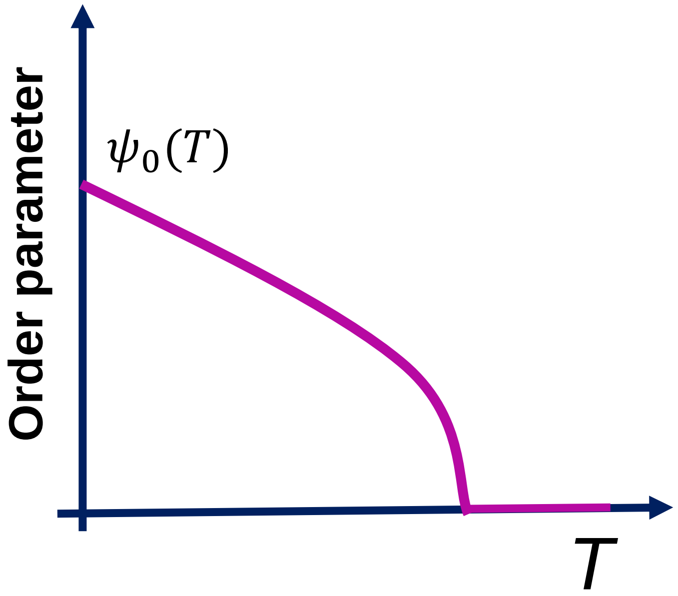
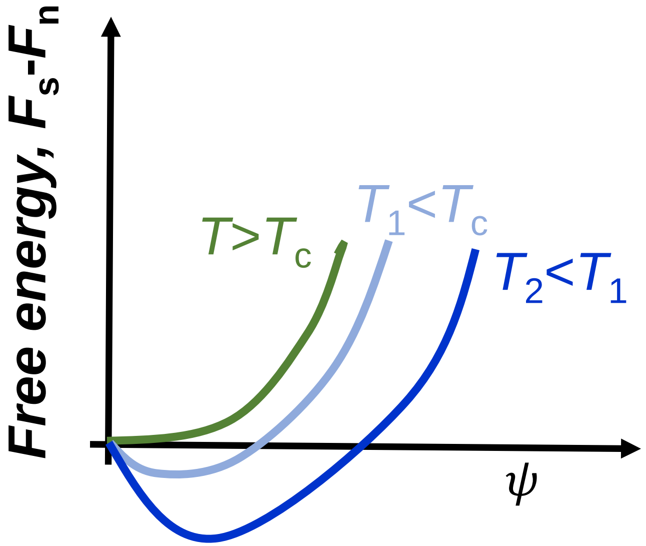
- ✓ The theory assumes that superconducting state is more ordered compared to the normal state and the transition to superconducting state (in zero magnetic field) is a second order transition.
- ✓ In the GL theory the field $\psi(\boldsymbol{r})$ (made of Cooper pairs) takes a role of the order parameter.



Superconducting transition is continuous at T_c but results in spontaneous broken U(1) guage symmetry





$$\psi_0 = -\alpha / \beta$$

$$|\Psi(\mathbf{r})|^2 = n_s/2.$$

n_s - the density of superconducting electrons

Free energy close to T_c in zero magnetic field, where | $\psi(\boldsymbol{r})$ |<<1

$$F_{s0} = F_n + \alpha|\Psi|^2 + \frac{\beta}{2}|\Psi|^4.$$

$$\frac{dF_{s0}}{d|\Psi|^2} = 0$$

Ginzburg-Landau (GL) theory the first quantum phenomenological theory of superconductivity

For non-homogeneous superconductor in magnetic field, close to T_c free energy can be expressed:

$$\mathcal{G}_{sH} = \mathcal{G}_n + \int \left[\alpha|\Psi|^2 + \frac{\beta}{2}|\Psi|^4 + \frac{1}{4m}\left| -i\hbar\nabla\Psi - \frac{2e}{c}\mathbf{A}\Psi \right|^2 + \frac{(\nabla\times\mathbf{A})^2}{8\pi} - \frac{\nabla\times\mathbf{A}\cdot\mathbf{H}_0}{4\pi} \right] dV$$

$\delta_{\Psi^*}\mathcal{G}_{sH} = 0$

$\delta_{\mathbf{A}}\mathcal{G}_{sH} = 0$

minimization of the energy gives:

$$\alpha\Psi + \beta\Psi|\Psi|^2 + \frac{1}{4m}\left(i\hbar\nabla + \frac{2e}{c}\mathbf{A}\right)^2\Psi = 0$$ - first GL equation

$$\mathbf{j}_s = -\frac{i\hbar e}{2m}(\Psi^*\nabla\Psi - \Psi\nabla\Psi^*) - \frac{2e^2}{mc}|\Psi|^2\mathbf{A}$$ - second GL equation, expression for supercurrent

The one can define:

$$\xi^2 = \frac{\hbar^2}{4m|\alpha|},$$ - GL coherence length

$$\lambda^2 = \frac{mc^2}{4\pi n_s e^2} = \frac{mc^2\beta}{8\pi e^2|\alpha|}.$$ - magnetic penetration depth

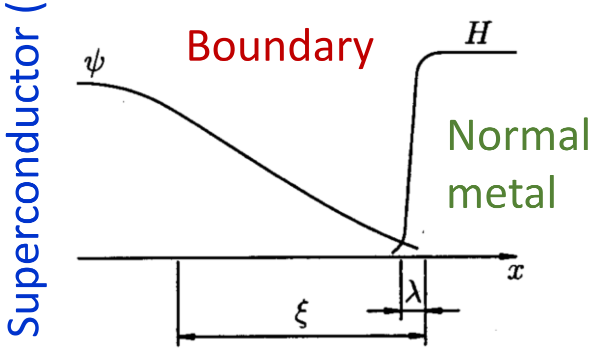
$\Phi_0 = \pi\hbar c/e$ - flux quantum

$$\xi^2\left(i\nabla + \frac{2\pi}{\Phi_0}\mathbf{A}\right)^2\psi - \psi + \psi|\psi|^2 = 0$$

$$\mathbf{j}_s = -i\frac{\Phi_0}{4\pi\lambda^2}(\psi^*\nabla\psi - \psi\nabla\psi^*) - \frac{|\psi|^2}{\lambda^2}\mathbf{A}.$$

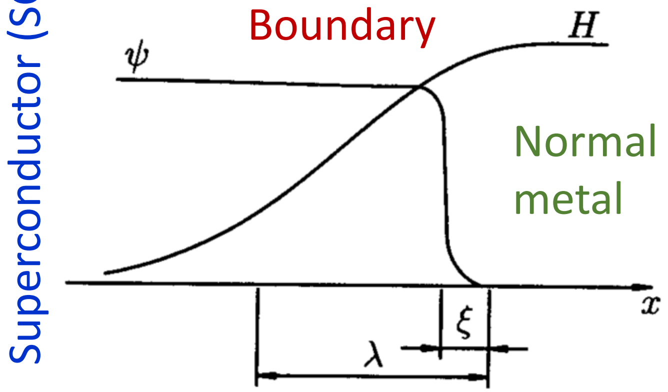
Type-I superconductor

$F_n - F_{SC} \propto \xi H_{cm}^2$



Type-II superconductor

$F_n - F_{SC} \propto \lambda H_{cm}^2$

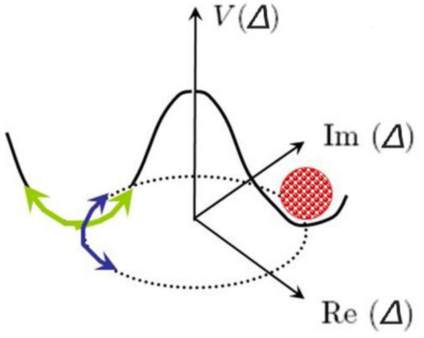


H_{cm} - thermodynamic critical field

Superconductors with broken time-reversal symmetry

At T_c **broken $U(1)$**

Order parameter $\Delta = |\Delta|\, e^{i\theta}$



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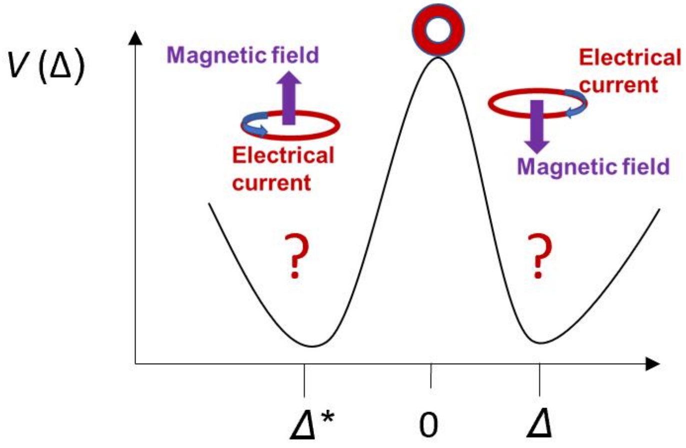
The fundamental symmetry which spontaneously violates by any superconducting state.

and

$T(Z_2)$ – time reversal symmetry: $\Delta \xrightarrow{T} \Delta^* \neq |\Delta_\alpha| e^{i\theta}$



Two energetically equivalent states with $\Delta \neq \Delta^*$ meaning spontaneously broken Z_2 symmetry.

- **Broken time-reversal symmetry (BTRS) state can be described by two component imaginary order parameter: $\Delta_1 \pm i\,\Delta_2$.**
- **Two different phase transitions → $T_c \neq T_{BTRS}$ and two different coherence lengths $\xi_{U(1)} \neq \xi_{Z2}$.**
- The Broken Time-Reversal Symmetry state results in the appearance of spontaneous currents and magnetic fields.

Where these currents come from?

Superconductors that break time-reversal symmetry (BTRS)

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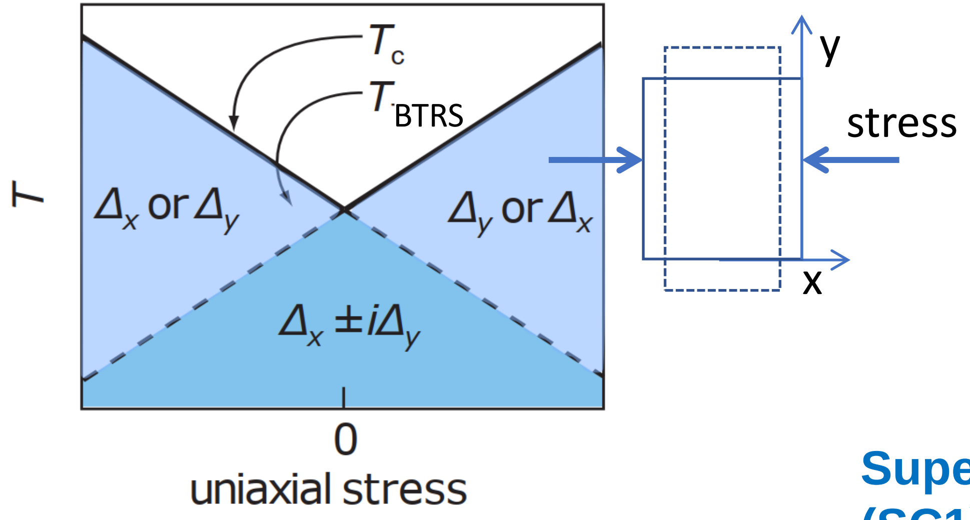
***Two-component** order parameter* $|\Delta_{1(2)}|e^{i\varphi 1(2)}$ **with double degenerate states:**

$$\{|\Delta_1|, |\Delta_2|, \varphi_1 - \varphi_2 = \theta_{12}\} \rightarrow \{|\Delta_1|, |\Delta_2|, \varphi_1 - \varphi_2 = -\theta_{12}\}$$

Degenerate (symmetry protected) order parameters

Intrinsically $|\Delta_1| = |\Delta_2|$, $\theta_{12} = \pm\pi/2$ ⟹ $T_c = T_{BTRS}$

Examples: $p_x \pm ip_y$, $d_{xz} \pm id_{yz}$, …



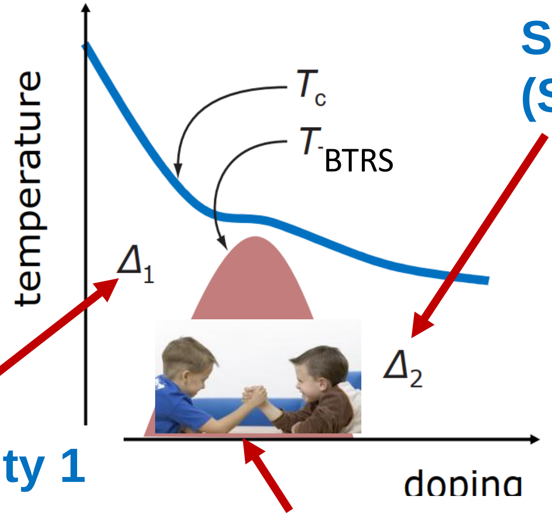
Lifting degeneracy by strain

V. Grinenko, S. Ghosh *et al.* *Nat. Phys.* **17,** 748–754 (2021).

Non-degenerate or accidentally degenerate order parameter

$|\Delta_1| \neq |\Delta_2|$, $\theta_{12} \neq 0,\pi$ ⟹ fine tuning for $T_c = T_{BTRS}$

Examples: $s \pm is'$, $s \pm id$, $d \pm ig$, …



Superconductivity 2 (SC2)

Superconductivity 1 (SC1)

Competition between SC1 and SC2

The degeneracy is lifted by any perturbation.

V. Grinenko *et al.*, *Nat. Phys.* **17,** 1254–1259 (2021).

Spontaneous currents appear as a result of a non-trivial phase difference between the components of the order parameter (internal Josephson effect).

What is the internal Josephson effect?

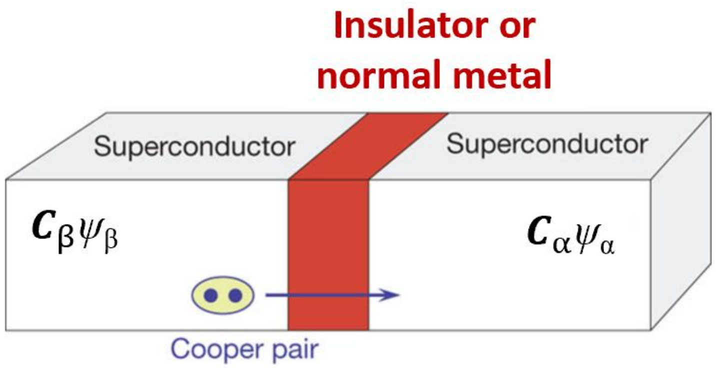
Phase of the order parameter

Phase of the order parameter φ is a key to define the properties of a superconductor and can be measured directly in the phase sensitive experiments such as Josephson effect.

Brian David Josephson

1962

Josephson junction between 2 superconductors



Nature 474, 589–597 doi:10.1038/nature10122

The evolution of the system can be described by time-dependent Schrodinger equation

$$i\hbar \frac{\partial \Psi}{\partial t} = \widehat{H}\Psi \quad \text{with a wave function} \quad \Psi(t) = \sum_\alpha C_\alpha(t)\psi_\alpha$$

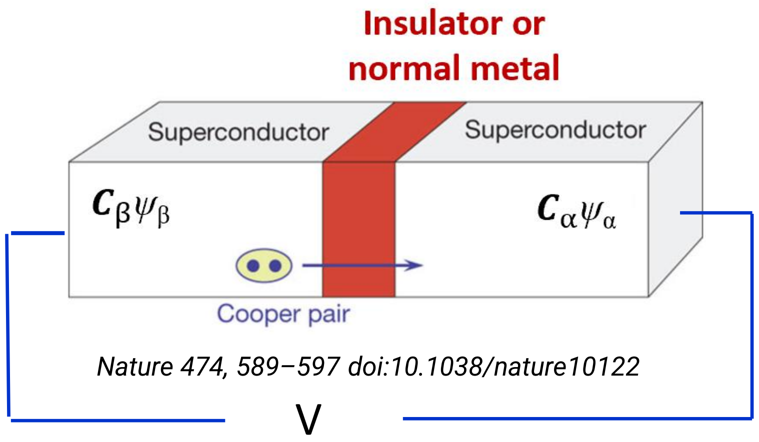
$$i\hbar \frac{dC_\beta}{dt} = \sum_\alpha H_{\beta\alpha} C_\alpha(t). \quad \text{with} \quad H_{\beta\alpha} = \int \psi_\beta^* \widehat{H} \psi_\alpha \, dV.$$

- ✓ Here we assume that the system can be in two discrete state ψ_α and ψ_β.
- ✓ $H_{\beta\beta}$ is the energy of the system at state ψ_β;
- ✓ $H_{\beta\alpha}$ is the matrix element characterizing the transition of the system from the state ψ_β to the state ψ_α ;
- ✓ C_α is the amplitude of the state ψ_α and $| C_\alpha |$ is the probability to find the system in the state ψ_α .

Phase of the order parameter

Phase of the order parameter φ is a key to define the properties of a superconductor and can be measured directly in the phase sensitive experiments such as Josephson effect.

Josephson junction between 2 superconductors

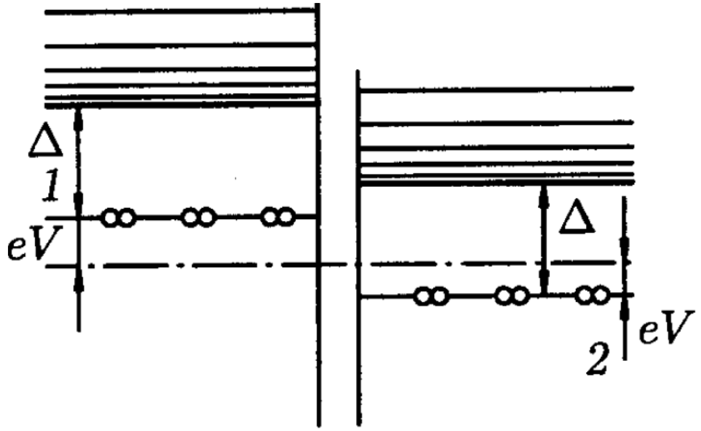


Nature 474, 589–597 doi:10.1038/nature10122

$$i\hbar \frac{dC_1}{dt} = eVC_1(t) + KC_2(t),$$
$$i\hbar \frac{dC_2}{dt} = KC_1(t) - eVC_2(t).$$

K is tunneling from energy level 1 to the energy level 2

Energetical equivalent schema of the junction



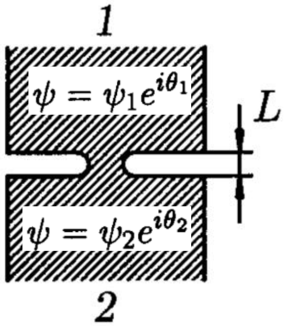
$C_\alpha = \sqrt{n_\alpha} e^{-i\varphi_\alpha}$ with n_α as a density of superconducting electrons at the level 1 and 2

$$J_s \propto \frac{dn_s}{dt} = \frac{2Kn_s}{\hbar} \sin(\varphi_2 - \varphi_1)$$

- Josephson current is propositional to the phase difference between left and right

Phase of the order parameter GL theory

It is useful to consider the Ginzburg-Landau (GL) theory to describe the Josephson effect.



$L \ll \xi$

$-cocherence\ length$

$$\xi^2 \left(i\nabla + \frac{2\pi}{\Phi_0}\mathbf{A} \right)^2 \psi - \psi + \psi|\psi|^2 = 0$$

If $L \ll \xi$ then $\nabla^2\psi \sim \psi/L^2$. Since $|\psi| \sim 1$ in the bulk, one get $\nabla^2 f(r) = 0$.

With a possible solution: $\psi = \psi_1 e^{i\theta_1} f(r) + \psi_2 e^{i\theta_2}(1 - f(r))$ where $f(r) = 1$ in 1 and $f(r) = 0$ in 2.

$$\mathbf{j}_s = -i\frac{\Phi_0}{4\pi\lambda^2}(\psi^*\nabla\psi - \psi\nabla\psi^*) - \frac{|\psi|^2}{\lambda^2}\mathbf{A}$$

- expression for supercurrent

In zero field and for θ_1 and θ_2 are independent on ***r*** we can get

$$\mathbf{j}_s = \frac{\Phi_0}{2\pi\lambda^2}\,\mathrm{Im}\left(\psi_1\psi_2^* e^{-i(\theta_2-\theta_1)}\right) f'(r)\hat{\mathbf{r}} = \frac{\Phi_0}{2\pi\lambda^2}\left(-\psi_1\psi_2\sin(\theta_2-\theta_1)\right) f'(r)\hat{r}$$

- the result depends on the phase difference between left and right and the gradient of the amplitude.

For $\theta_1(r)$ and $\theta_2(r)$.

$$\mathbf{j}_s = \frac{\Phi_0}{2\pi\lambda^2}\left[\psi_1^2 f(r)\nabla\theta_1 + \psi_2^2(1-f(r))\nabla\theta_2 + \psi_1\psi_2\sin(\theta_2-\theta_1)f'(r)\hat{r}\right]$$

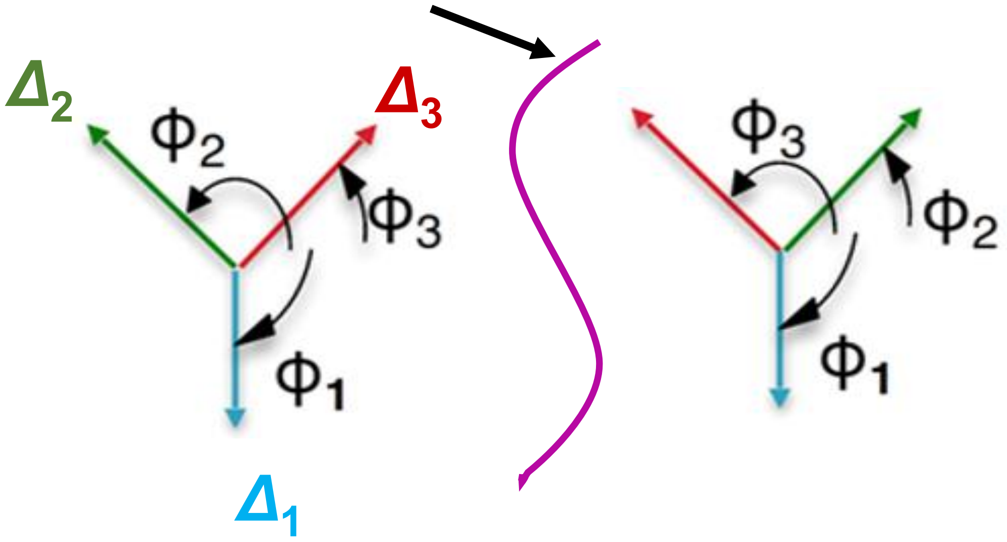
- this adds gradients of the phases

Currents in a superconductor appear spontaneously when there is a variation of phase and amplitude of the order parameter!

Phase of the order parameter GL theory

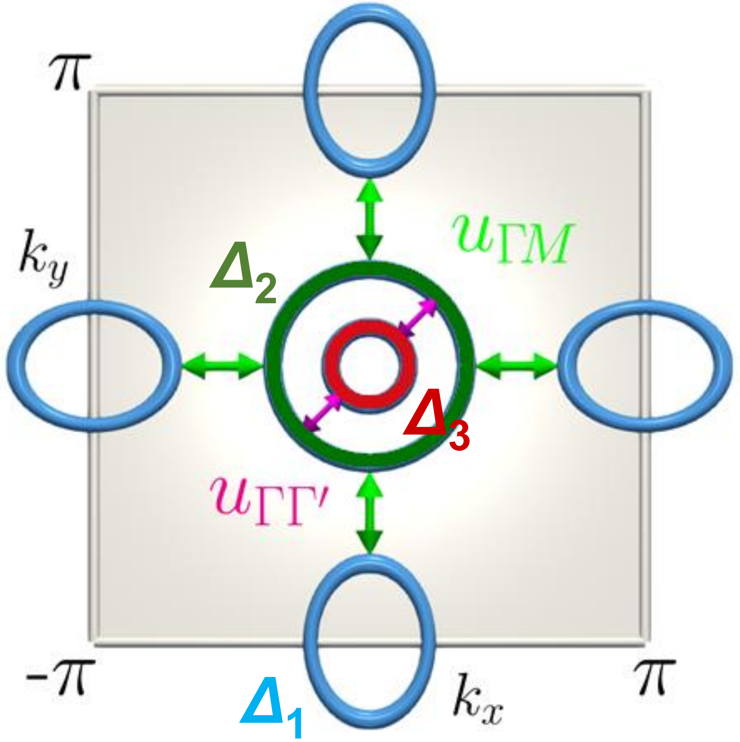
Now we can understand what will happened if there is a superconductor with a multicomponent order parameter in the real space $\psi_\alpha = |\Delta_\alpha| e^{i\Phi_\alpha}$ **where α= 1,2,3 and** $\Phi_\alpha - \Phi_\beta = \Phi_{\alpha\beta}$

Domain wall between regions of different chirality of the phase



(s-is)	**(s+is)**
m = 1	*m* = -1

The order parameter components are separated in the momentum space.



Z_2 Ising order parameter *m* characterizes an inter-band phase locking

Ginzburg-Landau description of the multicomponent s+is state

Free energy for a multicomponent order parameter $\psi_\alpha = |\Delta_\alpha| e^{i\Phi_\alpha}$ **where** $\alpha = 1,2,3$ **and** $\Phi_\alpha - \Phi_\beta = \varphi_{\alpha\beta}$ (an *s*+is' sate).

$$f = \frac{1}{2\rho^2}\left[\left(\sum_{i=1,2,3} |\Delta_i|^2 \vec{\nabla}\phi_i\right) - e\rho^2 \vec{A}\right]^2 + \sum_{i>j} \frac{|\Delta_i|^2 |\Delta_j|^2}{2\rho^2}\left[\vec{\nabla}(\phi_i - \phi_j)\right]^2 + \sum_{i>j} \eta_{ij} |\Delta_i||\Delta_j| \cos(\phi_i - \phi_j) + \sum_i V(|\Delta_i|^2, |\Delta_i|^4) + \frac{\mathbf{B}^2}{2}, \text{ where } \rho^2 = \Delta_1^2 + \Delta_2^2 + \Delta_3^2$$

SC

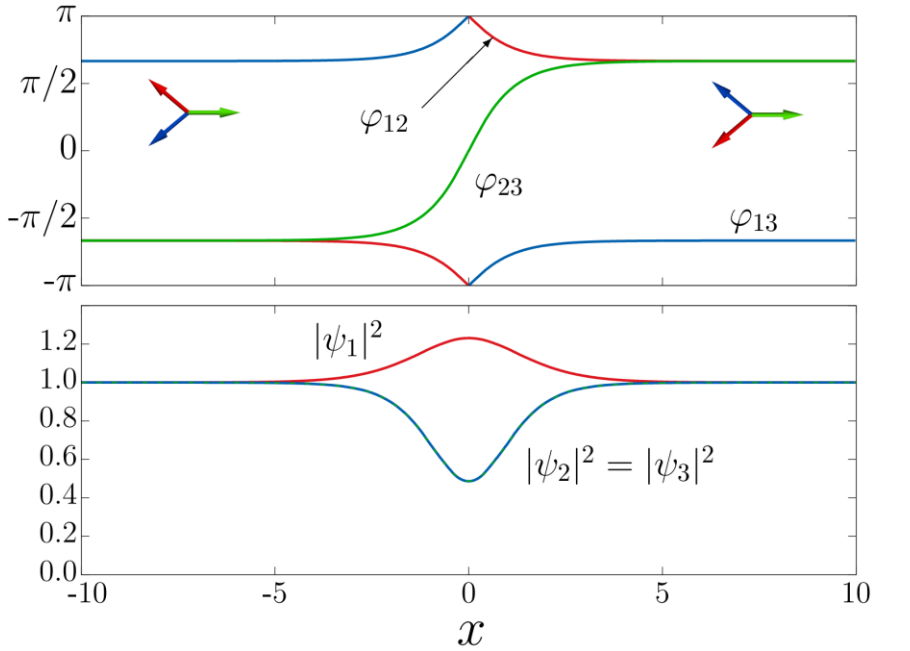
TRSB

Internal Josephson coupling

Case	Sign of $\eta_{12}, \eta_{13}, \eta_{23}$	Ground State Phases
1	s_{++} $- - -$	$\phi_1 = \phi_2 = \phi_3$
2	s+is' $- - +$	Frustrated
3	$s_{\pm}$ $- + +$	$\phi_1 = \phi_2 = \phi_3 + \pi$
4	s+is' $+ + +$	Frustrated

V Stanev, Z Tešanović PRB 81 (13), 134522 (2010)
J Carlström, J Garaud, E Babaev PRB 84 (13), 134518 (2011)
J Garaud, M Silaev, E Babaev Physica C: 533, 63-73 (2017)
…

Change of the phases and amplitudes at the domain walls

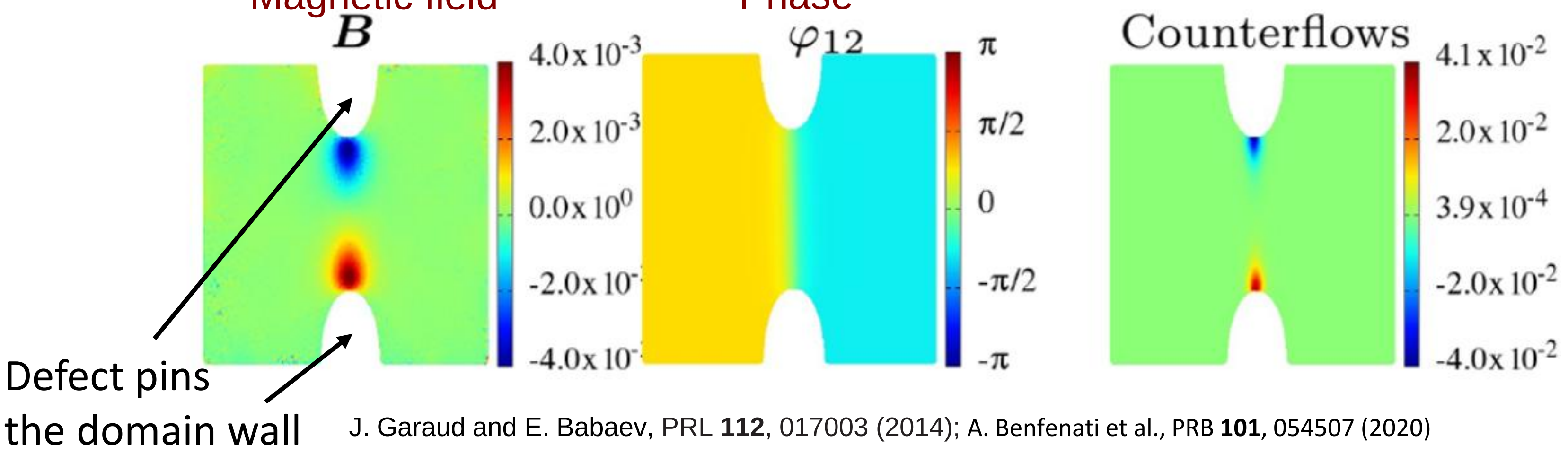


Julien Garaud, thesis, 2022

We also assume that $|\Delta_\alpha|$ = ***const*** but this is not the case on the domain walls or around inhomogeneities!

This *s+is'* state breaks time reversal symmetry and induces fields at domain walls and around inhomogeneities.

Spontaneous currents in *s+is* superconductor



J. Garaud and E. Babaev, PRL **112**, 017003 (2014); A. Benfenati et al., PRB **101**, 054507 (2020)

$$B_k = \partial_l A_m - \partial_m A_l = -\epsilon_{lm}\partial_l\left(\frac{J_m}{e|\Psi|^2}\right) \boxed{-\frac{i\epsilon_{lm}}{e^2|\Psi|^4}\left[|\Psi|^2\partial_l\Psi^\dagger\partial_m\Psi + \Psi^\dagger\partial_l\Psi\partial_m\Psi^\dagger\Psi\right]}$$

, where $\varepsilon_{ij} = \begin{cases} +1 & \text{if } (i,j)=(1,2) \\ -1 & \text{if } (i,j)=(2,1) \\ 0 & \text{if } i=j \end{cases}$

The spontaneous fields originate from a second term having the **Skyrmionic form** of CP^2 topological charge density. [E. Babaev, L. D. Faddeev, and A. J. Niemi, Phys. Rev. B65, 100512 (2002).]

In 2- band case: $\psi = (\psi_1, \psi_2)$

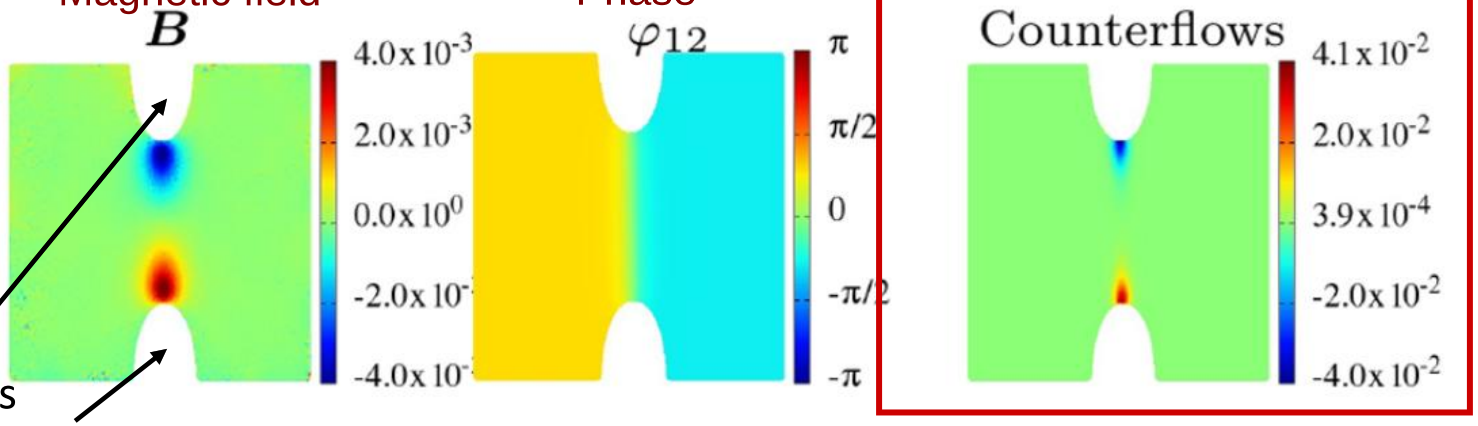
$$\mathbf{B} = \mathrm{curl}\,\mathbf{A} = \mathrm{curl}\frac{i}{e^2|\psi|^2}[(\psi_1\nabla\psi_1^* - \psi_1^*\nabla\psi_1) + (\psi_2\nabla\psi_2^* - \psi_2^*\nabla\psi_2)] - \mathrm{curl}\frac{1}{e|\psi|^2}\mathbf{J}.$$

B contains products of density gradients and gradients of the phases of the form

$$\partial_i\left(\frac{|\psi_\nu|^2}{|\psi_\nu|^2+|\psi_\mu|^2}\right)\partial_j\theta_\nu.$$

Spontaneous currents in *s+is* superconductor

Magnetic field Phase



Defect pins
the domain wall

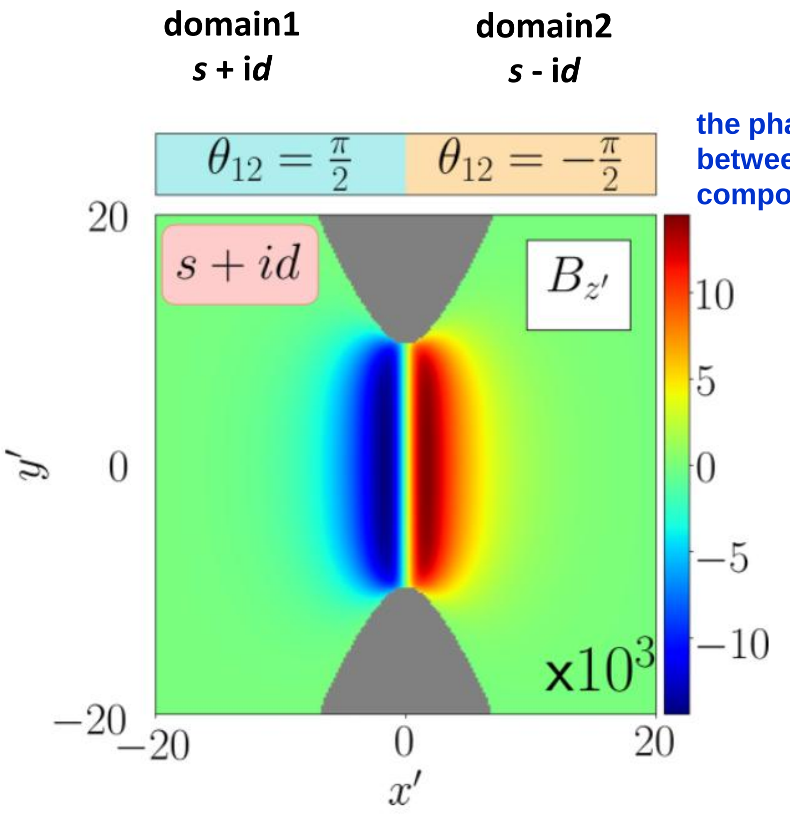
J. Garaud and E. Babaev, PRL **112**, 017003 (2014); A. Benfenati et al., PRB **101**, 054507 (2020)

$$B_k = \partial_l A_m - \partial_m A_l = -\epsilon_{lm}\partial_l\left(\frac{J_m}{e|\Psi|^2}\right) - \frac{i\epsilon_{lm}}{e^2|\Psi|^4}\left[|\Psi|^2\partial_l\Psi^\dagger\partial_m\Psi + \Psi^\dagger\partial_l\Psi\partial_m\Psi^\dagger\Psi\right]$$

The spontaneous fields originate from a second term having the **Skyrmionic form** of CP^2 topological charge density. [E. Babaev, L. D. Faddeev, and A. J. Niemi, Phys. Rev. B65, 100512 (2002).]

- ✓ The fields associated with the magnetic field induced by internal Josephson currents between different bands (in momentum space), in the presence of relative density gradients caused by inhomogeneities.
- ✓ Without inhomogeneities, spontaneous fields are perfectly compensated (counter-flow) since in momentum space all directions are equivalent, and there is no net current transfer.

Spontaneous fields in non-chiral pairing states, domain walls

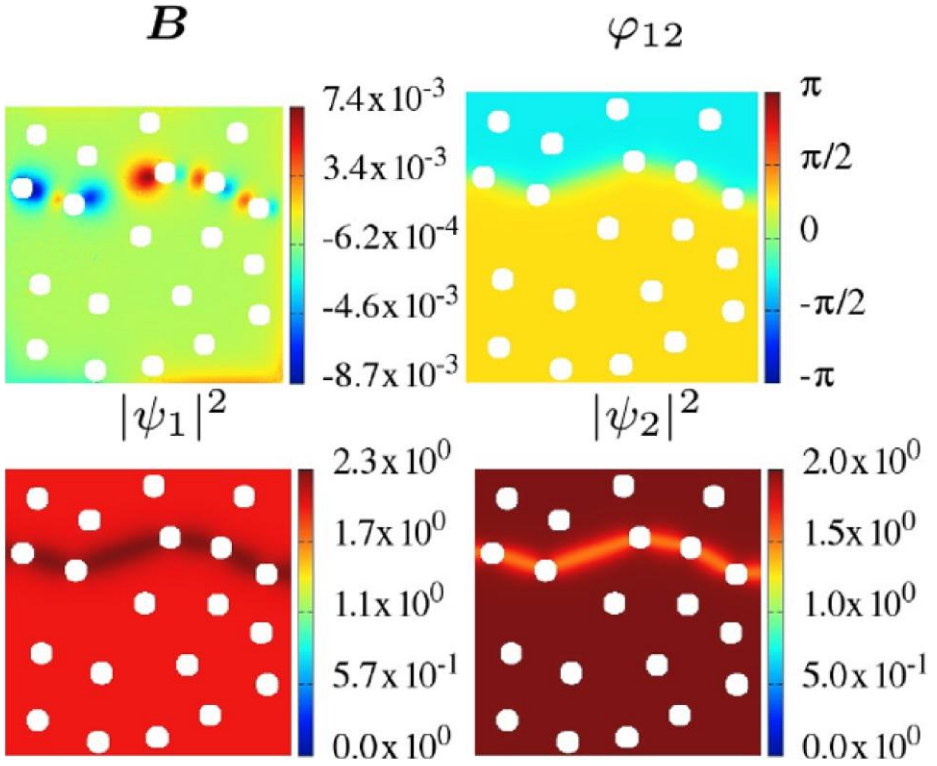


the phase difference between *s* and *d* components

A. Benfenati *et al.*, PRB **101**, 054507 (2020).

Domain boundary pinned by defects produces dipolar spontaneous fields.

A domain wall stabilized by randomly located pinning centers for an *s*+*is* state



J. Garaud and E. Babaev, PRL 112, 017003 (2014)

Dipolar structure of the field → the net magnetic flux is zero

Spontaneous fields in non-chiral pairing states due to inhomogeneity

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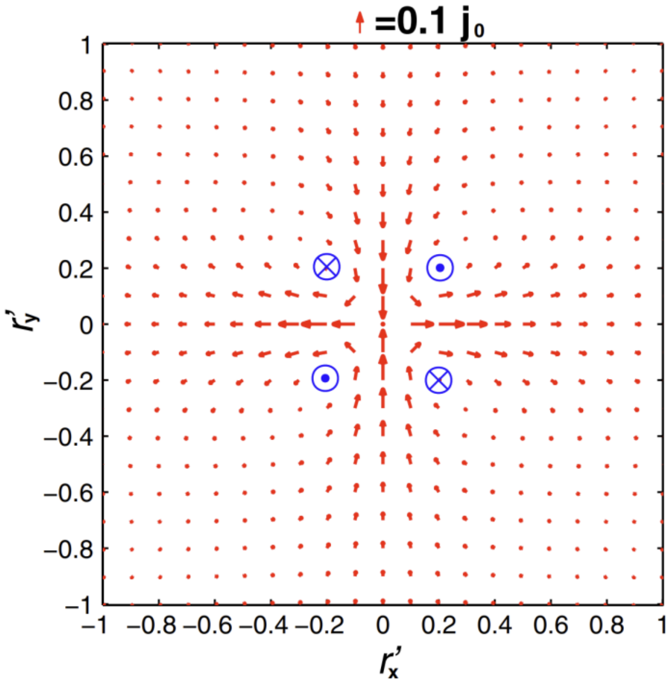
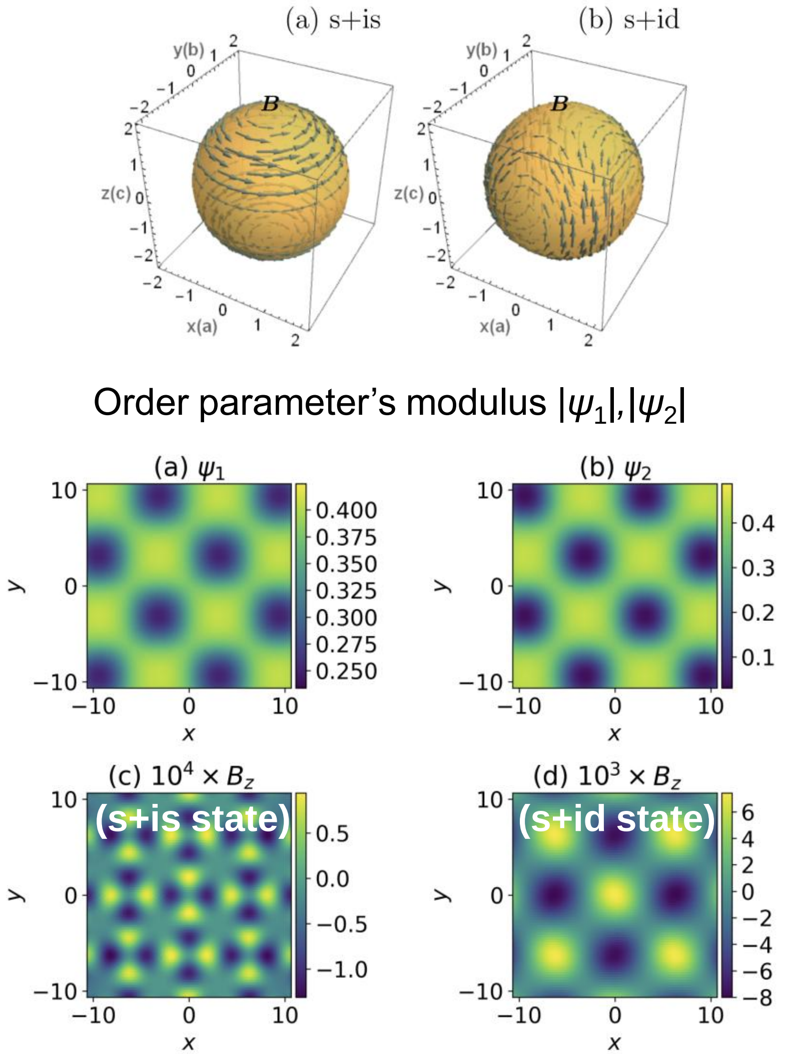
Spontaneous magnetic in *s+id* superconductor



W.-C. Lee, S.-C. Zhang, and C. Wu PRL 102, 217002 (2009)

Spontaneous magnetic field due to spherical inhomogeneity

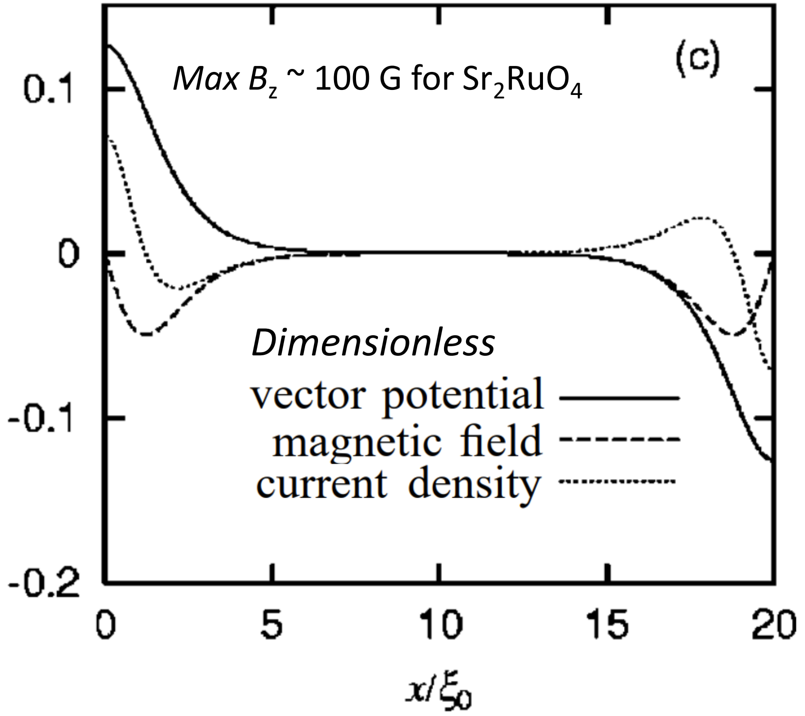


V. L. Vadimov and M.A. Silaev, PRB 98, 104504 (2018)

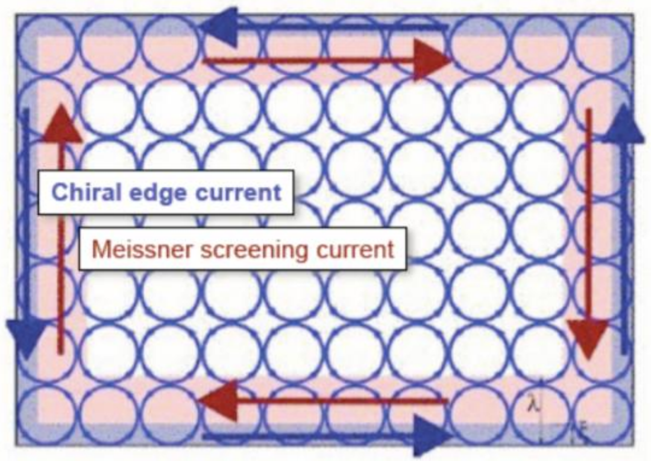
Non-chiral superconductivity has similarities with ***antiferromagnetic*** state **-> the average field is zero.**

Chiral $p_x \pm ip_y$ superconductivity

Distribution of spontaneous fields and currents in one domain ($p_x + ip_y$)



Furusaki, M. Matsumoto, and M. Sigrist: Phys. Rev. B 64 054514 (2001)
M. Matsumoto and M. Sigrist, J. Phys. Soc. Jpn. 68, 994 (1999)



C. Kallin http://www.icmr.ucsb.edu/programs/documents/Kallin.pdf

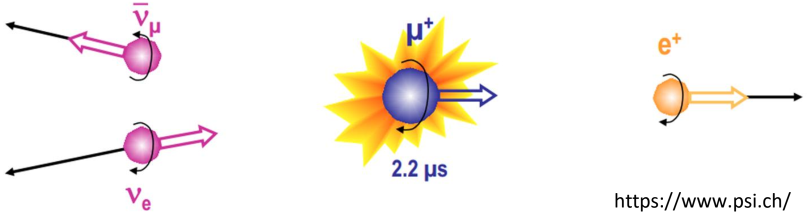
J. R. Kirtley et al., Phys. Rev. B 76, 014526 (2007).

Spontaneous currents flow along the surfaces, domain boundaries, and crystalline defects.

Chiral superconductivity has similarities with ***ferromagnetic*** state → **spontaneous bulk magnetization.**

Experimental methods (bulk) sensitive to BTRS superconductivity

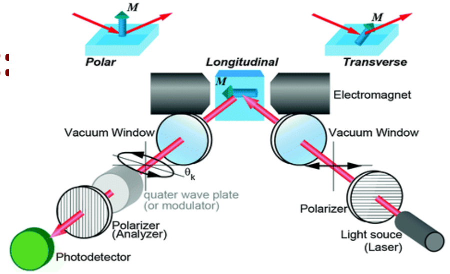
Muon spin rotation/relaxation (μSR)



https://www.psi.ch/

μSR is a local probe is sensitive to **both chiral and non-chiral states** with **broken time reversal symmetry (BTRS)**

Polar Kerr effect:

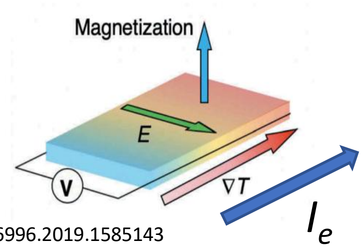


A change of the reflected light polarization → "reliably" sensitive to chiral superconductivity, only.

https://doi.org/10.1007/978-981-10-6156-1_108

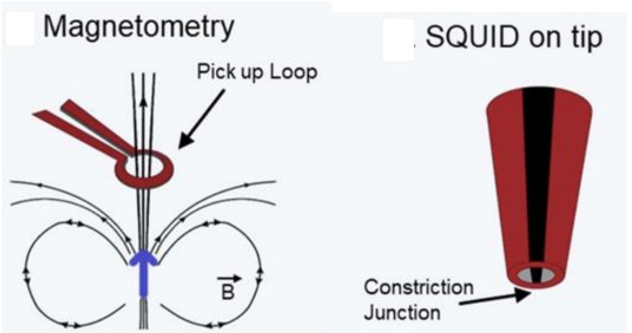
The most direct are
Scanning SQUID or NV center magnetometry

Spontaneous Nernst and Hall effects in zero magnetic field
→ *due to averaging can be zero in a BTRS state*

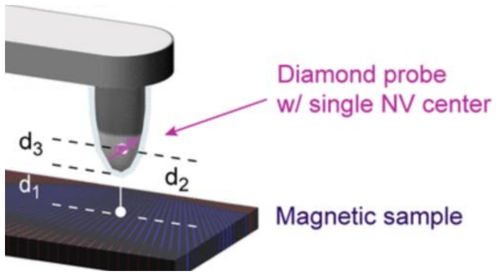


Heat or electrical charge flow

https://doi.org/10.1080/14686996.2019.1585143



J. Phys. Mater. 7 (2024) 032501

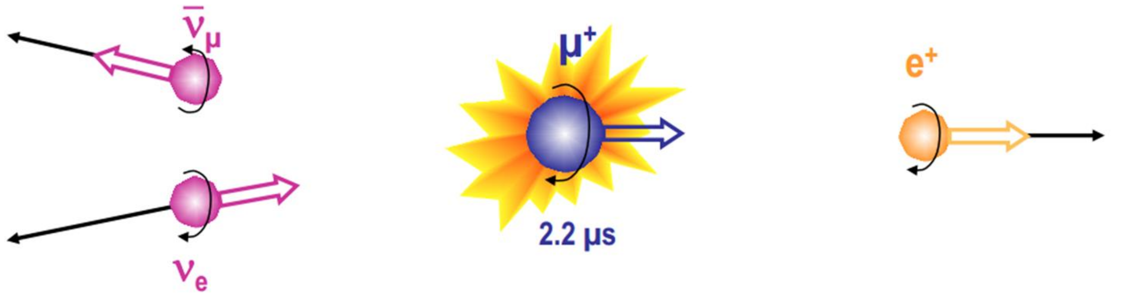


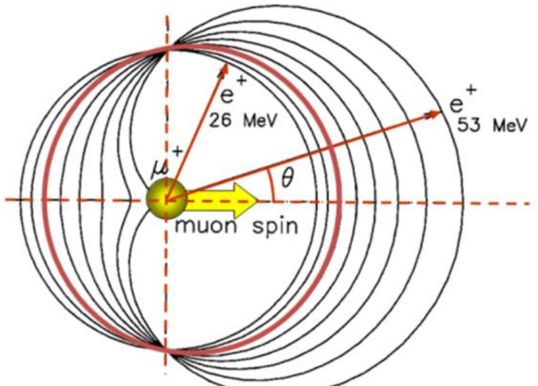
ACS Nano 2025, 19, 8, 8255-8265

High sensitivity >~ 10 nm resolution

Surface fields in BTRS state are not yet reported

Muon spin rotation relaxation (μSR)





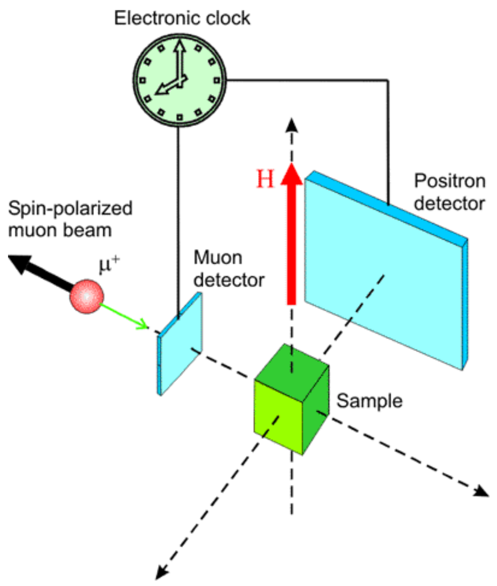
Angular distribution of positrons from the parity violating muon decay:

$$W(E,\theta) = 1 + a(E)\cos(\theta)$$

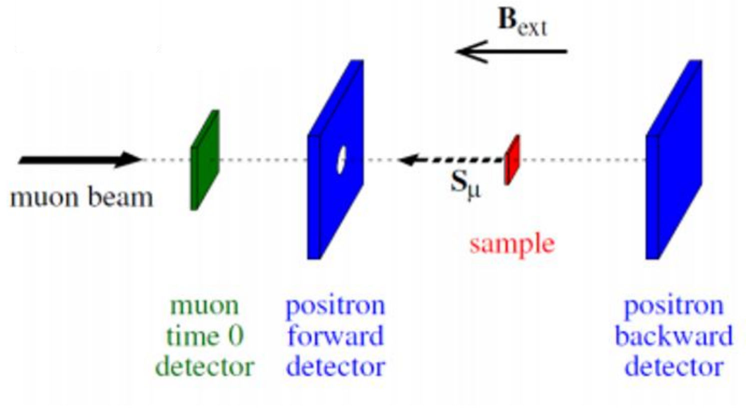
The asymmetry parameter *a* = 1/3 when all positron energies *E* are sampled with equal probability.

Positrons preferentially emitted along direction of muon spin at decay time

By detecting spatial positron emission as function of time → time evolution of muon spin



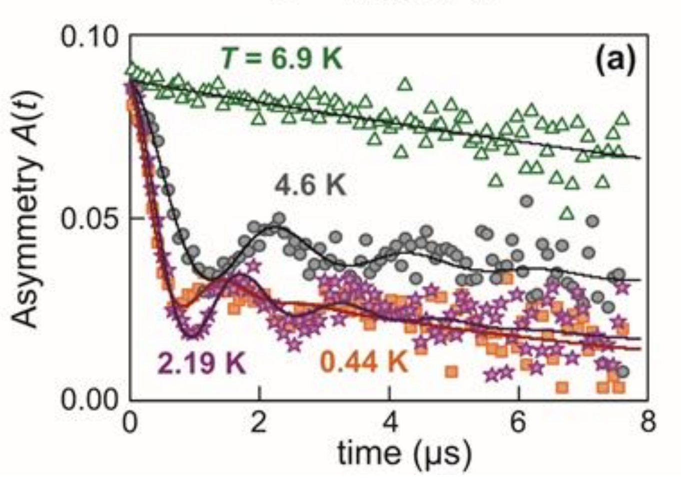
In typical experiments used 2 detectors schema



$$\frac{N_F(t) - N_B(t)}{N_F(t) + N_B(t)} = A(t)$$

Minimum sample thickness ~ 100 μm

AFM transition in Sr_2RuO_4

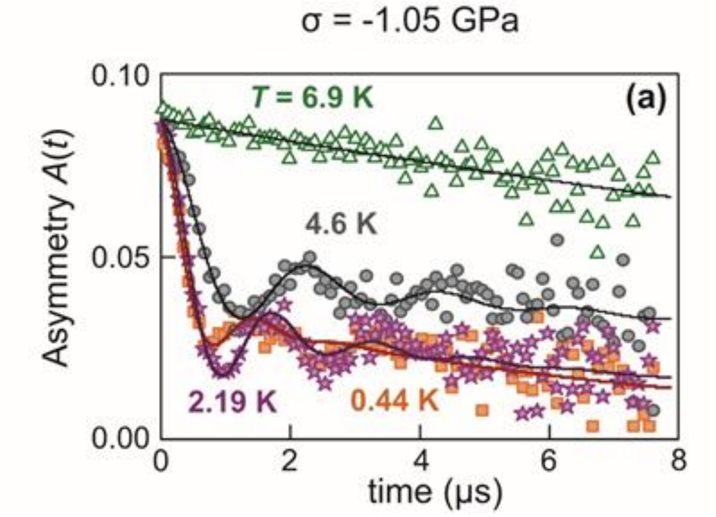


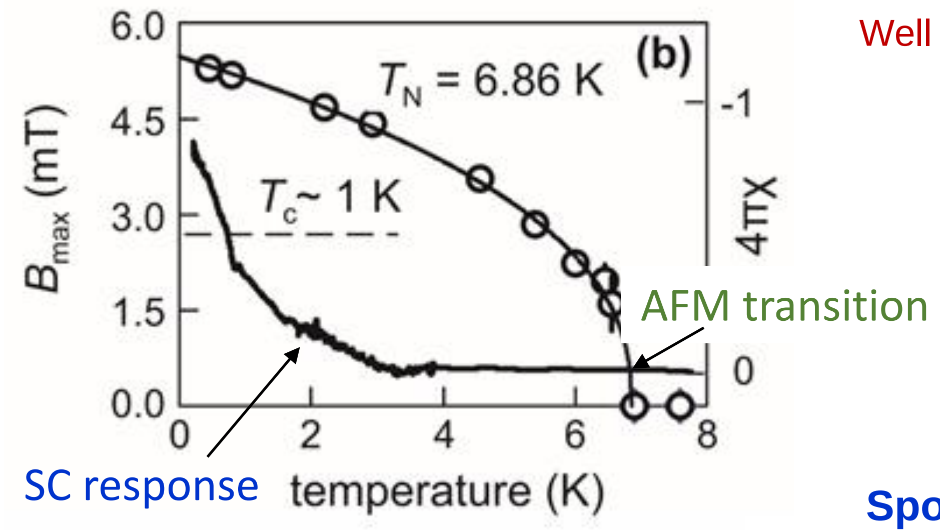
V. Grinenko, S. Ghosh *et al.* *Nat. Phys.* **17,** 748–754 (2021).

3D dynamic zero field compensation better than 0.02 Oe

Ordered vs. BTRS magnetism

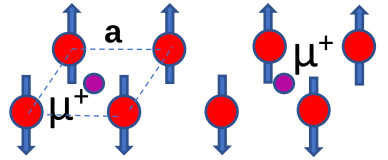
AFM transition in Sr_2RuO_4





V. Grinenko, S. Ghosh *et al.* *Nat. Phys.* **17,** 748–754 (2021).

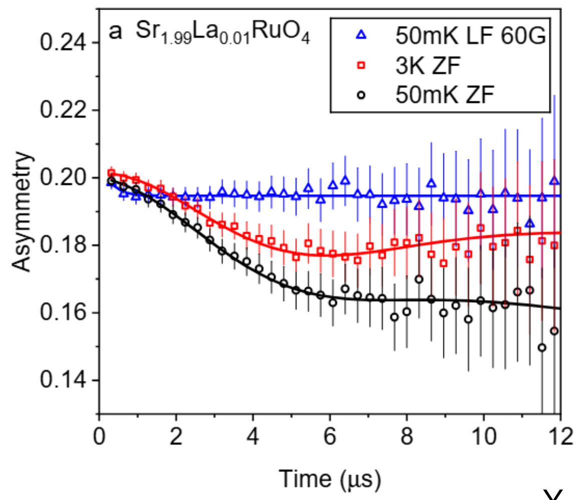
Well defined fields in every unit cell due to long-range AFM structure

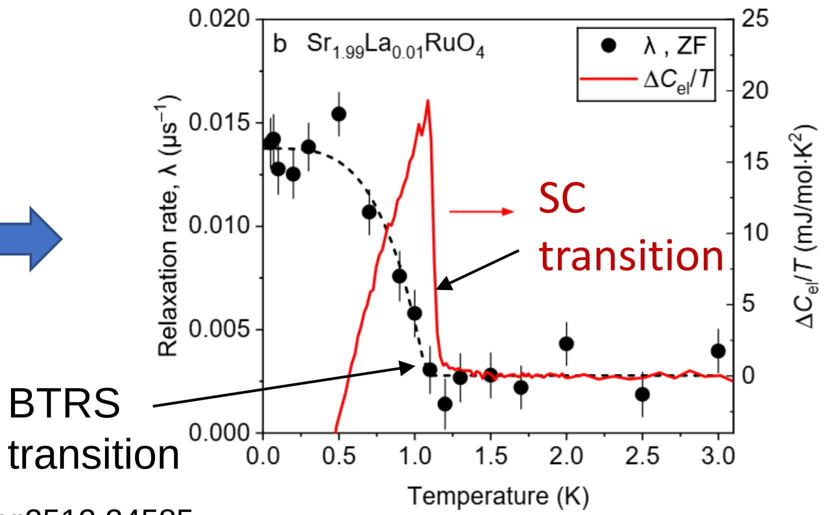


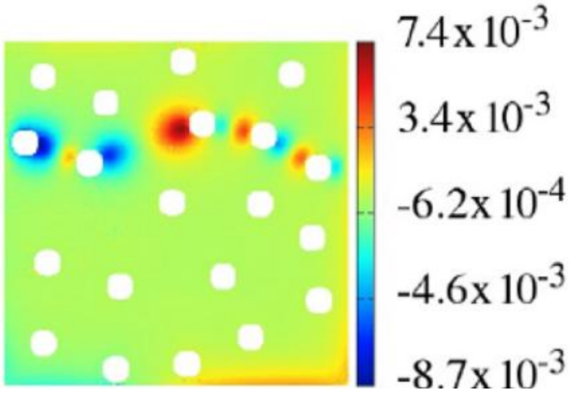
Spontaneous fields in BTRS SC

Random fields decay as $\sim e^{-\frac{r}{\lambda}}$

where unit size a << λ SC penetration depth

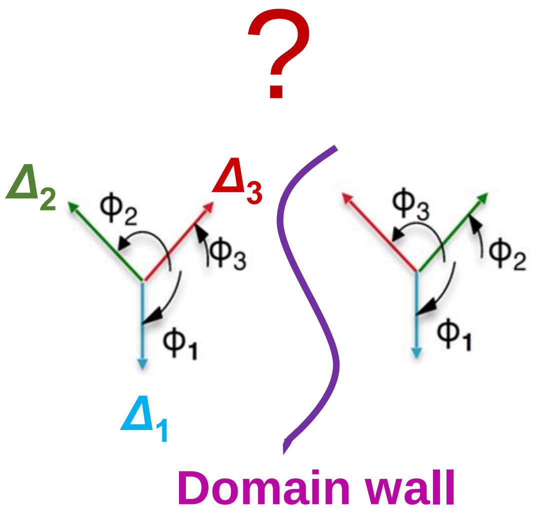






Y. Li et al. arXiv:2512.24585

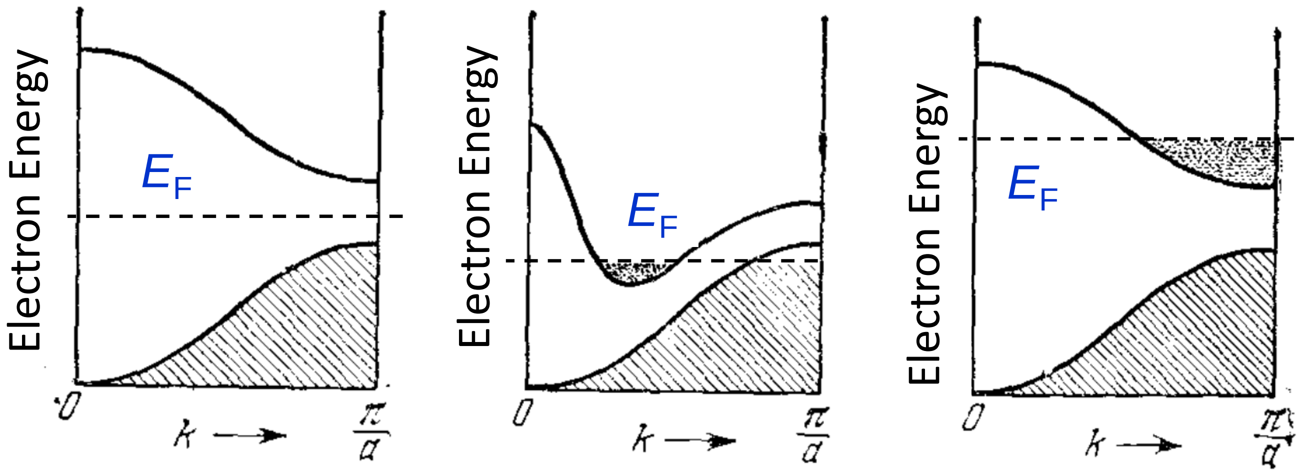
Where to look for multicomponent states?



We are looking for multicomponent superconductors in multiband metals.

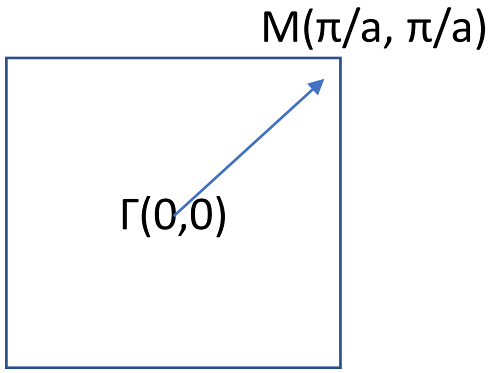
Band structure

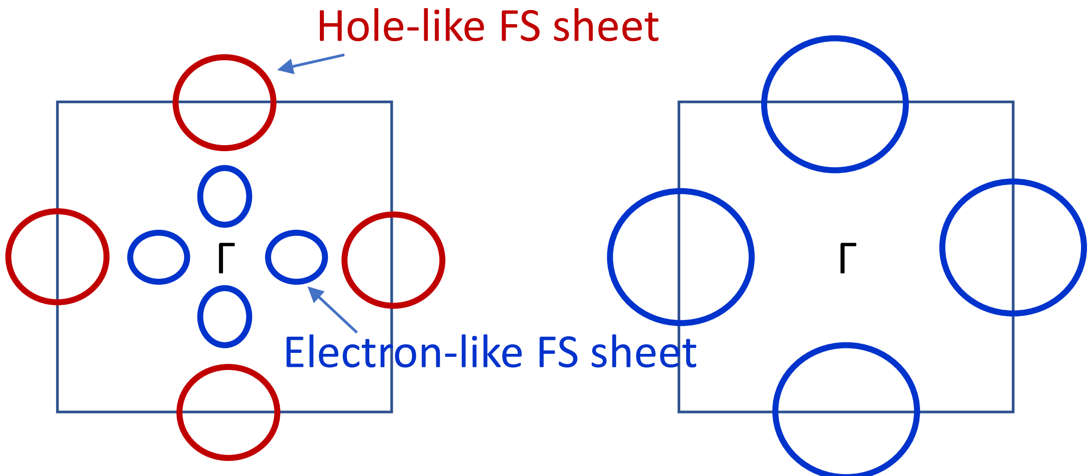
Insulator **Metal – 2 band** **Metal - single band**



The ***electronic band structure*** of the crystals results from electrons moving in the periodic potential of atoms.

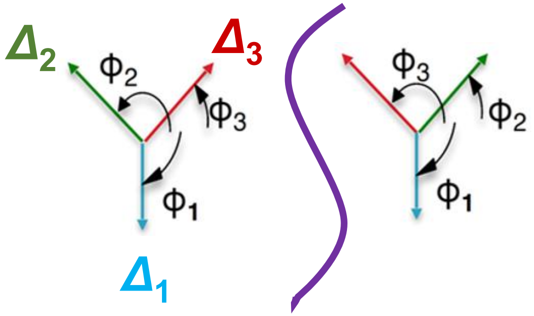
Fermi surface (FS)





Where to find multiband superconductors?

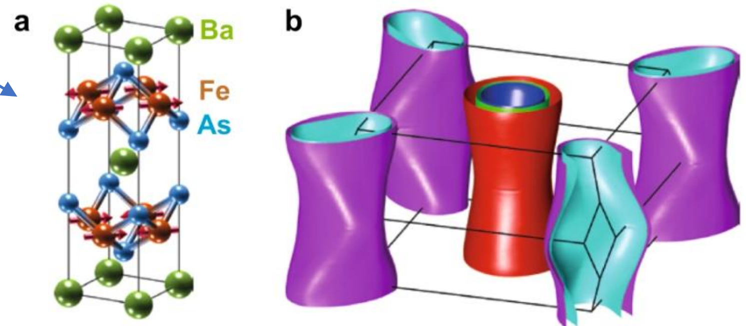
?



Domain wall

Fe base superconductors

Crystal structure



Fermi surface (electronic sates at the Fermi energy)

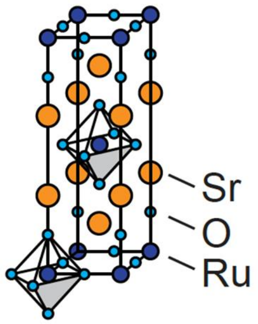
T_c^{max} ~ 60 K

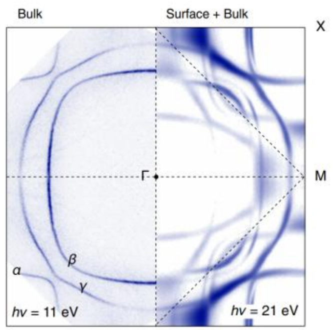
Watson, M.D., et al. npj Quantum Mater. 4, 36 (2019).

Possibilities: *s*+*is'* and *s*+*id*

T_c^{max} ~ 3.5 K

Sr_2RuO_4





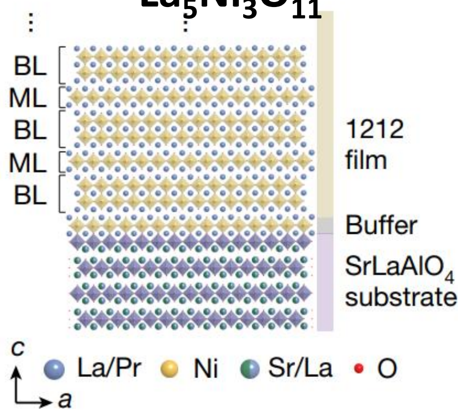
A. Tamai et al., Phys. Rev. X **9**, 021048 (2019).

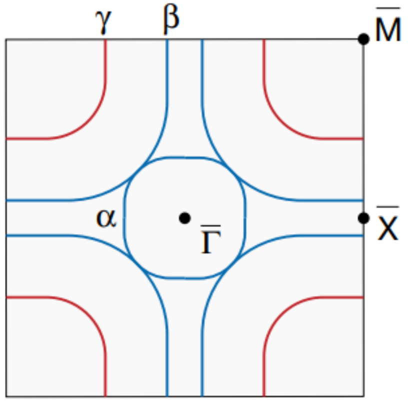
Possibilities: *s*+*id*, *d*+*ig*, *d*+*id*

Nickelates

T_c^{max} ~ 50 K

$La_5Ni_3O_{11}$





Z. Nie et al., Nature 652, 628–634 (2026).

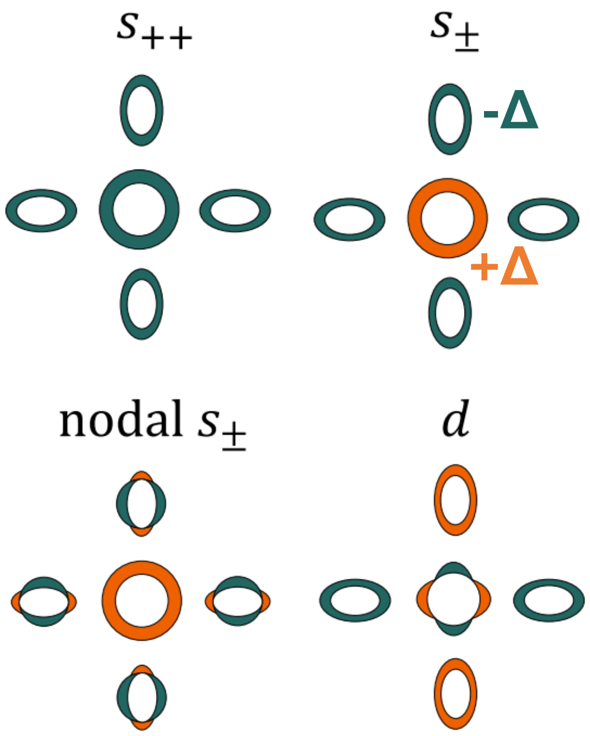
Possibilities: *s*+*id*

Multiband unconventional – *s*-wave superconductivity in iron based superconductors

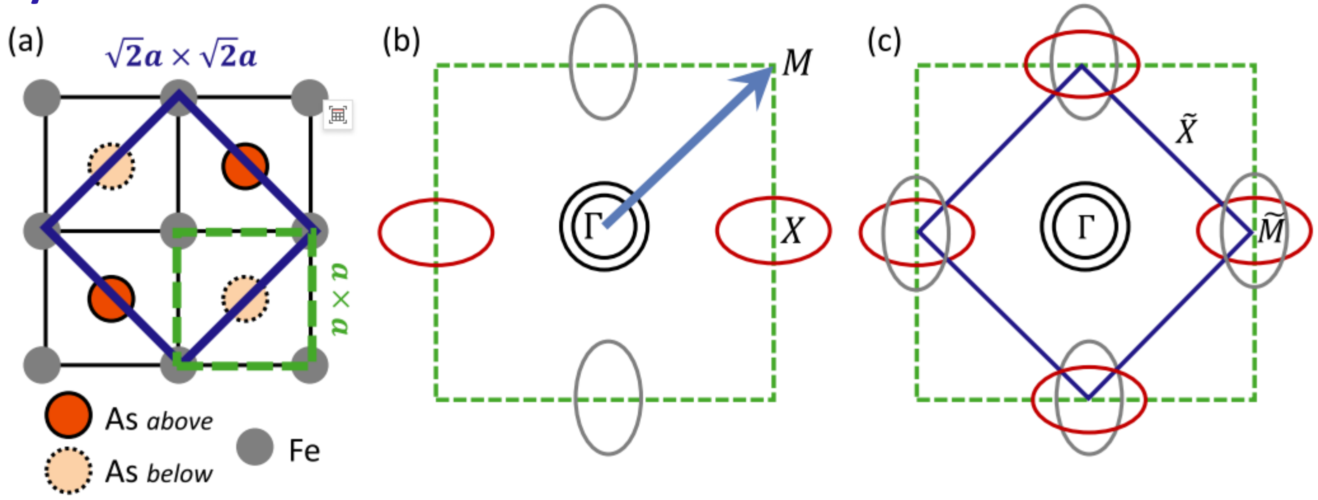
Tsung-Dao Lee Institute

Proposal for *s*-wave superconductivity mediated by spin fluctuation

Schematic representation of the superconducting gaps at the Fermi surface.



Crystalline lattice **Fermi surface**



Theoretical treatment of the interactions in multiorbital system using Coulomb (U) and Hund's (J) couplings.

$$H = H_0 + \bar{U} \sum_{i,\ell} n_{i\ell\uparrow} n_{i\ell\downarrow} + \bar{U}' \sum_{i,\ell'<\ell} n_{i\ell} n_{i\ell'} + \bar{J} \sum_{i,\ell'<\ell} \sum_{\sigma,\sigma'} c^{\dagger}_{i\ell\sigma} c^{\dagger}_{i\ell'\sigma'} c_{i\ell\sigma'} c_{i\ell'\sigma} + \bar{J}' \sum_{i,\ell'\neq\ell} c^{\dagger}_{i\ell\uparrow} c^{\dagger}_{i\ell\downarrow} c_{i\ell'\downarrow} c_{i\ell'\uparrow},$$

P. J. Hirschfeld *et al*., Rep. Prog. Phys. 74 124508 (2011)

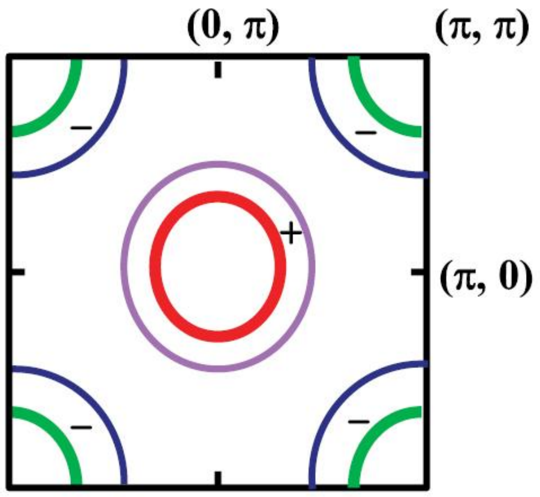
Multiband unconventional – $s_{\pm}$-wave superconductivity in iron based superconductors

Free energy for a multicomponent order parameter $\psi_\alpha = |\Delta_\alpha| e^{i\Phi_\alpha}$ **where α= 1,2,3 and $\Phi_\alpha - \Phi_\beta = \theta_{\alpha\beta}$** (an s+is' sate).

$$f = \frac{1}{2\rho^2}\left[\left(\underbrace{\sum_{i=1,2,3} |\Delta_i|^2 \vec{\nabla}\phi_i\right) - e\rho^2 \vec{A}}_{\text{SC}}\right]^2 + \underbrace{\sum_{i>j} \frac{|\Delta_i|^2|\Delta_j|^2}{2\rho^2}\left[\vec{\nabla}(\phi_i - \phi_j)\right]^2 + \sum_{i>j} \eta_{ij}|\Delta_i||\Delta_j|\cos(\phi_i - \phi_j)}_{\text{TRSB}} + \sum_i V(|\Delta_i|^2, |\Delta_i|^4) + \frac{\mathbf{B}^2}{2}$$

Interband Josephson coupling results in $s_{\pm}$ superconductivity at optimal doping

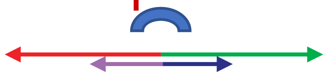
Case	Sign of $\eta_{12}, \eta_{13}, \eta_{23}$		Ground State Phases
1	s_{++}	$- - -$	$\varphi_1 = \varphi_2 = \varphi_3$
2	s+is	$- - +$	Frustrated
3	s_{+}	$- + +$	$\varphi_1 = \varphi_2 = \varphi_3 + \pi$
4	s+is	$+ + +$	Frustrated



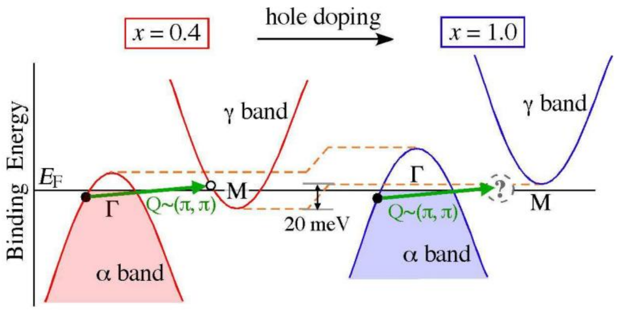
Yunkyu Bang and G R Stewart 2017 *J. Phys.: Condens. Matter* **29** 123003

$Ba_{1-x}K_xFe_2As_2$

$\Phi_1 = \pi$



Only two leading bands for optimal doping!



T. Sato *et al*., Phys. Rev. Lett. **103** (2009) 047002

It is broadly accepted that interband interactions between electron and hole Fermi pockets are responsible for the magnetism and superconductivity.

Why do we choose the $Ba_{1-x}K_xFe_2As_2$ system?

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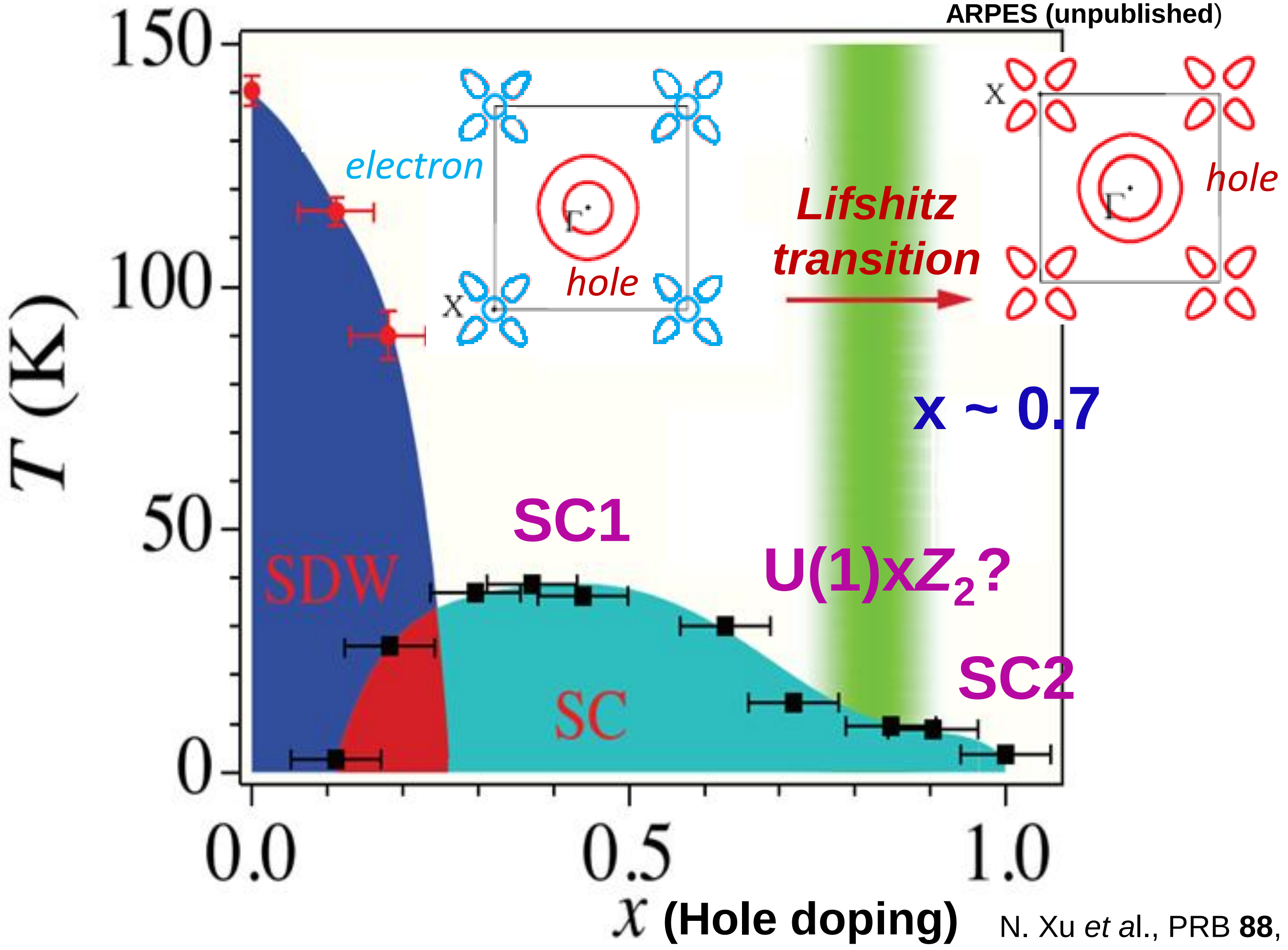
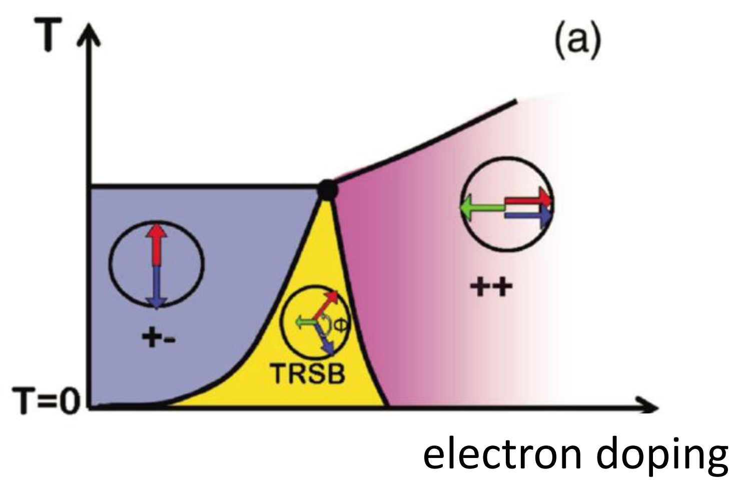
D. Evtushinsky at al., ARPES (unpublished)



N. Xu *et al.*, PRB **88**, 220508(R) (2013)

- The system is multiband with more then 3 disconnected Fermi pockets.
- Hole doping results in topological changes of the Fermi surface **(Lifshitz transition)** at $x_L = 0.7$.

Predictions for the phase diagram of $Ba_{1-x}K_xFe_2As_2$



S. Maiti and A.V. Chubukov PRB **87**, 14451 (2013).

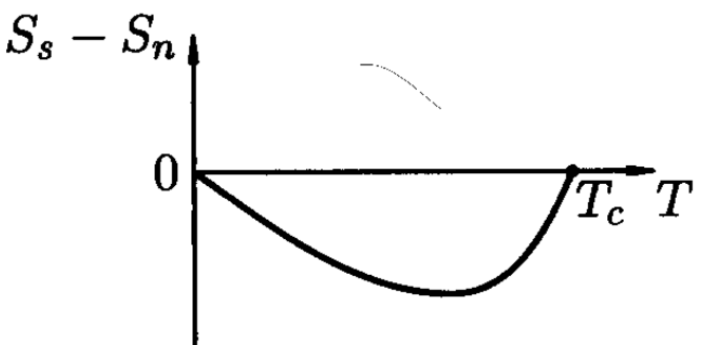
- **The superconducting dome is continuous across the Lifshitz transition.**

Is the BTRS sate possible close to Lifshitz transition?

Specific heat jump in superconductors direct probe of the order parameter structure

$$F_n - F_{s0} = H_{cm}^2/8\pi$$

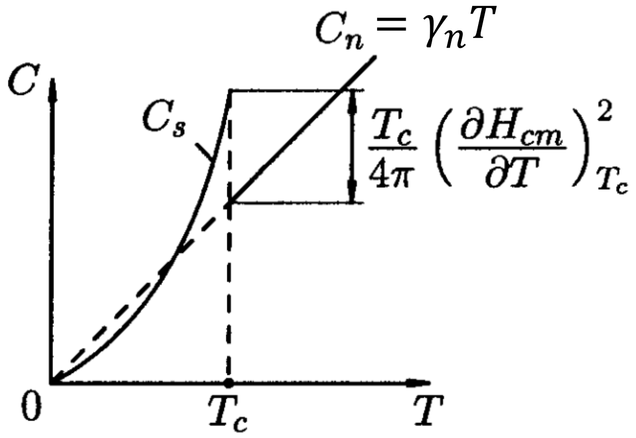
The free energy of the superconductor is related to thermodynamic critical field H_{cm} of a superconductor.



Using: $S = -(\partial F/\partial T)_R$, we get $S_s - S_n = \frac{H_{cm}}{4\pi}\left(\frac{\partial H_{cm}}{\partial T}\right)_R$.

The specific heat is given $C = T(\partial S/\partial T)$, resulting in

$$\Delta C_{el} = C_s - C_n = \frac{T_c}{4\pi}\left(\frac{\partial H_{cm}}{\partial T}\right)^2_{T_c}$$



Within the **BCS theory the free energy of the superconductor** is related to the energy gap Δ_0 (the order parameter amplitude at $T \to 0$).

$$\frac{H_{cm}^2(0)}{8\pi} = \frac{1}{2}N(0)\Delta_0^2,$$

In a conventional *s* –wave superconductor $\Delta C_{el}/\gamma_n T_c = 1.43$, where $\frac{2\Delta_0}{k_B T_c} = 3.52$

For, an *d* –wave superconductor $\Delta C_{el}/\gamma_n T_c = 0.95$ with $\frac{2\Delta_0}{k_B T_c} = 4.1$ (for weak coupling)

The jump reflects the order parameter structure!

Specific heat of $Ba_{1-x}K_xFe_2As_2$

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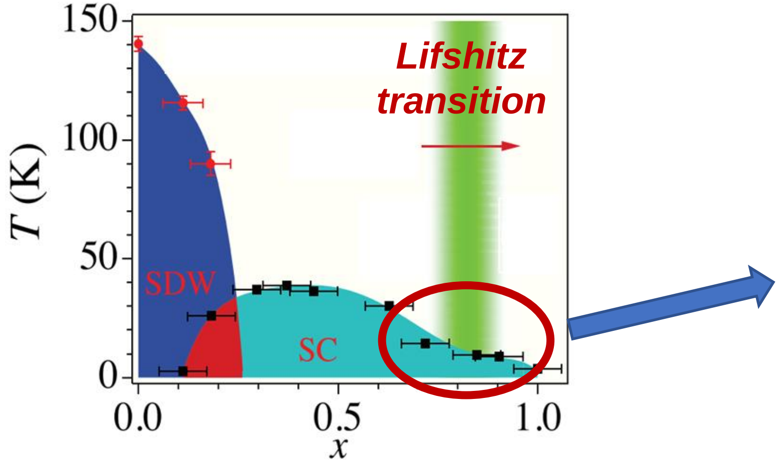
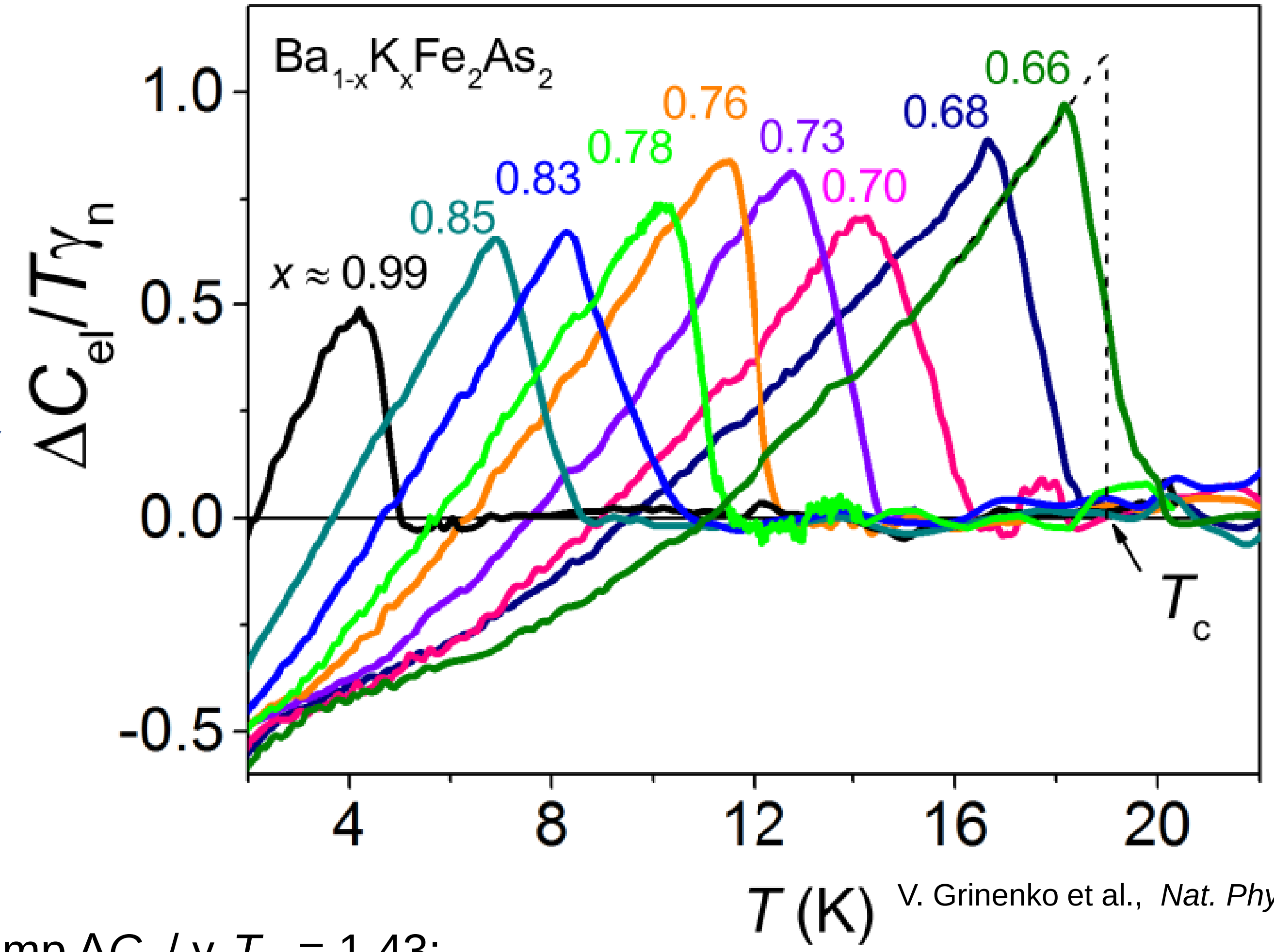
TSUNG-DAO LEE INSTITUTE





V. Grinenko et al., *Nat. Phys.* **16**, 789–794 (2020).

- In the BCS theory the specific heat jump $\Delta C_{el}/ \gamma_n T_c = 1.43$;
- $\Delta C_{el}/ \gamma_n T_c$ is below BCS value at all doping levels -> multiband behaviour;
- Non-monotonic dependence on T_c.

Thermodynamic signature of two superconducting states in $Ba_{1-x}K_xFe_2As_2$

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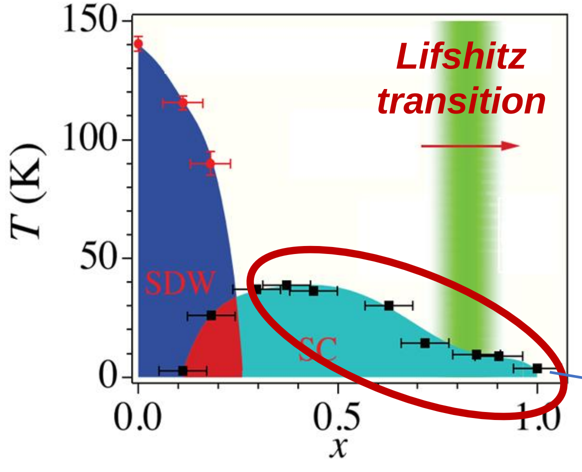
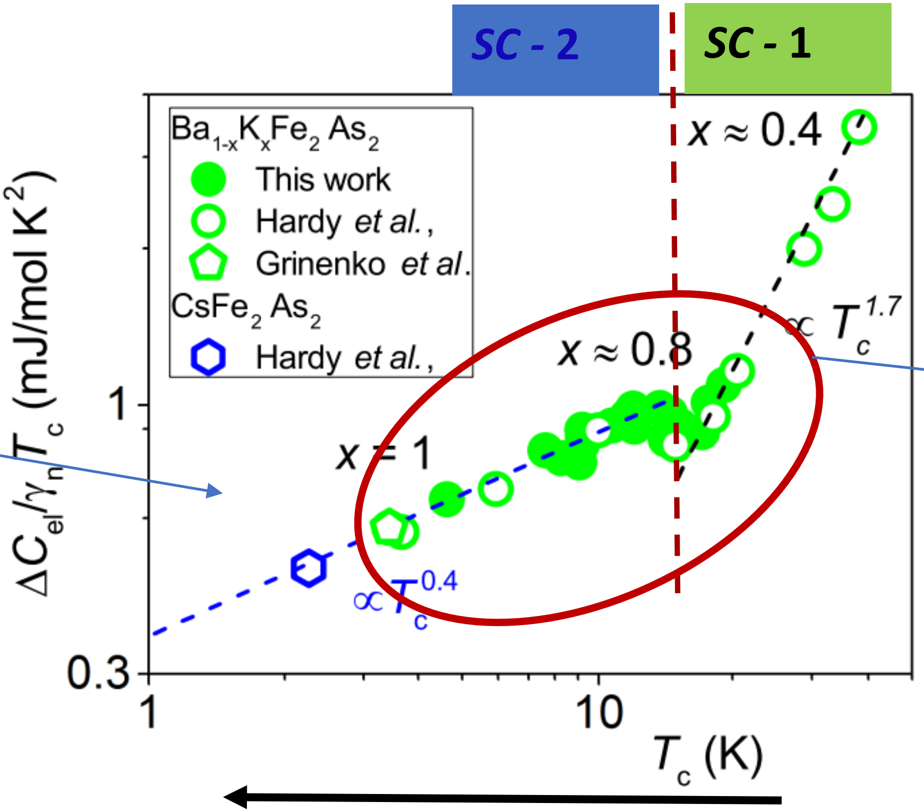
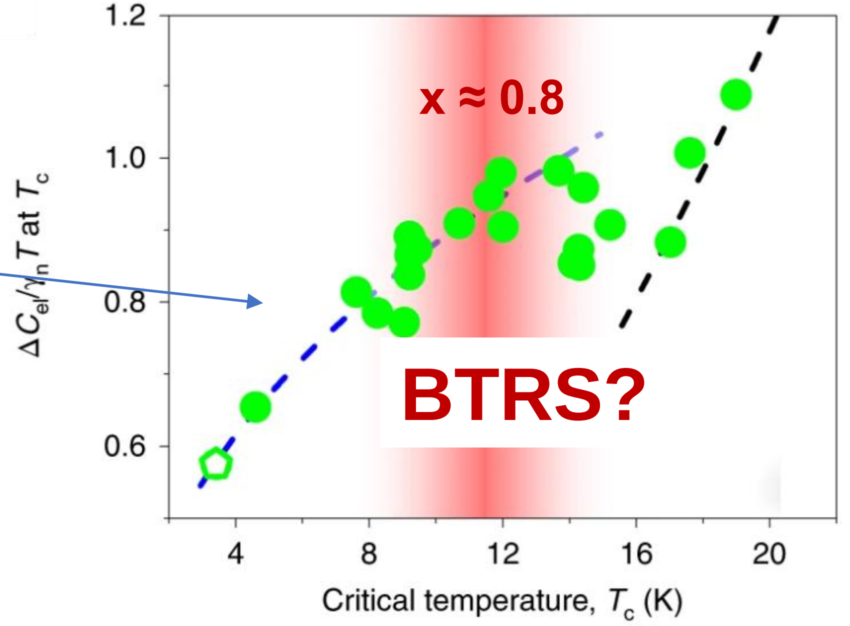




K doping

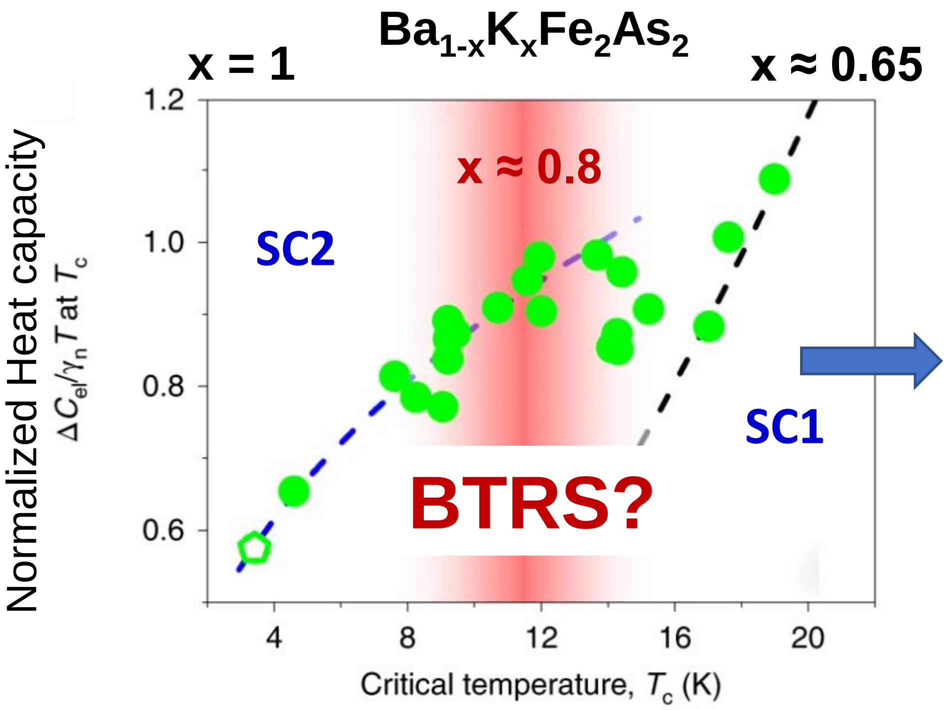


V. Grinenko et al., *Nat. Phys.* **16**, 789–794 (2020).

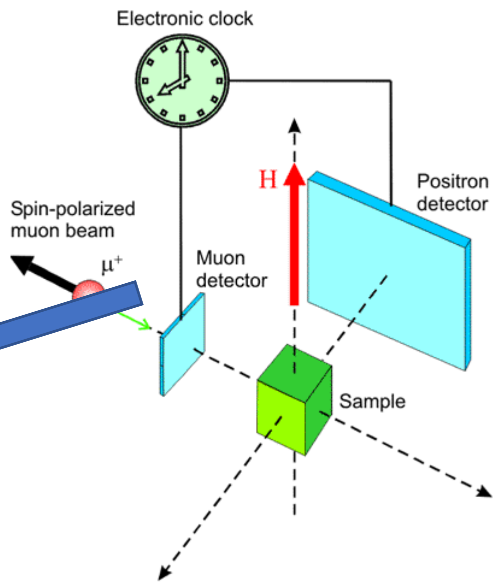
- Non-monotonous doping dependence of $\Delta C_{el}/\gamma_n T_c$ indicates changes of the electronic or the superconducting gap structure;
- Two different scaling behaviors -> two qualitatively different superconducting states.

Possible BTRS state around x ≈ 0.8!

Search for BTRS superconductivity in $Ba_{1-x}K_xFe_2As_2$ with μSR



SμS – Swiss Muon Source at High Intensity Proton Accelerator Facility



Proton Accelerator

V. Grinenko et al., Phys. Rev. B 95, 214511 (2017).
V. Grinenko et al., *Nat. Phys.* 16, 789 - 794 (2020).

Search for BTRS superconductivity in $Ba_{1-x}K_xFe_2As_2$ with μSR

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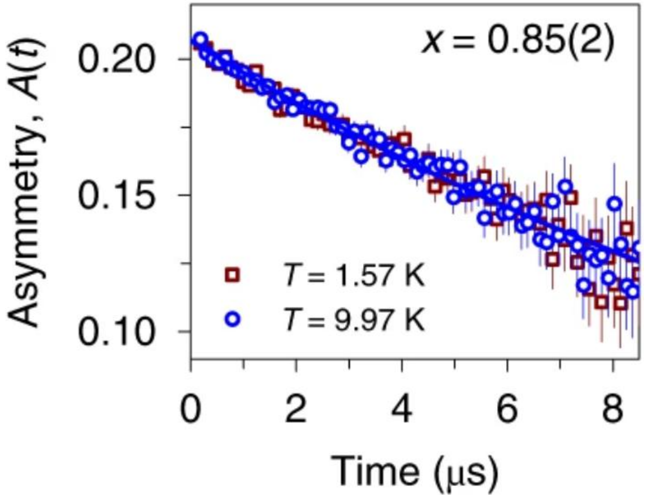
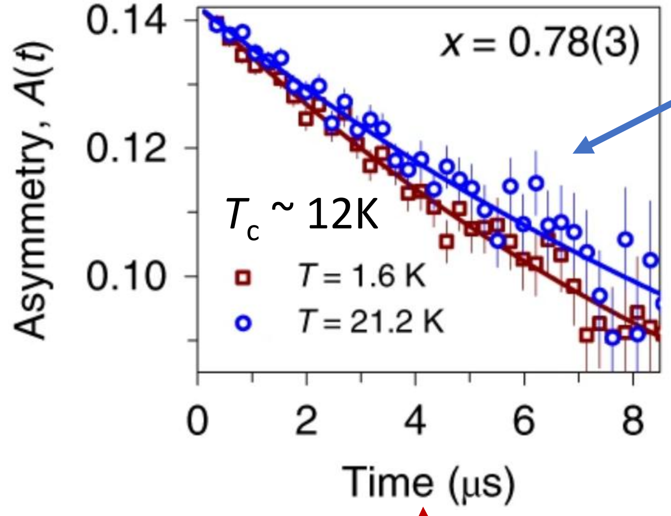
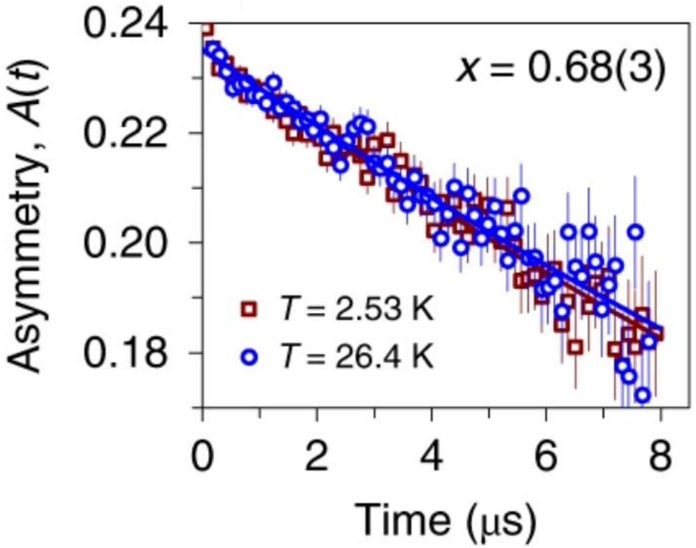
Muon-spin relaxation rate doesn't change across T_c ~ 9K

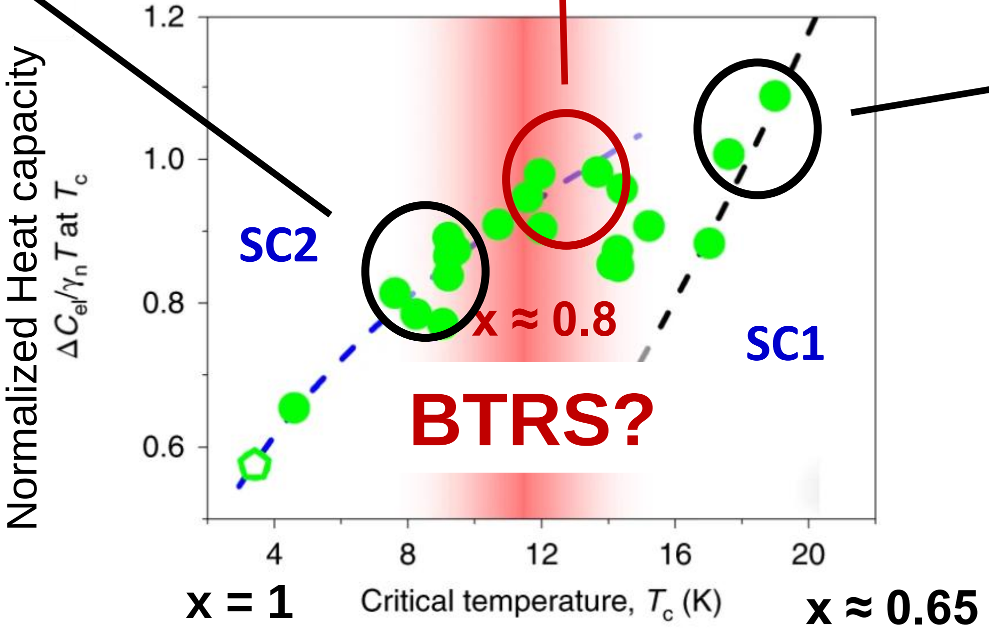




Muon-spin relaxation rate is enhanced in the superconducting state!

Relaxation doesn't change across T_c ~ 19K

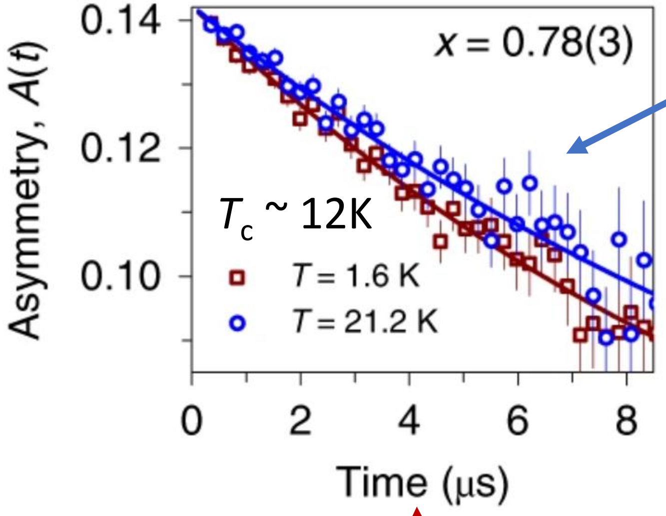




V. Grinenko et al., Phys. Rev. B 95, 214511 (2017).

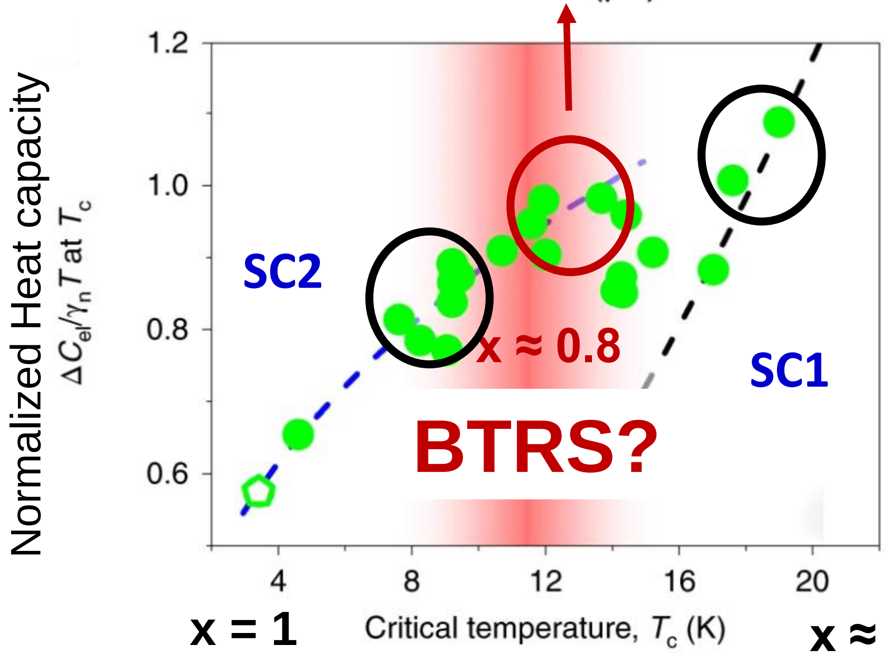
V. Grinenko et al., *Nat. Phys.* 16, 789 - 794 (2020).

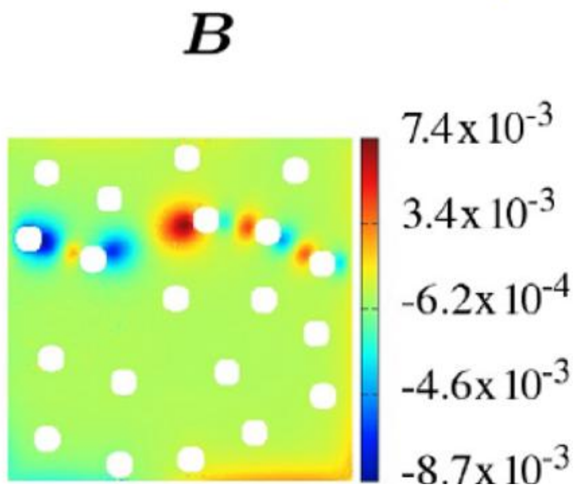
Search for BTRS superconductivity in $Ba_{1-x}K_xFe_2As_2$ with μSR



Muon-spin relaxation rate is enhanced in the superconducting state!

No oscillations of the Asymmetry -> consistent with dilute field sources.



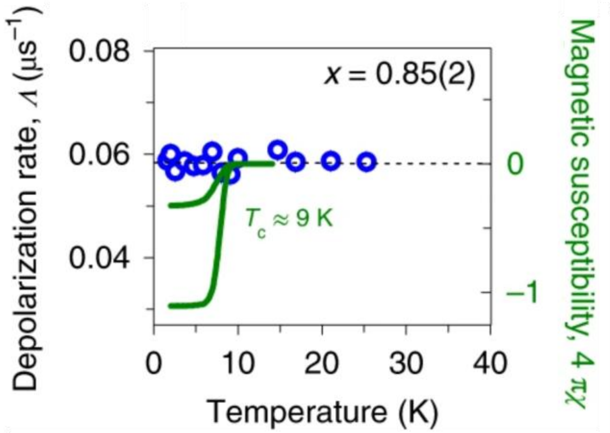


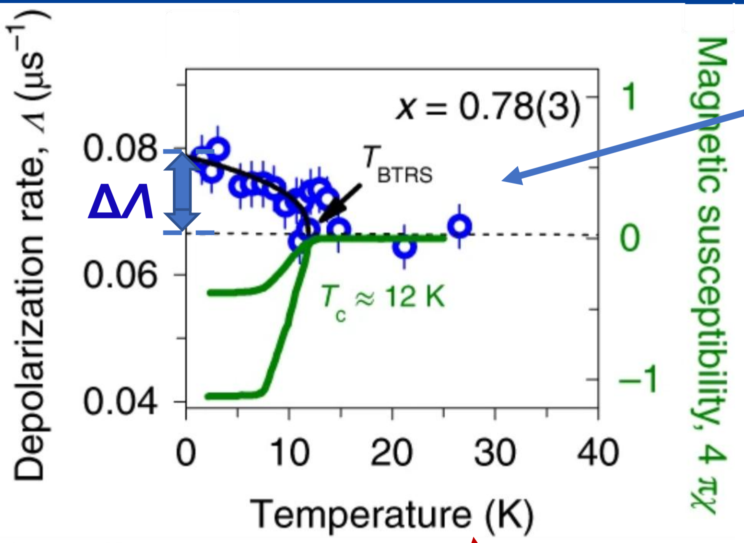
J. Garaud and E. Babaev, PRL 112, 017003 (2014)

V. Grinenko et al., Phys. Rev. B 95, 214511 (2017).
V. Grinenko et al., *Nat. Phys.* 16, 789 - 794 (2020).

Search for BTRS superconductivity in $Ba_{1-x}K_xFe_2As_2$ with μSR

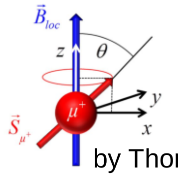
Muon-spin relaxation rate doesn't change across T_c



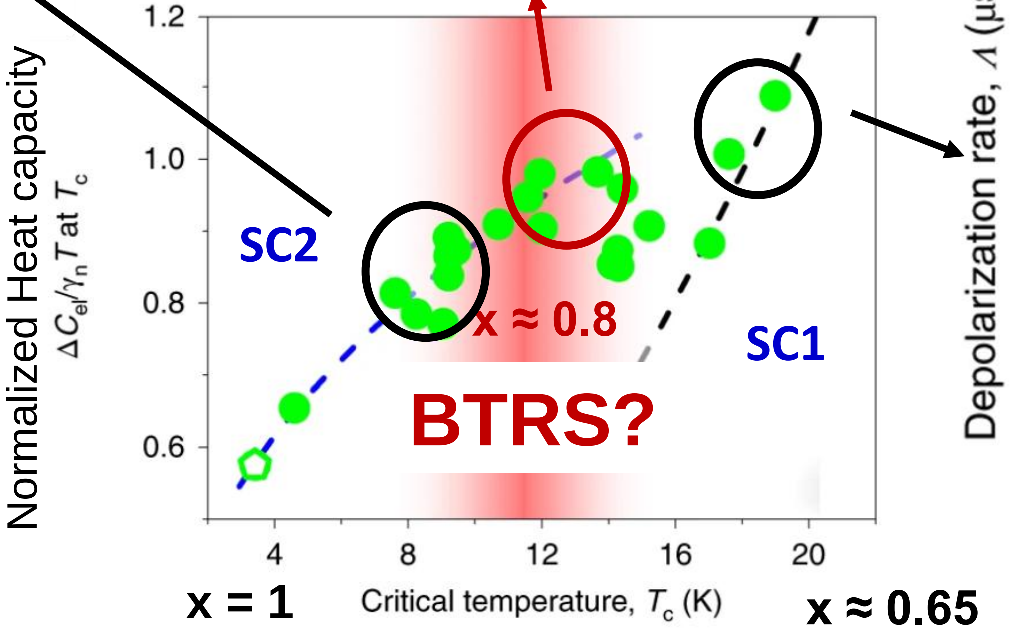


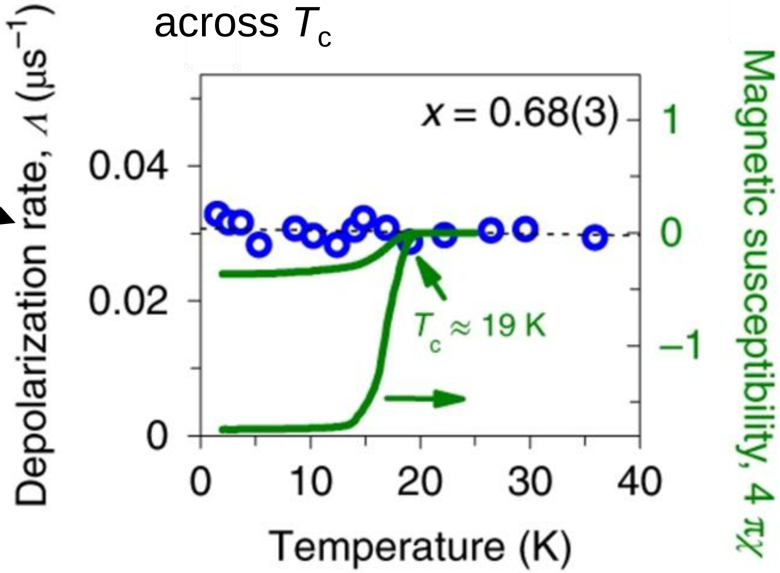
Muon-spin relaxation rate is enhanced in the superconducting state!

Average spontaneous field $\langle B_{int} \rangle = \Delta\Lambda/\lambda_\mu$ ~ 0.1 G.



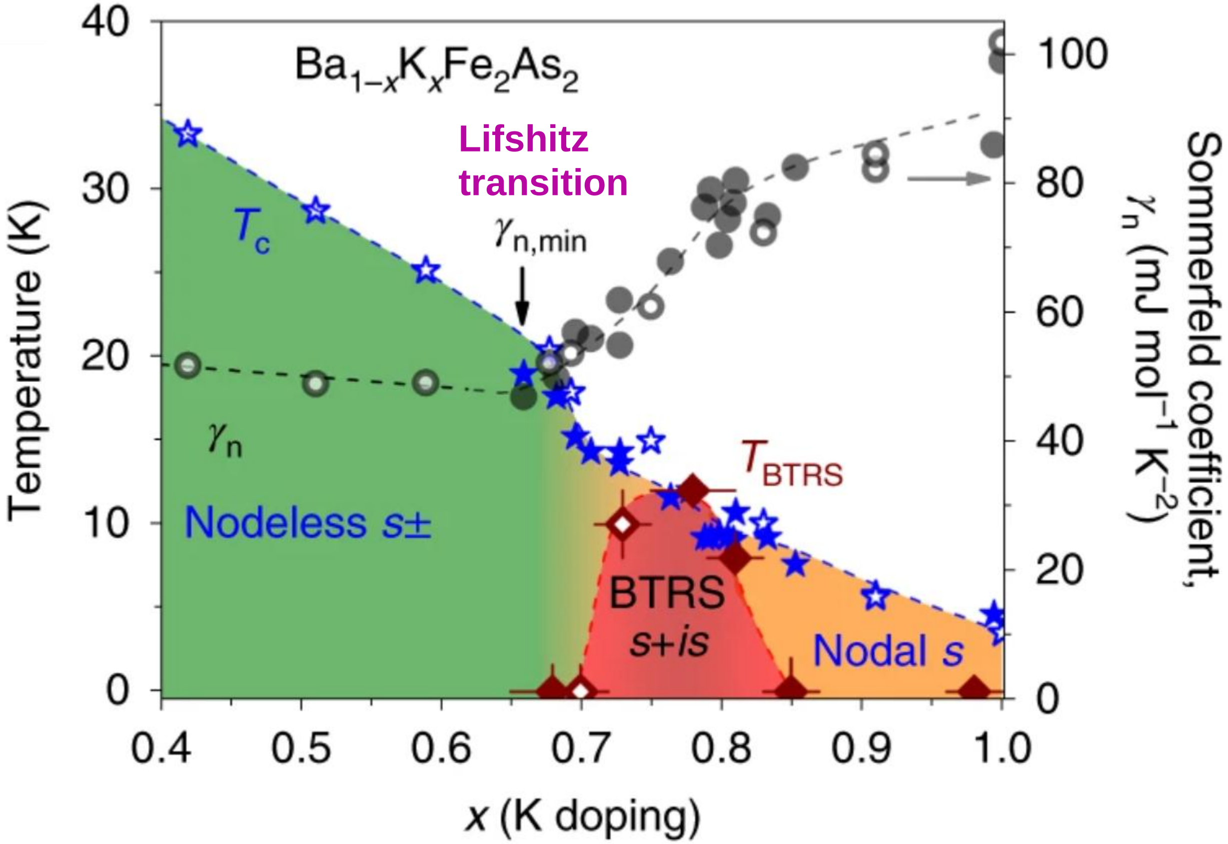
Relaxation doesn't change across T_c



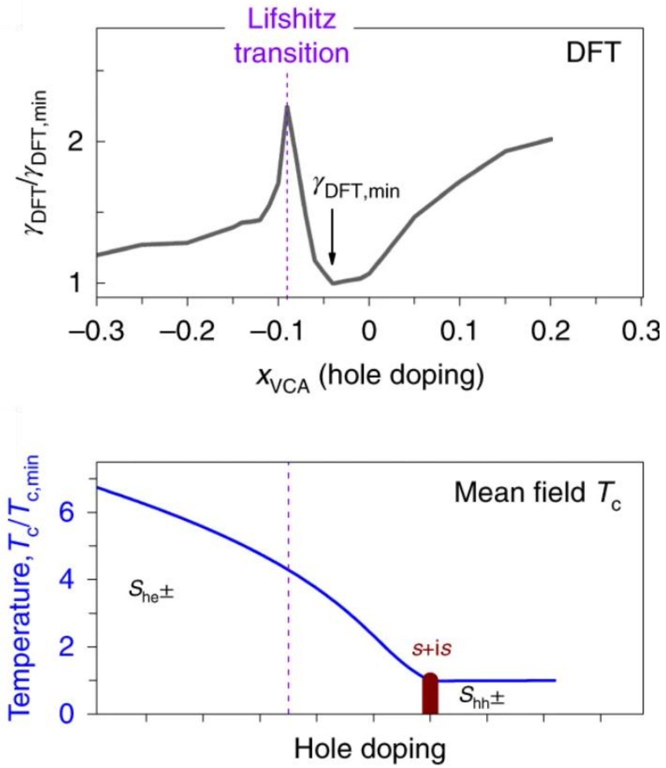


V. Grinenko et al., Phys. Rev. B 95, 214511 (2017).
V. Grinenko et al., *Nat. Phys.* 16, 789 - 794 (2020).

BTRS superconductivity in $Ba_{1-x}K_xFe_2As_2$



Density Functional Theory calculations of the effect of Lifshitz traction



The Moun-spin depolarization rate is enhanced around T_c in a limited doping range only.

A narrow *BTRS* dome ($0.7 \lesssim x \lesssim 0.85$) close to the Lifshitz transition between two different superconducting states.

V. Grinenko et al., Phys. Rev. B 95, 214511 (2017).
V. Grinenko et al., *Nat. Phys.* 16, 789 - 794 (2020).

BTRS superconductivity driven by Lifshitz transition

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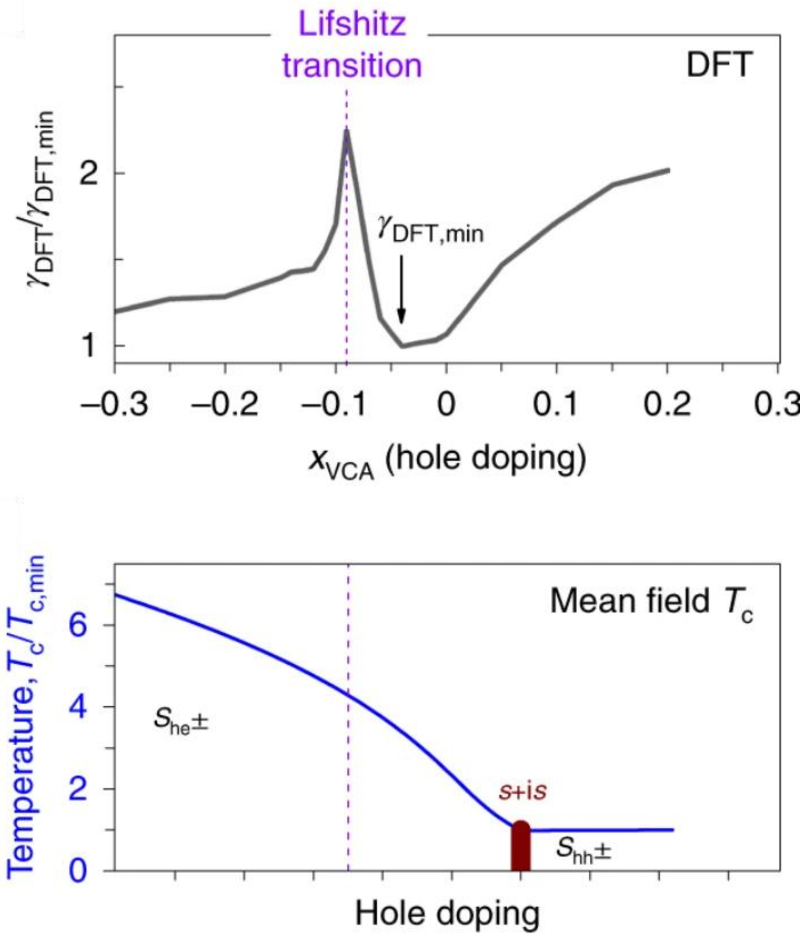
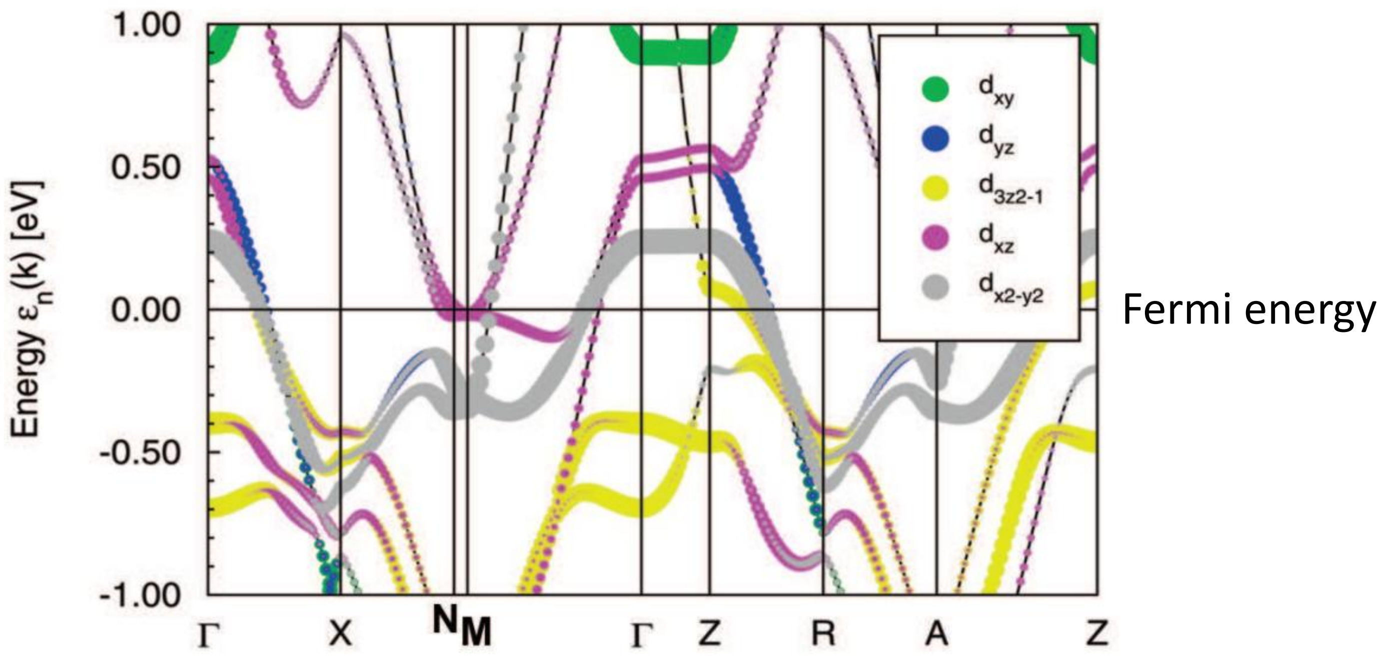


Density Functional Theory calculations of the doping effect – predication of the Lifshitz transition effect.



The result is consistent with a scenario where *s+is* superconductivity is driven by a Lifshitz transition.

J. Böker et al., Phys. Rev. B 96, 014517 (2017).
V. Grinenko et al., Phys. Rev. B 95, 214511 (2017).
V. Grinenko et al., *Nat. Phys.* 16, 789 - 794 (2020).

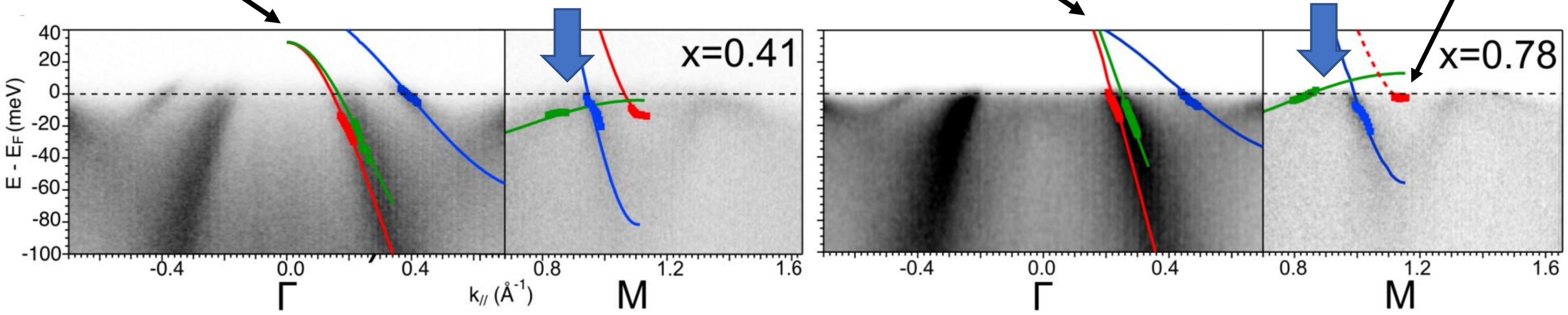
ARPES measurements of $Ba_{1-x}K_xFe_2As_2$

Three hole pockets

Three hole pockets

Lifshitz transition

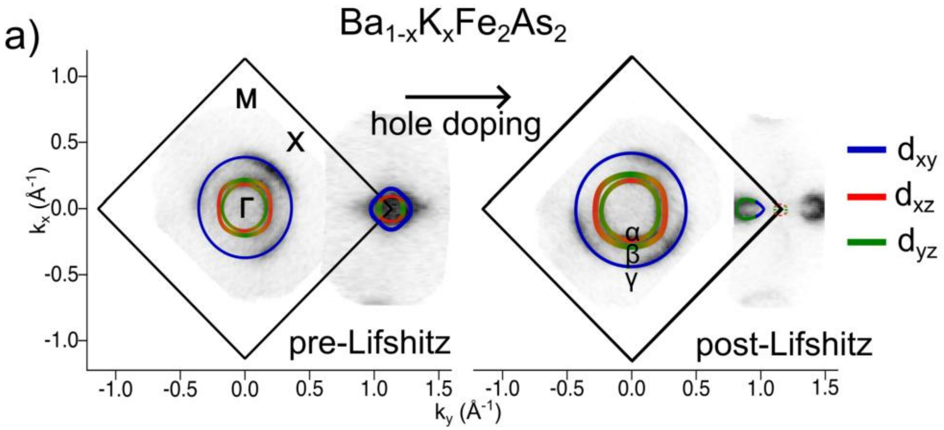
The electron pocket is still present in BTRS state.

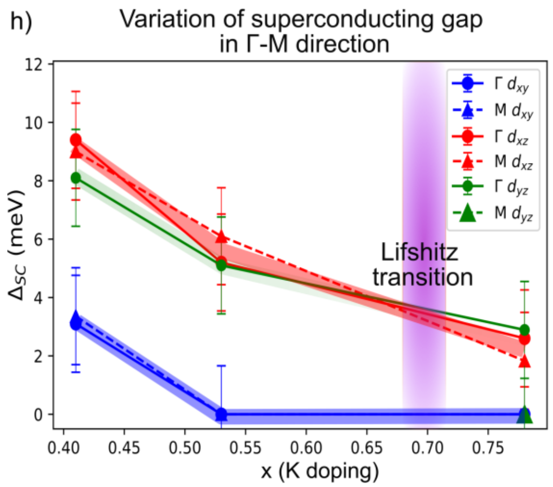


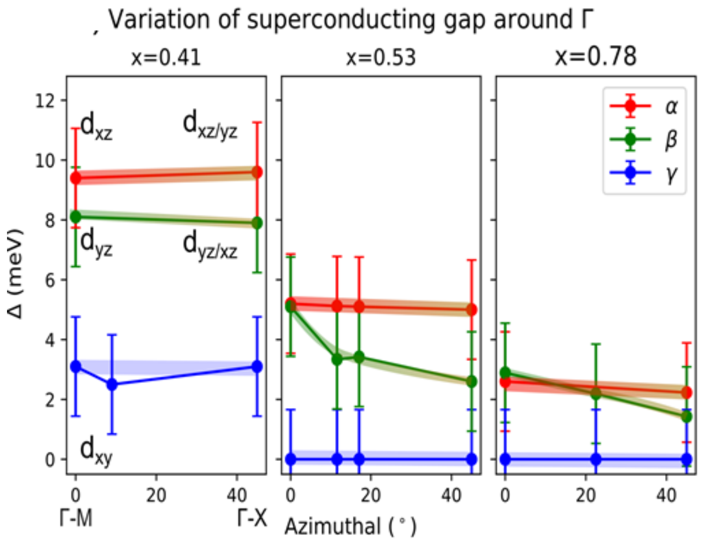
Stanford University

Zhi-Xun Shen

Elena Corbae







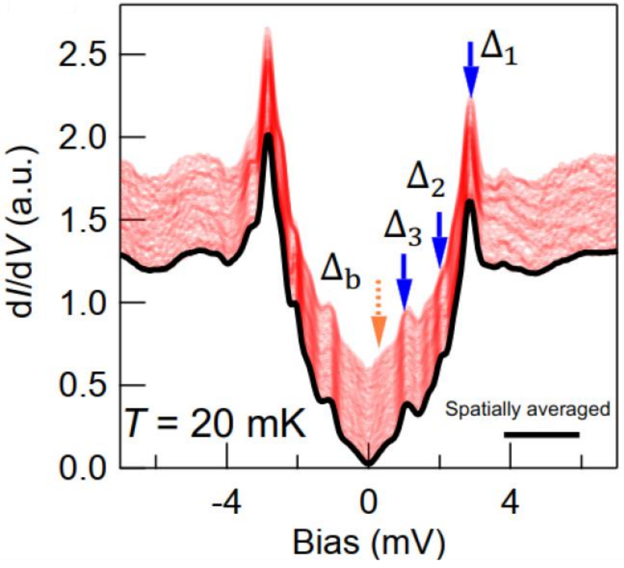
3 nearly equal components for BTRS state

Elena Corbae *et al.*, arXiv:2510.06435

Evidence for 3-gap superconductor in BTRS state

STM

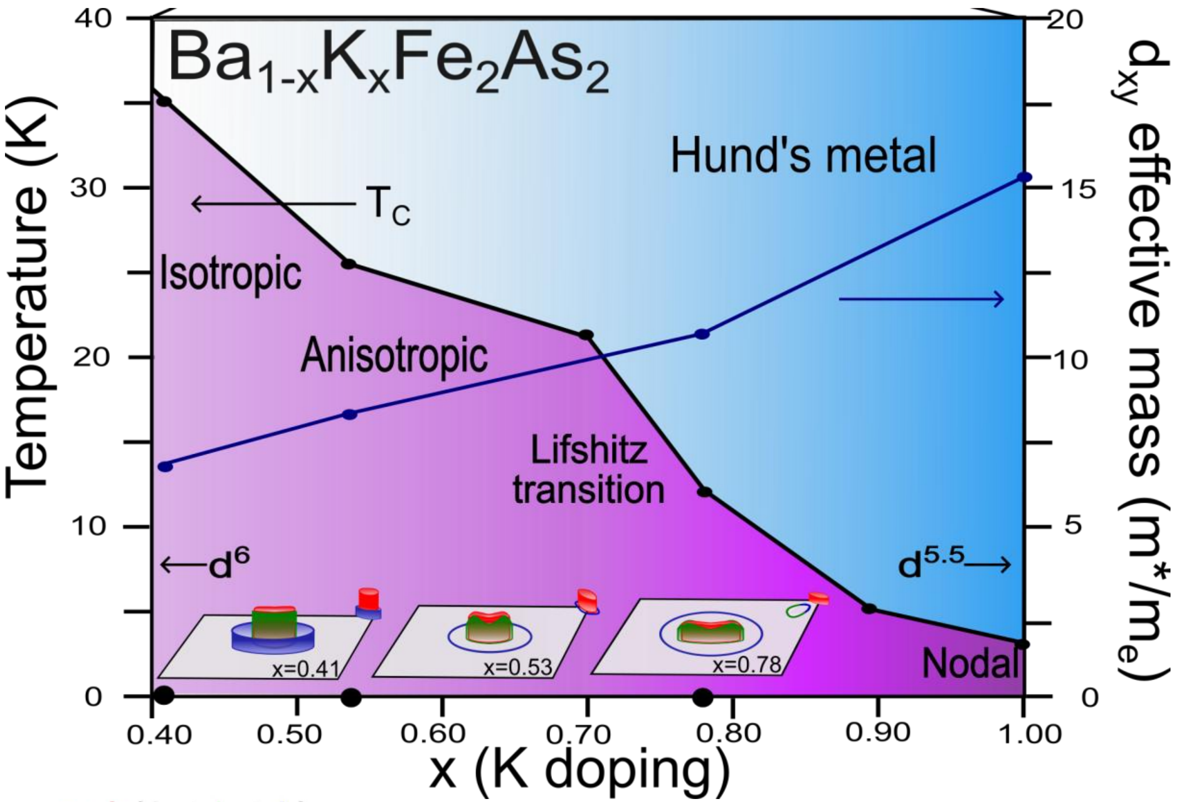
Quanxin Hu

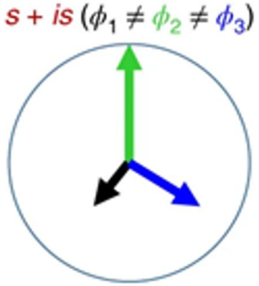


K1x1 surface of KFe_2As_2
effective doping $Ba_{0.25}K_{0.75}Fe_2As_2$

Yu Zheng, Quanxin Hu *et al.*, Science (2026).

ARPES





Stanford University

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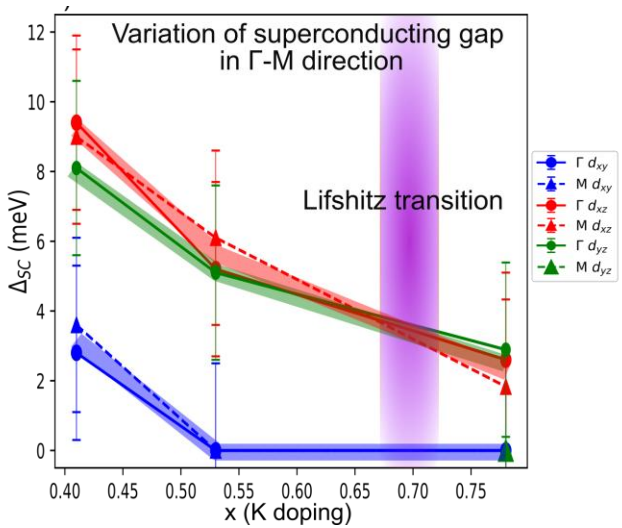
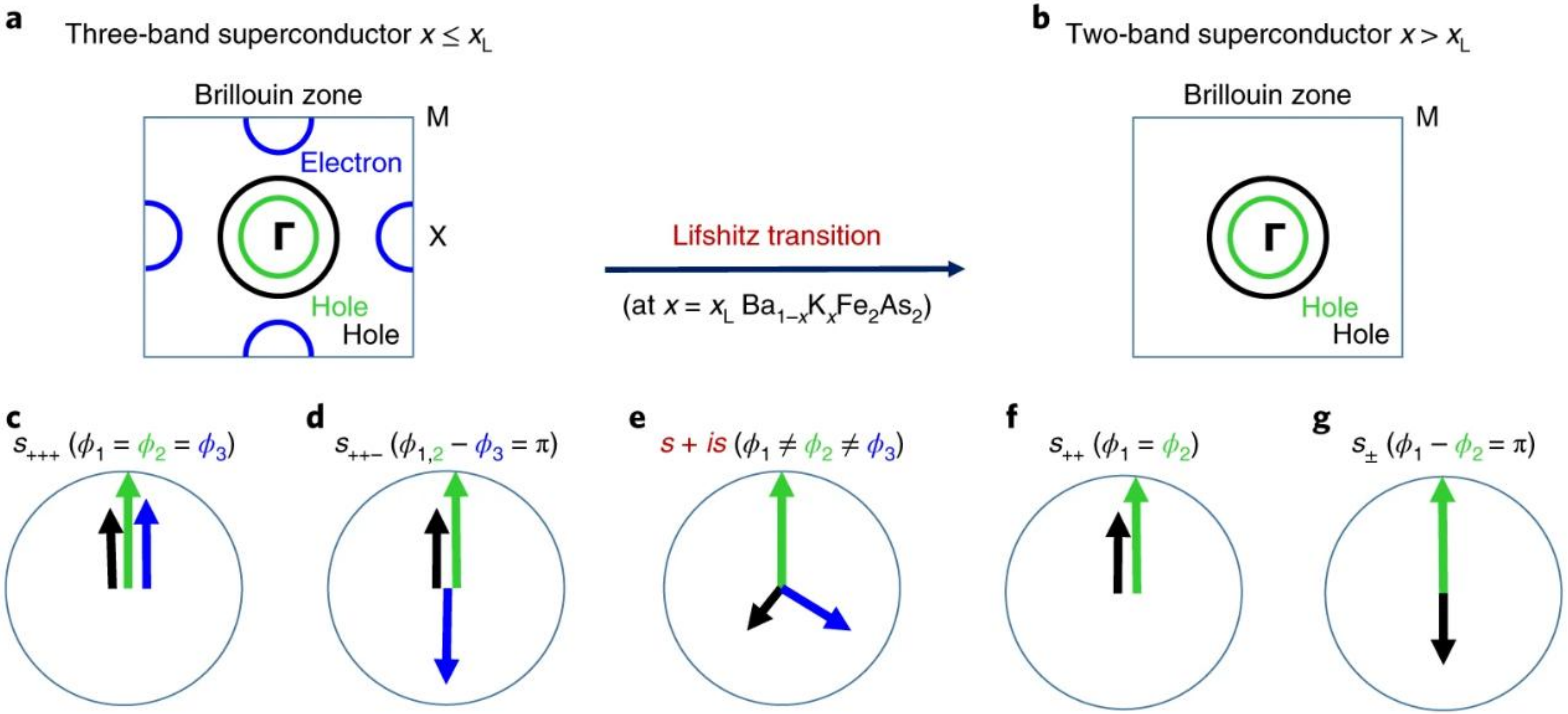


Experimental results are consistent with 3 component superconducting state!

Elena Corbae *et al.*, arXiv:2510.06435

BTRS superconductivity in $Ba_{1-x}K_xFe_2As_2$



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V. Grinenko et al., Phys. Rev. B 95, 214511 (2017).
V. Grinenko et al., *Nat. Phys.* 16, 789 - 794 (2020).

Summary of the first part

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- ✓ Superconductors with multicomponent order parameters $\psi_\alpha = |\Delta_\alpha| e^{i\Phi_\alpha}$, where **α= 1,2,3,…** can break more then one symmetry:
- ✓ We considered superconducts with broken time-reversal symmetry → **broken U(1) and Z_2** symmetries.
- ✓ **Two different phase transitions → $T_c \neq T_{BTRS}$ and two different coherence lengths $\xi_{U(1)} \neq \xi_{Z2}$.**
- ✓ Phases of the order parameter Φ_α play an important role and result in formation superconducting domains and spontaneous currents below T_{BTRS}.
- ✓ Spontaneous fields appear around inhomogeneities, at the domain walls and sample surefaces (*no spontaneous fields in infinite homogeneous superconductor*).

The most universal experimental method to detect spontaneous fields regardless the structure are the muon spin rotation/relaxation (μSR) experiments.

However, direct observations of the field sources with such probes as scanning SQUID magnetometry is still missing!

Evidence for multicomponent superconductivity

Tsung-Dao Lee Institute

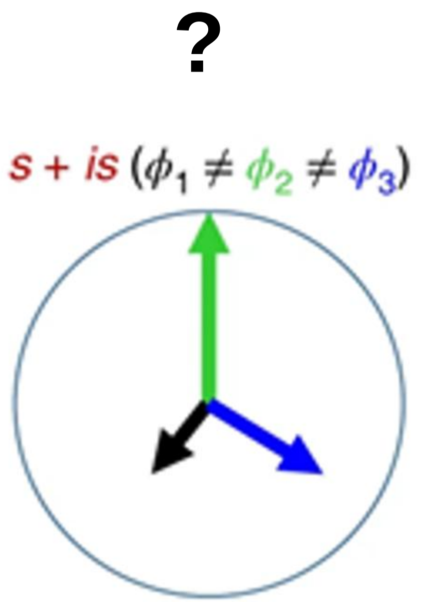
BTRS superconductivity is a multicomponent state.
Is there evidence of several components in the system?

1. Observation of the fractional vortices.

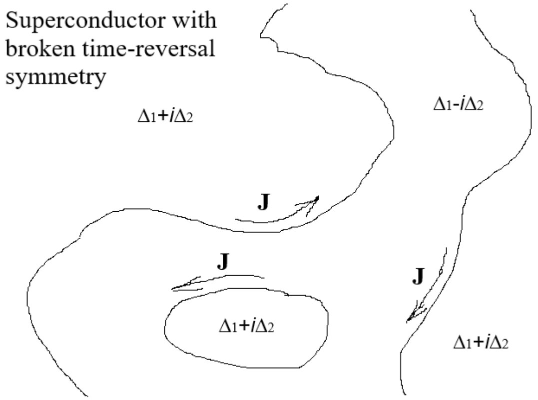
Y. Iguchi *et al. Science* 380,1244-1247(2023);
Yu Zheng, Quanxin Hu *et al*., Science (2026);
Q. Z. Zhou *et al*., arXiv:2408.05902.

2. The observation of three distinct superconducting gaps.
Yu Zheng, Quanxin Hu *et al*., Science (2026);
Elena Corbae *et al*., unpublished

3. Observations of two anomalies in the specific heat.
V. Grinenko *et al*., *Nat. Phys.* **17,** 1254–1259 (2021);
I. Shipulin *et al*., *Nat Commun* **14**, 6734 (2023).

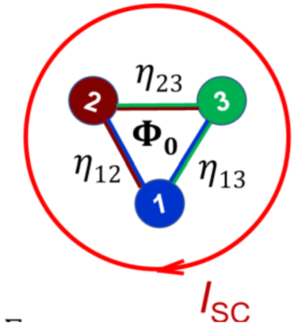


Outline of the course

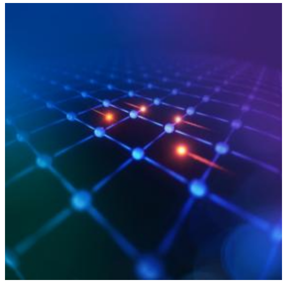


1. Superconductivity that breaks time reversal symmetry.

2. Flux quantum fractionalization and unquantized vortices in multicomponent superconductors.

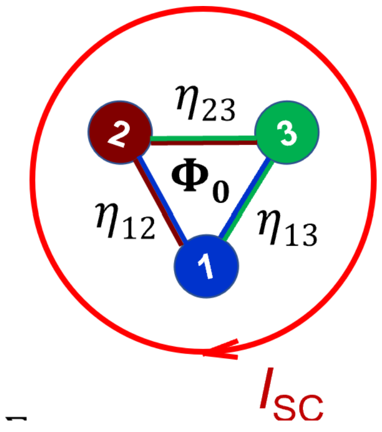


3. Electron-Quadrupling condensates in multicomponent superconductors with strong phase fluctuations.



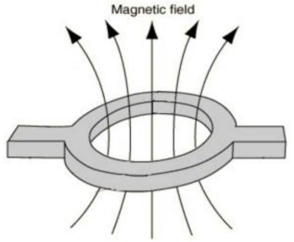
Aim of the lecture:

Provide a theoretical concept what unquantized vortices in superconductors are and show experimental evidence for their formation in multiband superconductors with broken time-reversal symmetry.



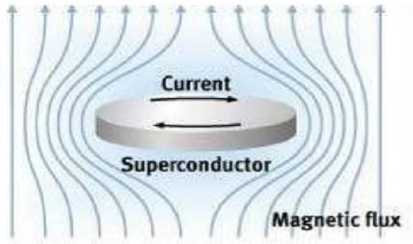
Superconductivity is a quantum phenomenon

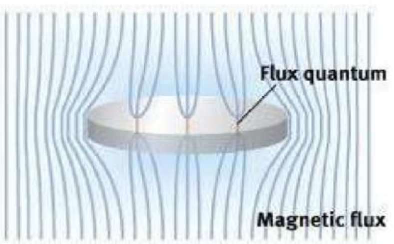
Flux quantization in superconductors



$$\Phi = n\Phi_0 = \frac{nh}{2e}$$

$H < H_{c1}$ $H > H_{c1}$





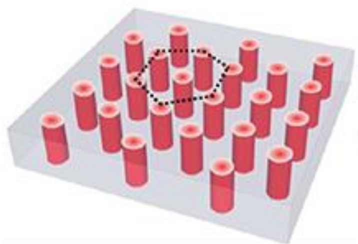
- Quantization of the frozen magnetic flux in superconductors was predicted by Fritz London in 1948

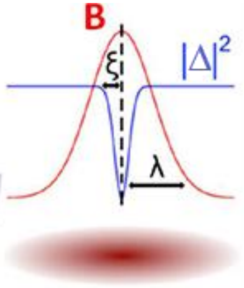
and discovered experimentally in 1961, only, by B. S. Deaver and W. M. Fairbank and, independently, by R. Doll and M. Näbauer

$$\Phi_0 = \frac{h}{2e} = 2.0679\times10^{-15}\ [\mathrm{W = T\ m^2}]$$

Onsager (1949) Feynman (1955) introduced quantum vortex states with cores, which was adopted by Abrikosov.

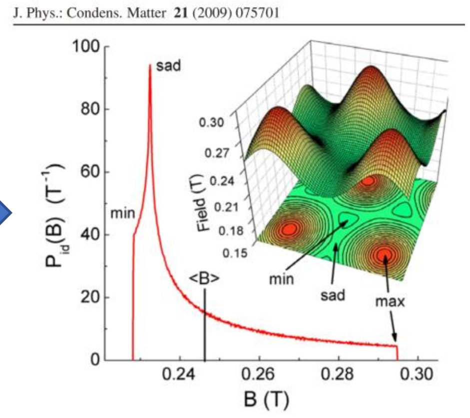
Vortex lattice in a type-II superconductor



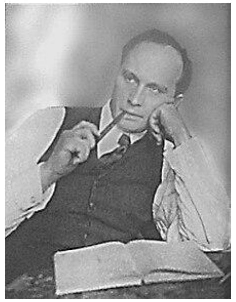


$$\vec{B} = \vec{B}_0 \exp\left(\frac{-x}{\lambda}\right)\hat{z}$$

Field distribution in a type-II superconductor



The discovery of type-II superconductivity in 1937 by Lev Vasilyevich Shubnikov



Theoretical explanation of superconducting vortex lattice 1952-1957 by Alexei Abrikosov

Vortices in superconductors

Flux-quantized vortices

Fritz London

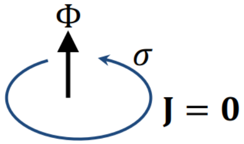
London –flux quantization

$$\Phi = \oint \mathbf{A}\cdot d\mathbf{r} = n\frac{h}{2e} = n\Phi_0$$

Wave function: $\psi = |\psi|e^{i\theta}$

Total current: $\mathbf{J} = \frac{\hbar e}{i2m}(\psi^*\nabla\psi - \psi\nabla\psi^*) - \frac{2e^2}{m}|\psi|^2\mathbf{A}$

$$= \frac{\hbar e}{m}|\psi|^2\nabla\theta - \frac{2e^2}{m}|\psi|^2\mathbf{A} = 0$$



$$\Phi = \oint_\sigma \mathbf{A}\cdot d\mathbf{r} = 2\pi n\frac{\hbar}{2e} = n\Phi_0$$

Defined by fundamental constants alone

Flux-unquantized vortices

Egor Babaev

Wave function at band j: $\psi_j = |\psi_j|e^{i\theta_j}$

Total current: $\mathbf{J} = \sum_j\left[\frac{\hbar e}{i2m}(\psi_j^*\nabla\psi_j - \psi_j\nabla\psi_j^*) - \frac{2e^2}{m}|\psi_j|^2\mathbf{A}\right]$

If phase winding only in one of the bands,

$$\mathbf{J} = \frac{\hbar e}{m}|\psi_1|^2\nabla\theta_1 - \sum_j\frac{2e^2}{m}|\psi_j|^2\mathbf{A}$$

$$\Phi = \oint_\sigma \mathbf{A}\cdot d\mathbf{r} = \frac{|\psi_1|^2}{\sum_j|\psi_j|^2}\Phi_0$$

If $|\psi_j|^2$ has different temperature dependence,
Φ is non-universally un-quantized

- Non-integer
- Temperature dependent

E. Babaev, *PRL* 89, 067001 (2002)

Defined by nonuniversal material parameters

Flux-unquantized vortices

Free energy in U(1)xU(1) model (no Josephson coupling terms)

$$F = \frac{1}{2}|(\nabla + iq\mathbf{A})\psi_1|^2 + \frac{1}{2}|(\nabla + iq\mathbf{A})\psi_2|^2 + \frac{(\nabla\times\mathbf{A})^2}{2} - a_1|\psi_1|^2 + \frac{b_1}{2}|\psi_1|^4 - a_2|\psi_2|^2 + \frac{b_2}{2}|\psi_2|^4 .$$

For supercurrent then we get $\mathbf{J} = \frac{iq}{2}(\psi_1^*\nabla\psi_1 - \psi_1\nabla\psi_1^*) + \frac{iq}{2}(\psi_2^*\nabla\psi_2 - \psi_2\nabla\psi_2^*) - q^2\rho^2\mathbf{A}$

, where $\rho^2 \equiv |\psi_1|^2 + |\psi_2|^2$

and superconducting penetration depth $\lambda = \frac{1}{|q|\rho}$.

$$F = \frac{1}{2\rho^2}|\psi_1|^2|\psi_2|^2\left[\nabla(\theta_1-\theta_2)\right]^2 + \frac{1}{2\rho^2}\left(|\psi_1|^2\nabla\theta_1 + |\psi_2|^2\nabla\theta_2 + q\rho^2\mathbf{A}\right)^2 + \frac{(\nabla\times\mathbf{A})^2}{2}$$

$U(1)\times U(1)$ Topological defects

	(M_1, M_2)	$\Phi_{(M_1,M_2)}$
Elementary vortices	$\pm(1,0)$	$\pm\sin^2(\frac{\beta}{2})\Phi_0$
	$\pm(0,1)$	$\pm\cos^2(\frac{\beta}{2})\Phi_0$
Composite vortices	$\pm(1,1)$	$\pm\Phi_0$
	$\pm(1,-1)$	$\mp\cos(\beta)\,\Phi_0$

E. Babaev, *PRL* 89, 067001 (2002)

- ✓ For (*1, 1*)-vortices, the first term is zero, while the second term acquires the form equivalent to a single-component superconductor.
- ✓ Then for a single component vortex (1,0) the current is $\mathbf{J} = -q\rho^2\left[\sin^2(\beta/2)\,\nabla\theta_1 + q\mathbf{A}\right]$

with $\sin(\beta/2) = |\psi_1|/\rho$, and $\cos(\beta/2) = |\psi_2|/\rho$. Then $\Phi_{(1,0)} = \oint_\sigma \mathbf{A}\cdot d\mathbf{l}$ ➡ $\Phi_{(1,0)} = \sin^2(\beta/2)\,\Phi_0$

with $\Phi_0 = -2\pi/q$

Repulsion force between fractions induced by domain walls in U(1)xZ_2 with Josephson coupling

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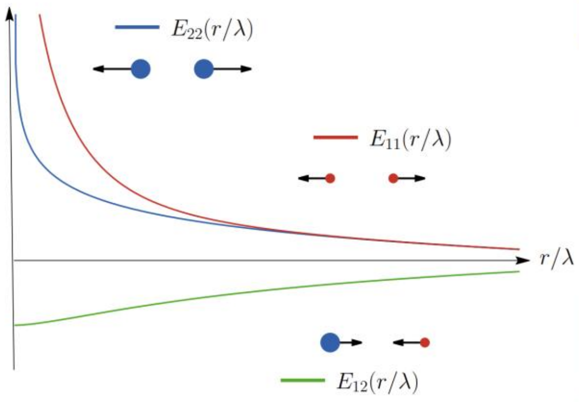
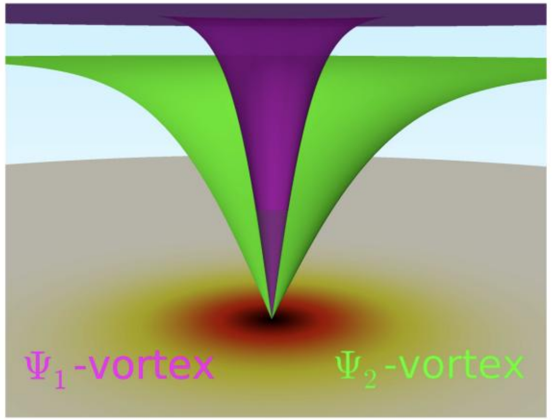
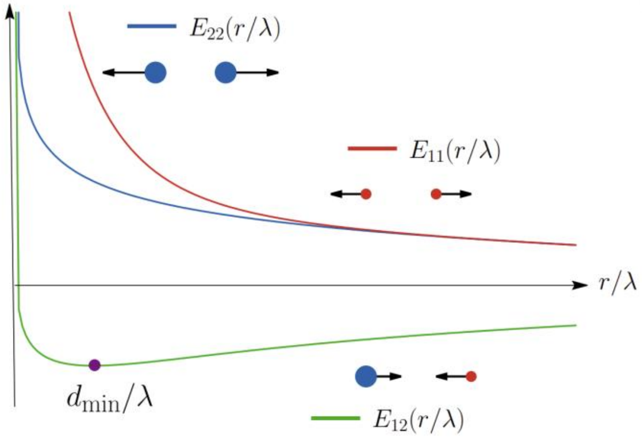
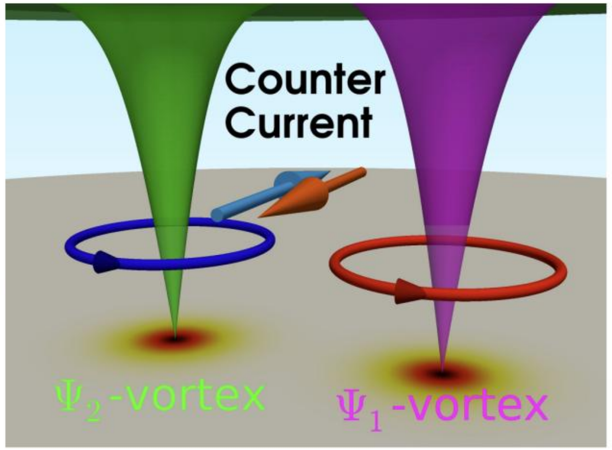
Normally, vortices neve split due to attractive potential between fractions.

The repulsion at short distances between different fractions appears when the vortex is located at the domain wall.









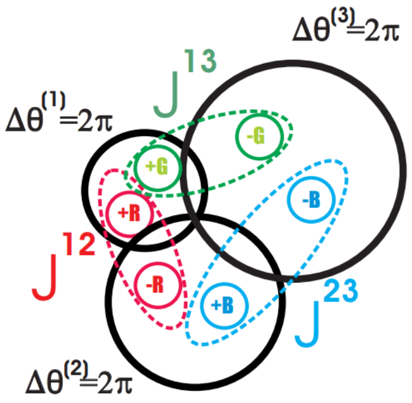
The blue (big) dot represents the vortex in ψ_1 while the red (small) dots represent the vortices in ψ_2.

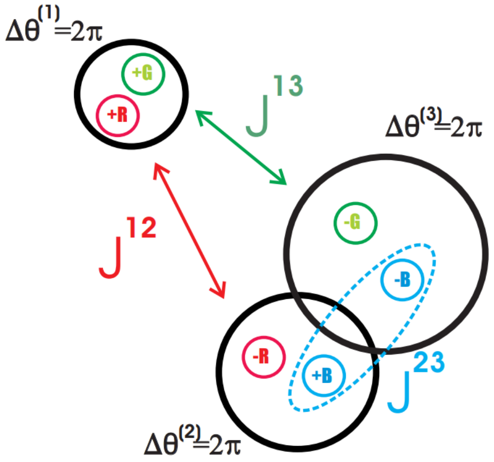
Julien Garaud, thesis, 2022

Vortices in more 3-component superconductors

Unquantized vortices are confined in analogy with quarks.

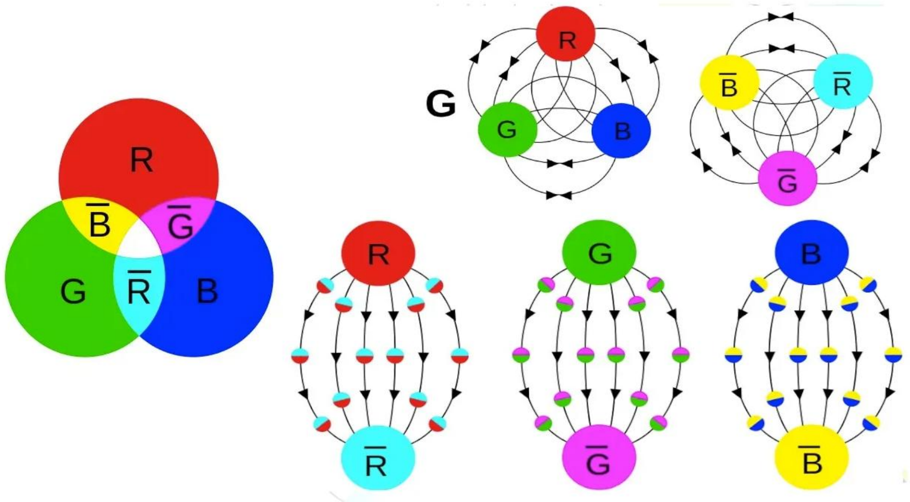
Composite vortex in 3-component superconductor





Physical Review B 71 (21), 214509 (2005)

Quarks confinement



https://medium.com/starts-with-a-bang/there-are-no-free-quarks-ddec8cb831ea

Unquantized vortices in multicomponent superconductors - historical outline

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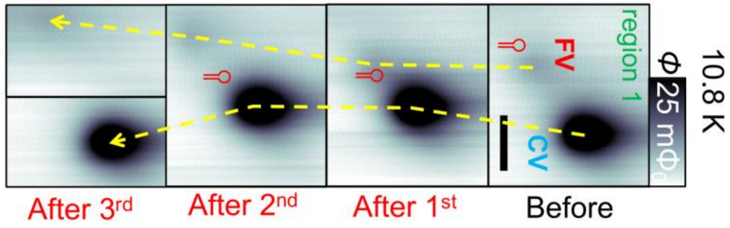
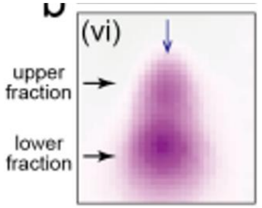
✓Flux unquantized vortices were predicted in 2002 [E. Babaev, PRL 89, 067001 (2002)].

$$\Phi = \oint_{\sigma} \mathbf{A} \cdot d\mathbf{r} = \frac{|\psi_1|^2}{\sum_j |\psi_j|^2} \Phi_0$$

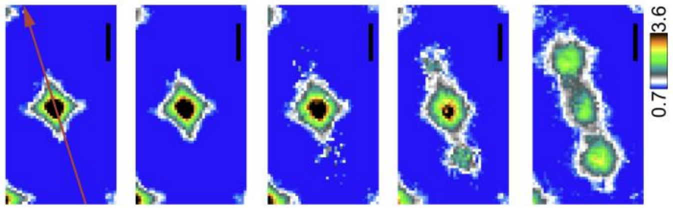
✓Flux unquailed vortices were discovered in 2023 using SQUID magnetometry [Y. Iguchi *et al. Science* **380**,1244-1247(2023)];



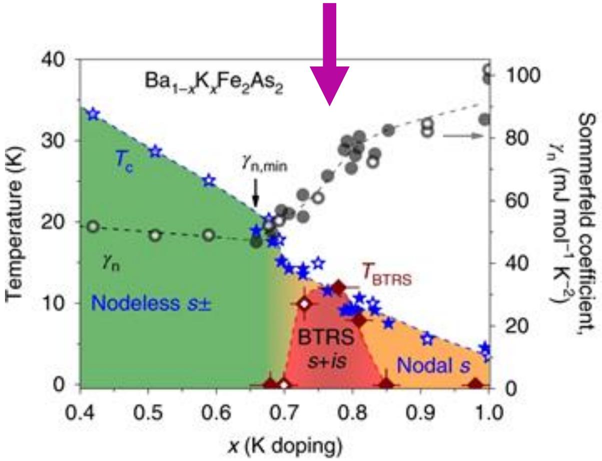
✓Integer-vortex flux fractionalization were observed in 2024 using SQUID magnetometry [Q. Z. Zhou *et al*., arXiv:2408.05902];



✓Integer-vortex core fractionalization were observed in 2024 using STM [Yu Zheng, Quanxin Hu *et al*., arXiv:2407.18610; *Science* (2026)];



Discovery of flux unquantized vortices in the BTRS state using Scanning SQUID Microscopy



V. Grinenko et al., *Nat. Phys.* 16, 789–794 (2020)

$$\Phi = \oint_\sigma \mathbf{A} \cdot d\mathbf{r} = \frac{|\psi_1|^2}{\sum_j |\psi_j|^2} \Phi_0$$

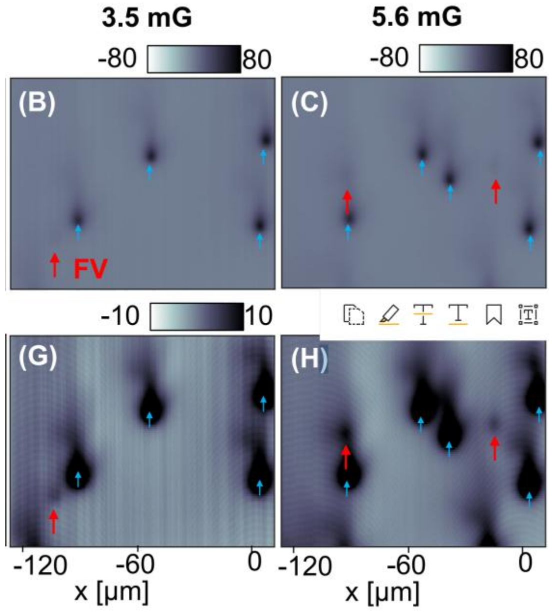
E. Babaev, *PRL* 89, 067001 (2002)

Stanford University

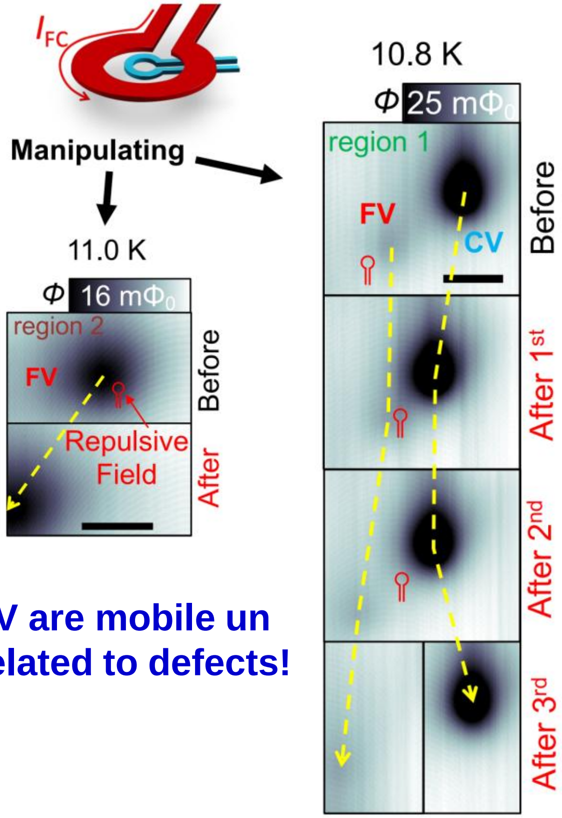
Yusuke Iguchi

Kathryn A. Moler

Support for multicomponent superconductivity!



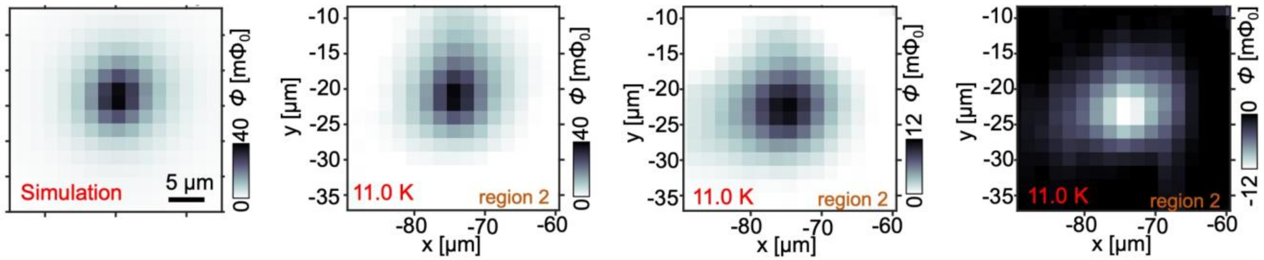
~ 10% of vortices are fractional vortices (FV)



FV are mobile un related to defects!

Abrikosov vortices

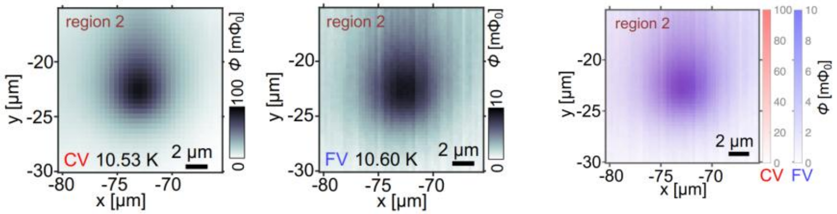
Fractional vortices



Total flux	1.02 Φ_0	0.99 Φ_0	0.36 Φ_0	-0.41 Φ_0

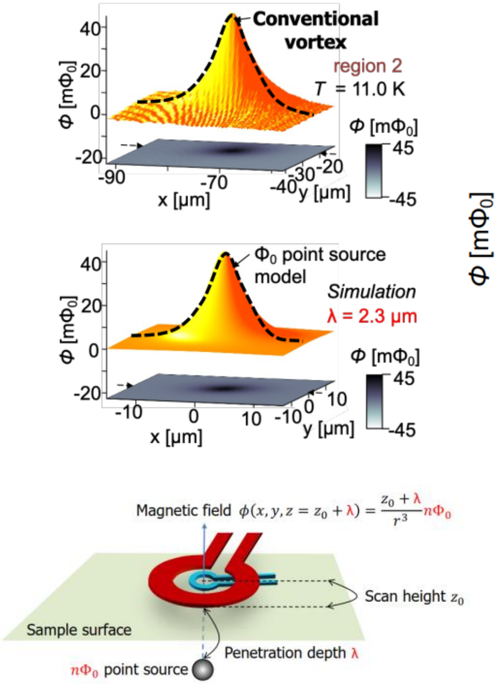
Y. Iguchi *et al.* *Science* **380**,1244-1247(2023).

Abrikosov and Fractional vortices pinned at the same location.

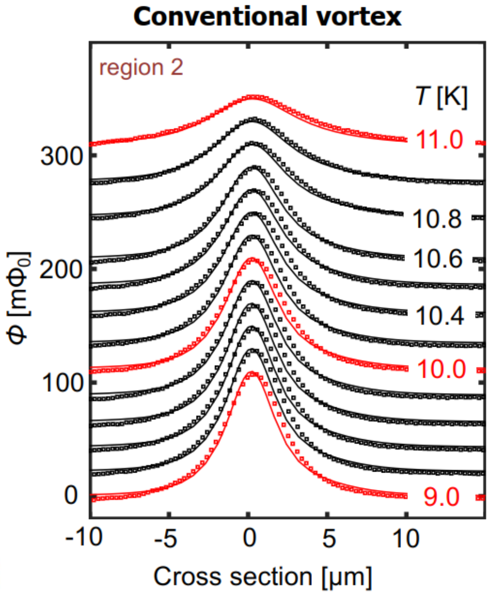


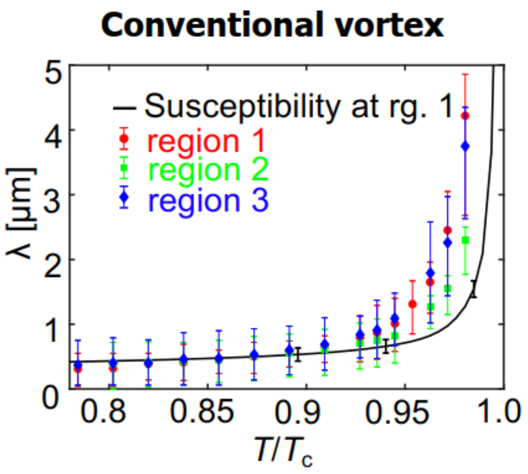
***FV* – quantum objects unrelated to specific defects in the sample.**

Temperature dependent fractions

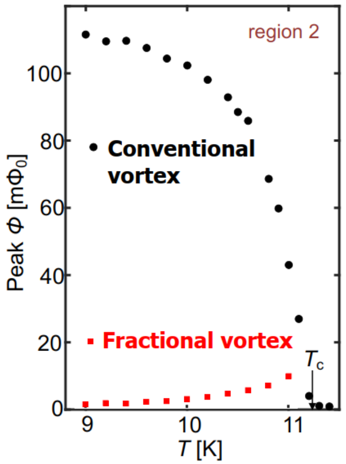


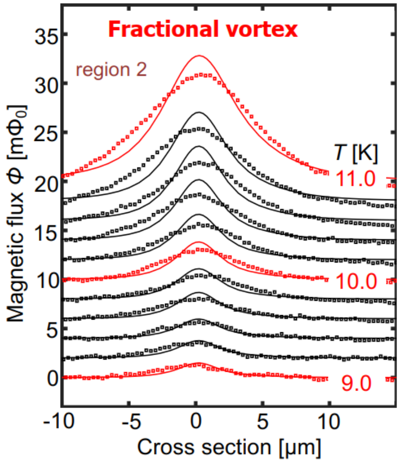
J.R. Kirtley *et al., Supercond. Sci. Technol.* 29, 124001(2016).

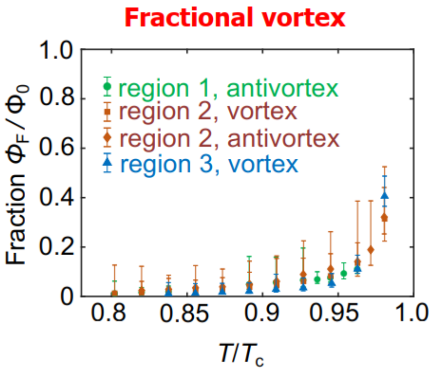




The expected temperature dependence for the penetration depth.







Reproducible result!

Point source model fitting is not perfect
→Delocalization of fractional vortex

→consistent with the theory: E. Babaev *et al.*, *PRL* 103, 237002 (2009)

The magnetic flux of fractional vortices is temperature dependent!

Is it consistent with a multiband model?

Y. Iguchi *et al. Science* **380**,1244-1247(2023)

Fractional vortices: Open questions.

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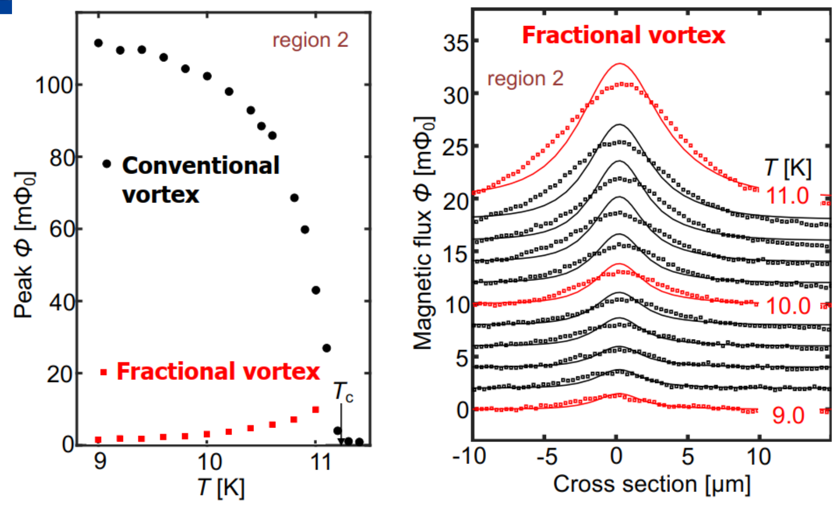
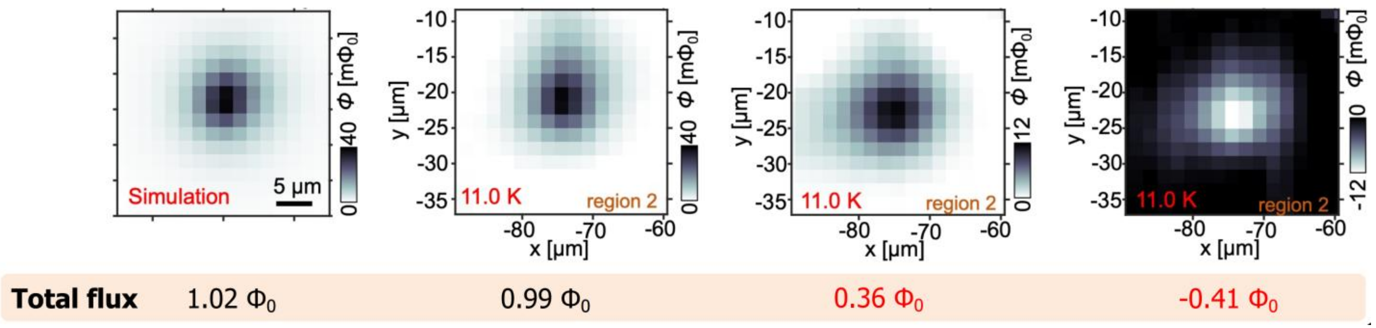
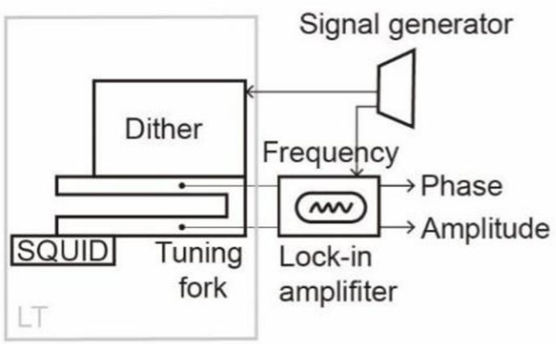
- Are the unquantized vortices stable at low temperatures?
- If unquantized vortices with $\Phi_{FV1} = a\Phi_0$ ($a < 1$) are the result of integer vortex fractionalization they must have counterparts with $\Phi_{FV2} = b\Phi_0$, where $a + b = 1$.

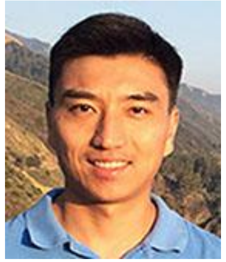


Y. Iguchi *et al.* *Science* **380**,1244-1247(2023).

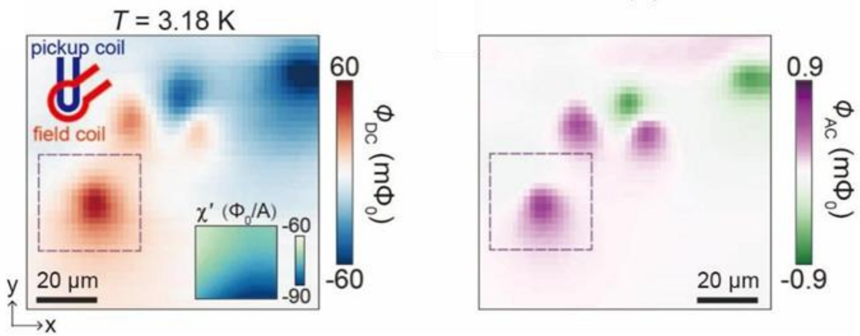


AC scanning SQUID magnetometry of $Ba_{0.23}K_{0.77}Fe_2As_2$

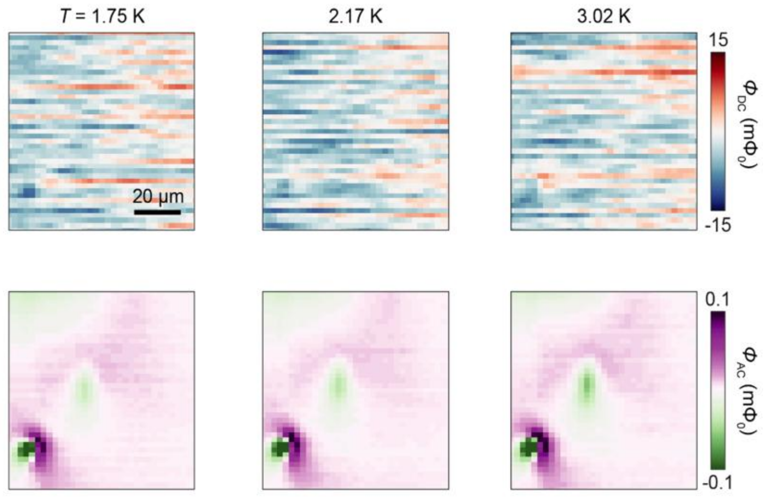




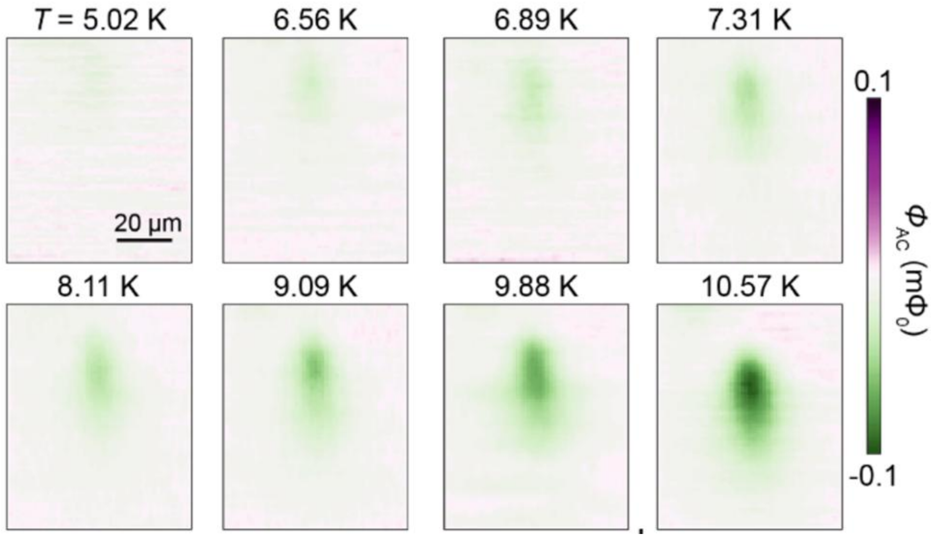
Yihua Wang



The nano-SQUID chip is mounted on a quartz tuning fork which oscillates at its resonant frequency under the drive of a dither piezo.



Fractional vortices at $T \ll T_c$ can be resolved in AC mode only.



The flux of unquantized vortices grows with temperature in accord with the previous study.

Integer vortex fractionalization in $Ba_{0.23}K_{0.77}Fe_2As_2$

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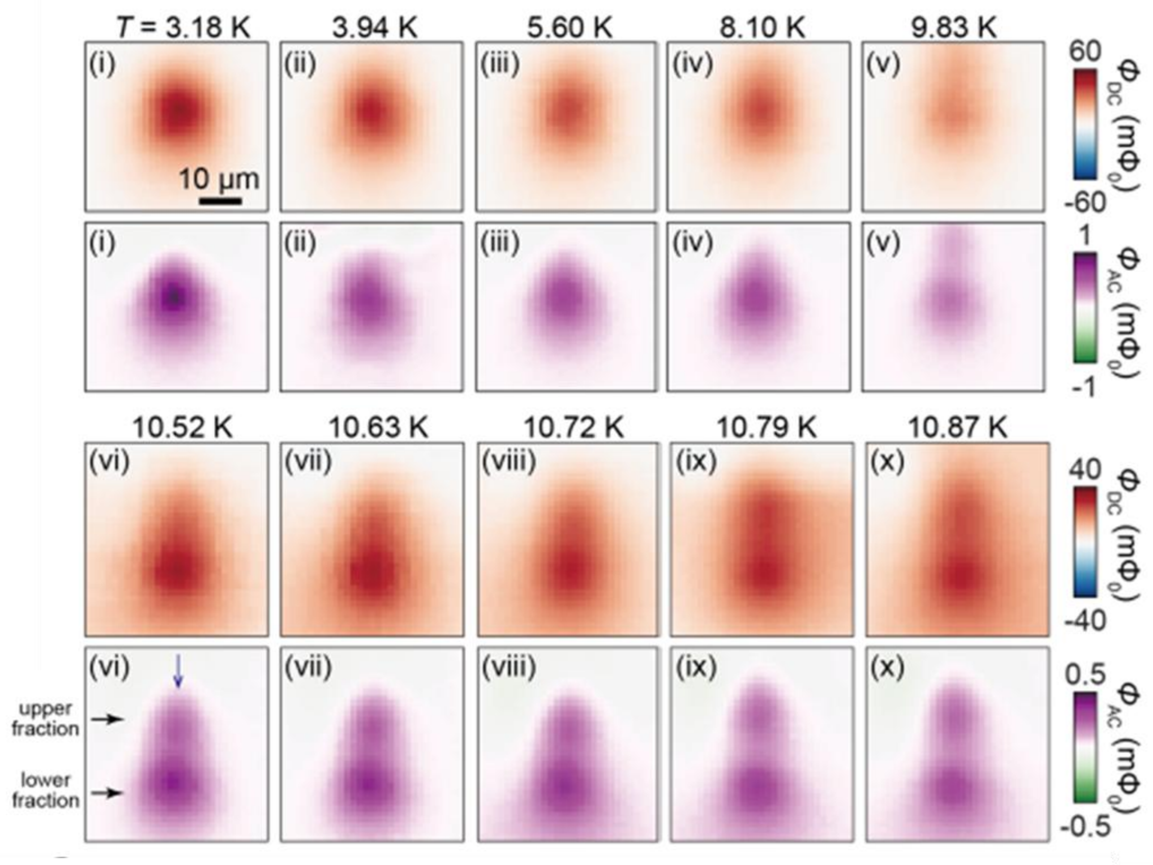
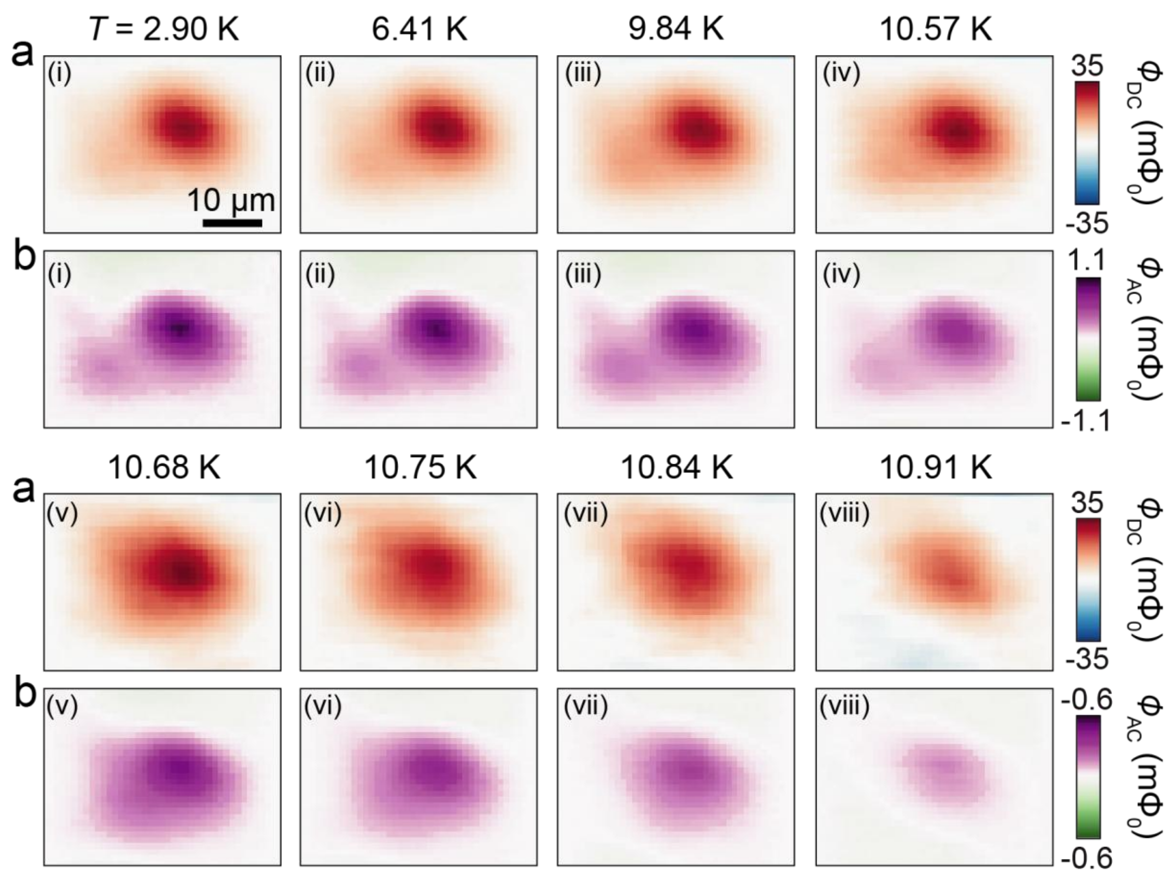
Integer vortex fractionalization with temperature increase.



Fractional vortices merging with temperature increase.



- Here the fractionalization is randomly process driven by thermal fluctuations and pinning.

Q. Z. Zhou *et al*., arXiv:2408.05902

Integer vortex fractionalization in $Ba_{0.23}K_{0.77}Fe_2As_2$

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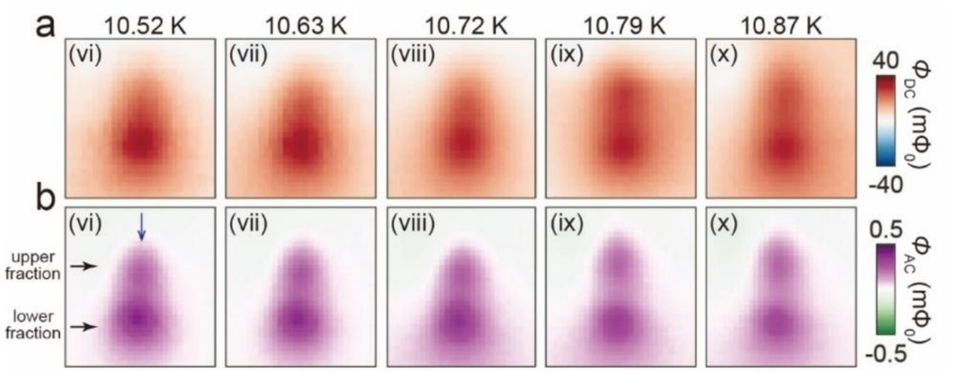
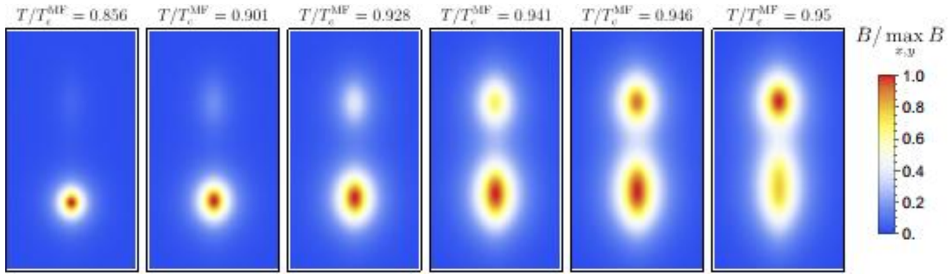
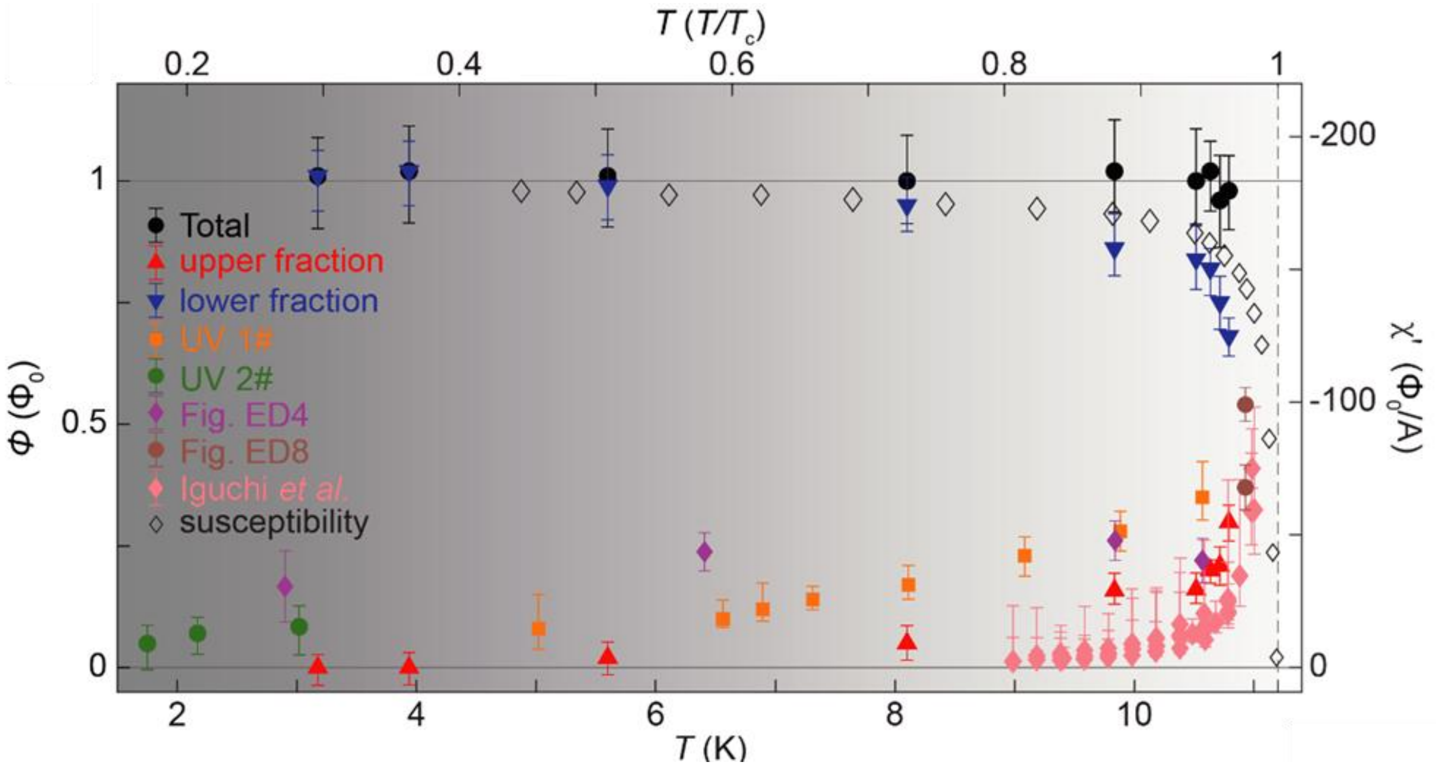
Seesaw effect (experiment)



Seesaw effect (GL theory)



Flux of fractionalized vortices vs. temperature

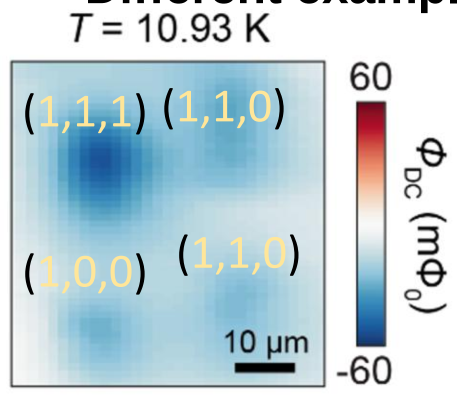


- The flux of individual fractions is strongly temperature depended but total flux of split fractions is equal to $\Phi_0 = h/2e$ (seesaw effect).

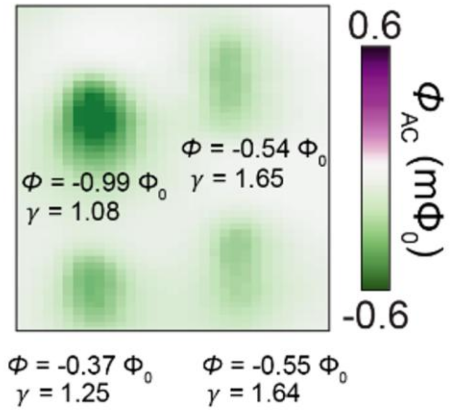
Q. Z. Zhou *et al.*, arXiv:2408.05902

Integer vortex fractionalization in $Ba_{0.23}K_{0.77}Fe_2As_2$

Different examples of unquantized vortices

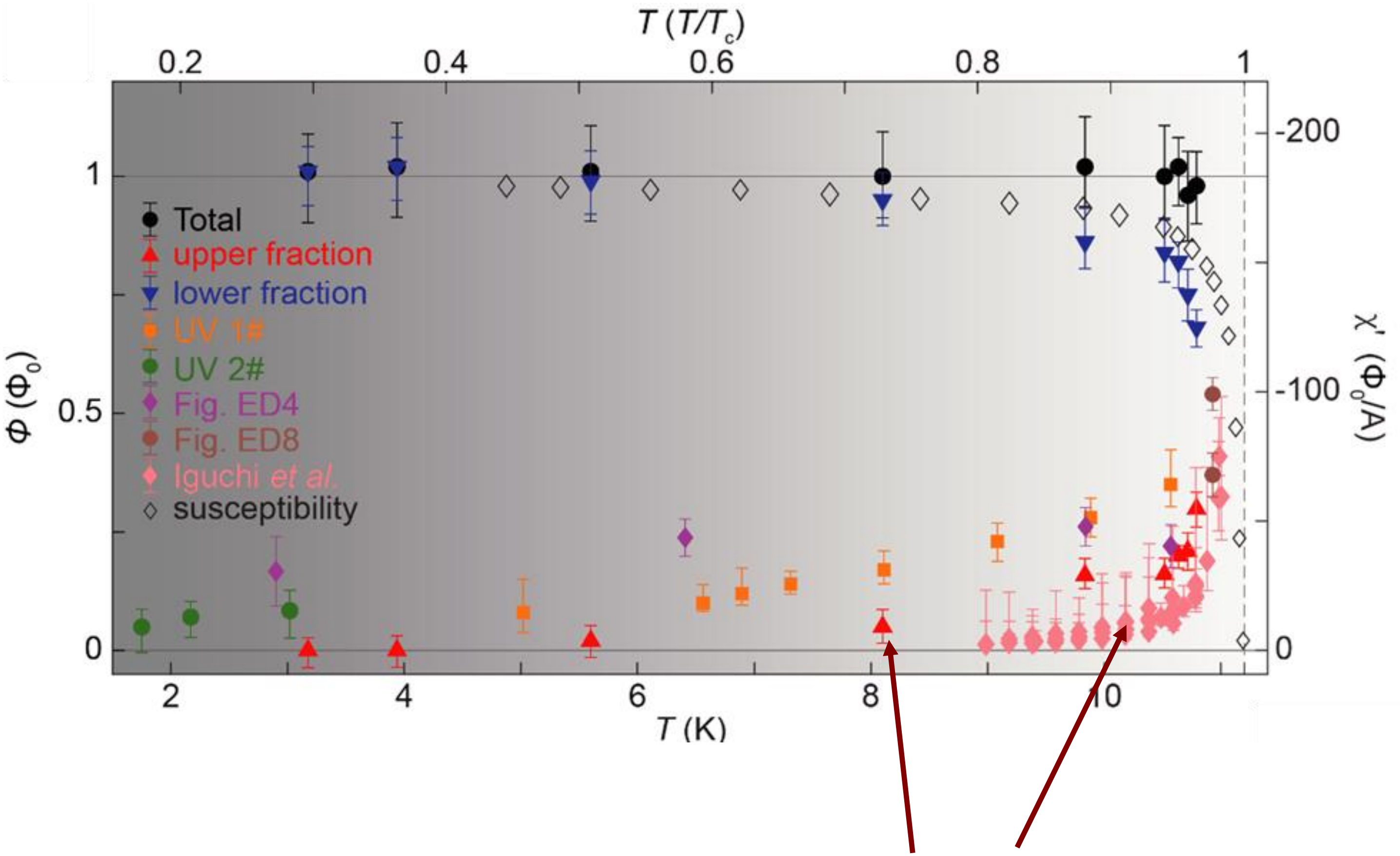


We observe two types fractions with nearly doubled flux values we attribute them to (1,1,0) and (1,0,0) vortices.



(1,1,0) fractions have more pronounced elongation presumably indicating two nearly split fractions.

Flux of fractionalized vortices vs. temperature

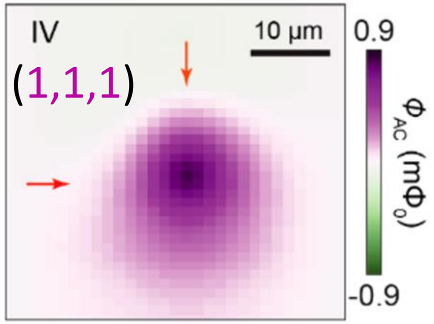


- The flux of isolated fractional vortices increases with temperature consistently with the previous observation, but there is no quantitative agreement → very high sensitivity to material parameters.

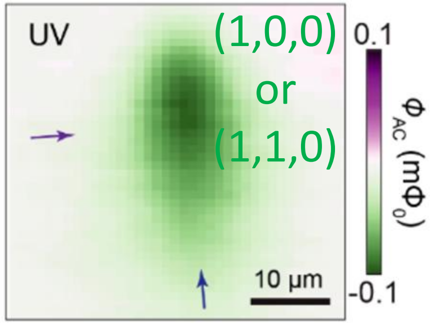
Q. Z. Zhou *et al.*, arXiv:2408.05902

Elongation of unquantized vortices

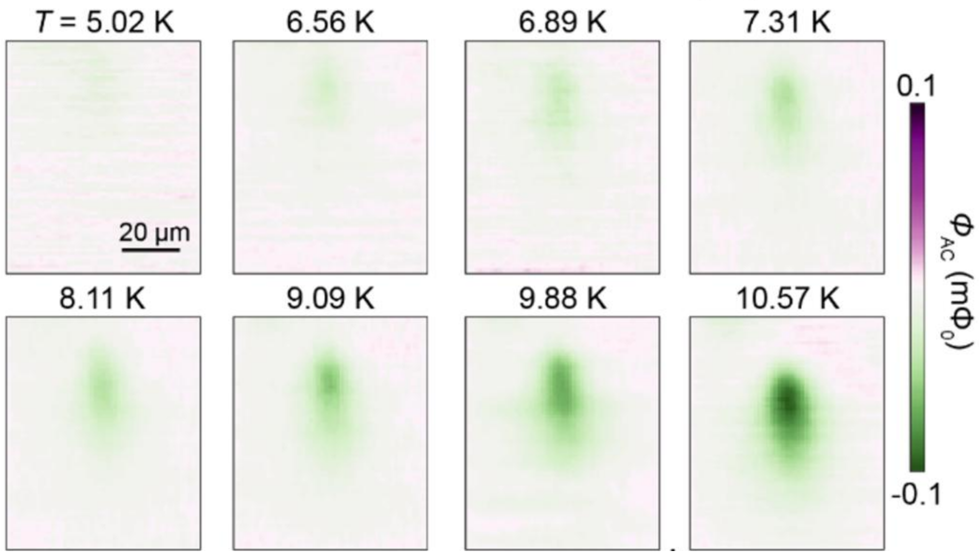
Integer vortex

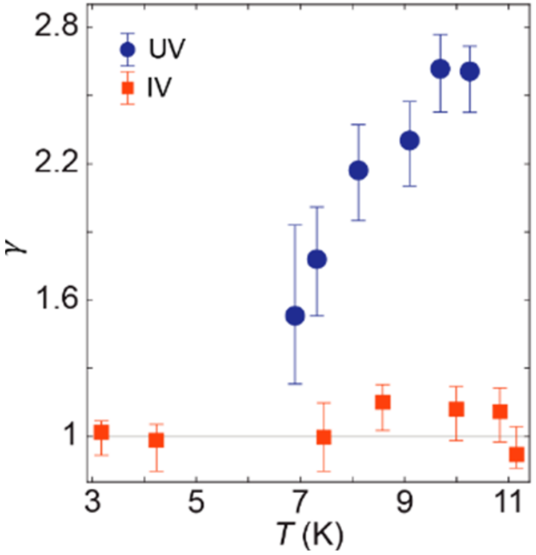


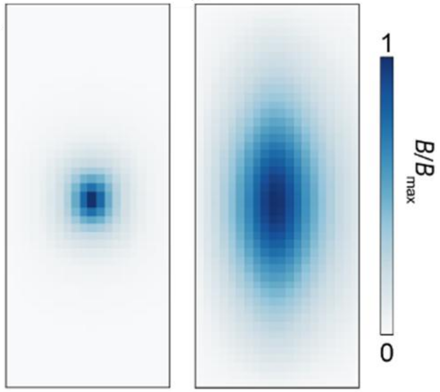
Unquantized vortex



Unquantized vortices elongates with increasing of temperature







Three-band Bogoliubov-de-Gennes model

Unquantized vortices must locate at the domain wall which results in elongation (intrinsic elongation)

Elongation due to incomplete fractionalization of composite fractions (1,1,0).

Integer vortex fractionalization in multicomponent superconductors

Tsung-Dao Lee Institute

Applied ⊗ magnetic field

Integer vortex

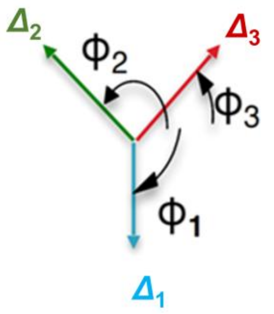
$\Phi_0 = h/2e$

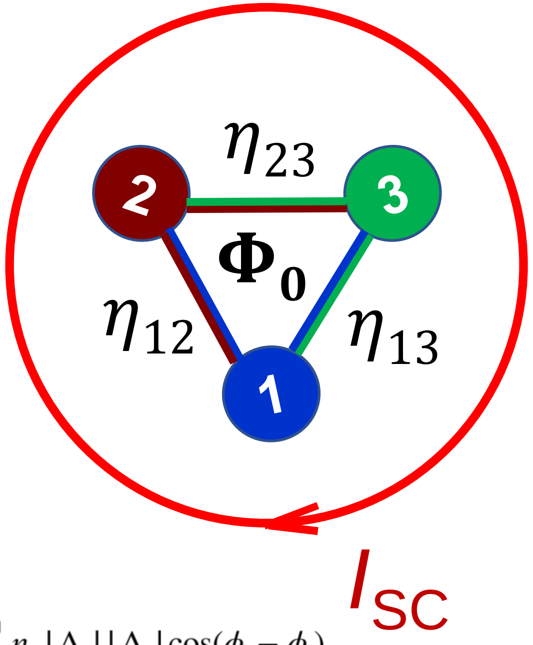
Fractional vortices

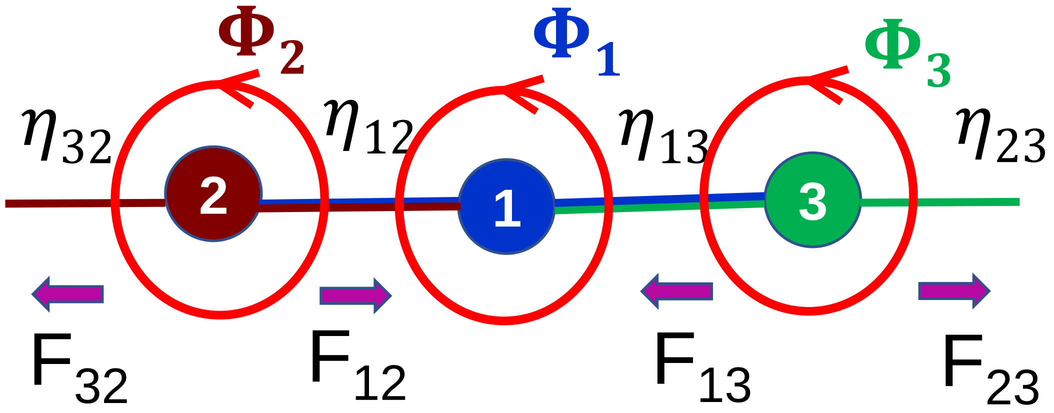
$\Phi_0 = \Phi_1 + \Phi_2 + \Phi_3$ and $\Phi_1 \neq \Phi_2 \neq \Phi_3$

Order parameter

$\psi_\alpha = |\Delta_\alpha| e^{i\Phi_\alpha}$







$$\sum_{i>j} \frac{|\Delta_i|^2 |\Delta_j|^2}{2\rho^2} \left[\vec{\nabla}(\phi_i - \phi_j) \right]^2 + \sum_{i>j} \eta_{ij} |\Delta_i| |\Delta_j| \cos(\phi_i - \phi_j)$$

TRSB

Internal Josephson coupling

- ✓ Josephson coupling of fractions, resulting in a strong attractive potential that holds the fractions together.

- ✓ The vortex splitting at the domain wall.

Integer vortex fractionalization in multicomponent superconductors

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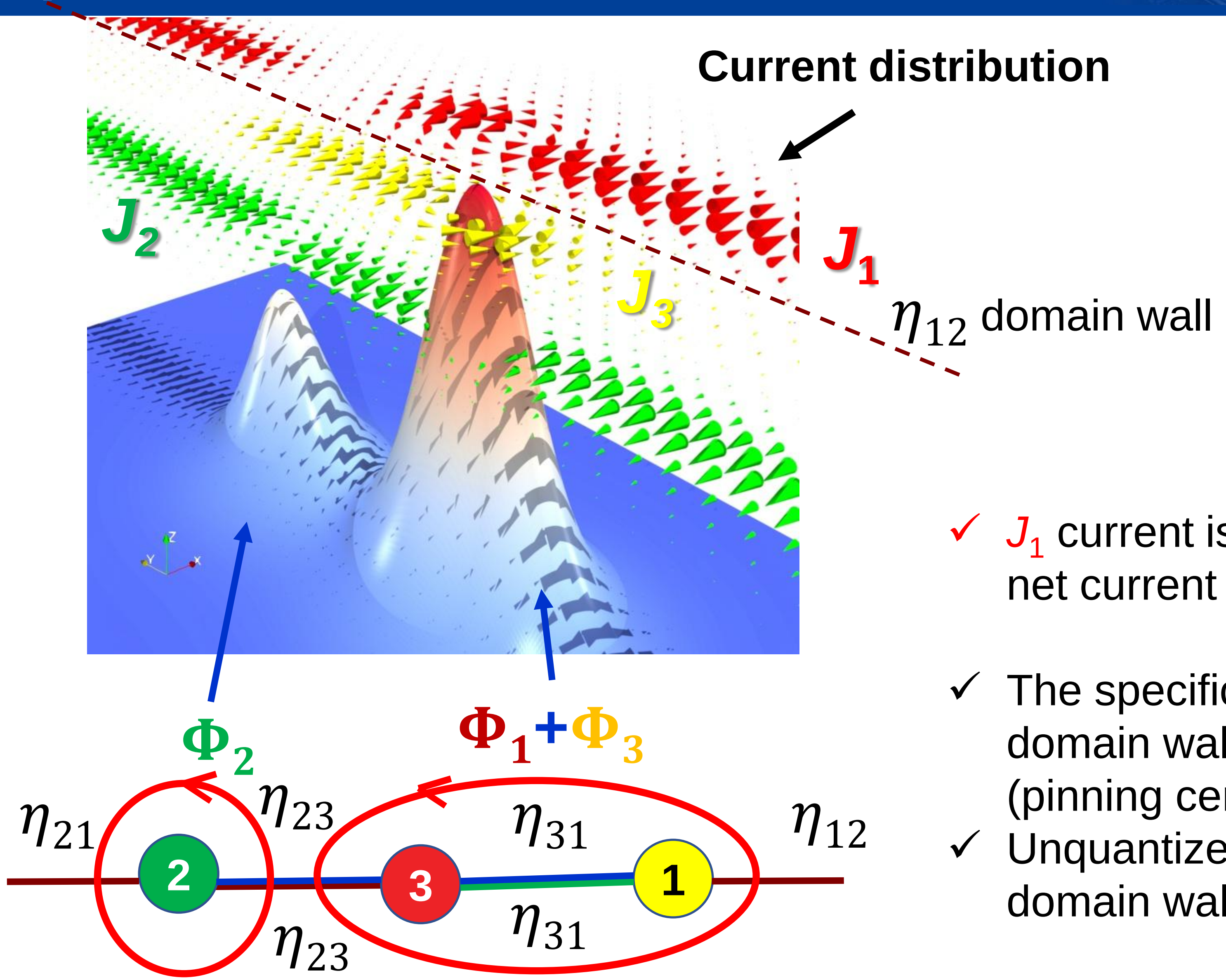

Julien Garaud,
CNRS-UMR, France

Anton Talkachov,
KTH

- ✓ J_1 current is coupled to J_2 current to keep overall net current along the domain wall zero.
- ✓ The specific position of the split fraction on the domain wall is stabilized by inhomogeneity (pinning center).
- ✓ Unquantized vortices are elongated along the domain wall.

Stable unquantized (1,0,0) vortex at the domain wall: microscopic demonstration (3-band Bogoliubov de Gennes model solution).

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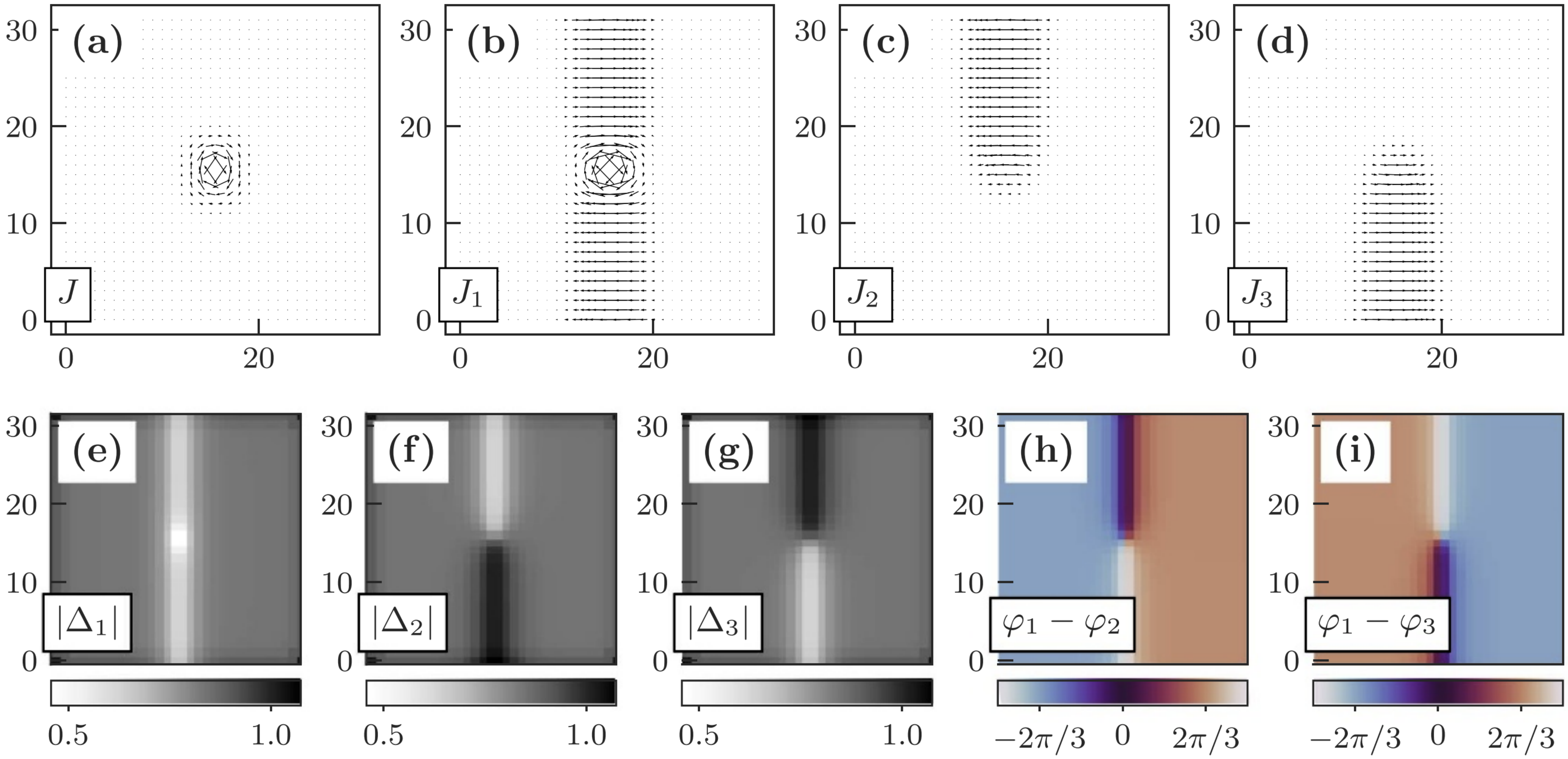

Igor Timoshuk and Egor Babaev, New J. Phys. 27 (2025) 123501

Unquantized vortices: Open questions.

Onsager-Feynman relation → one vortex core per flux quantum

What is about the unquantized vortex cores?

STM experiments: Surface reconstruction and disorder in $Ba_{0.23}K_{0.77}Fe_2As_2$

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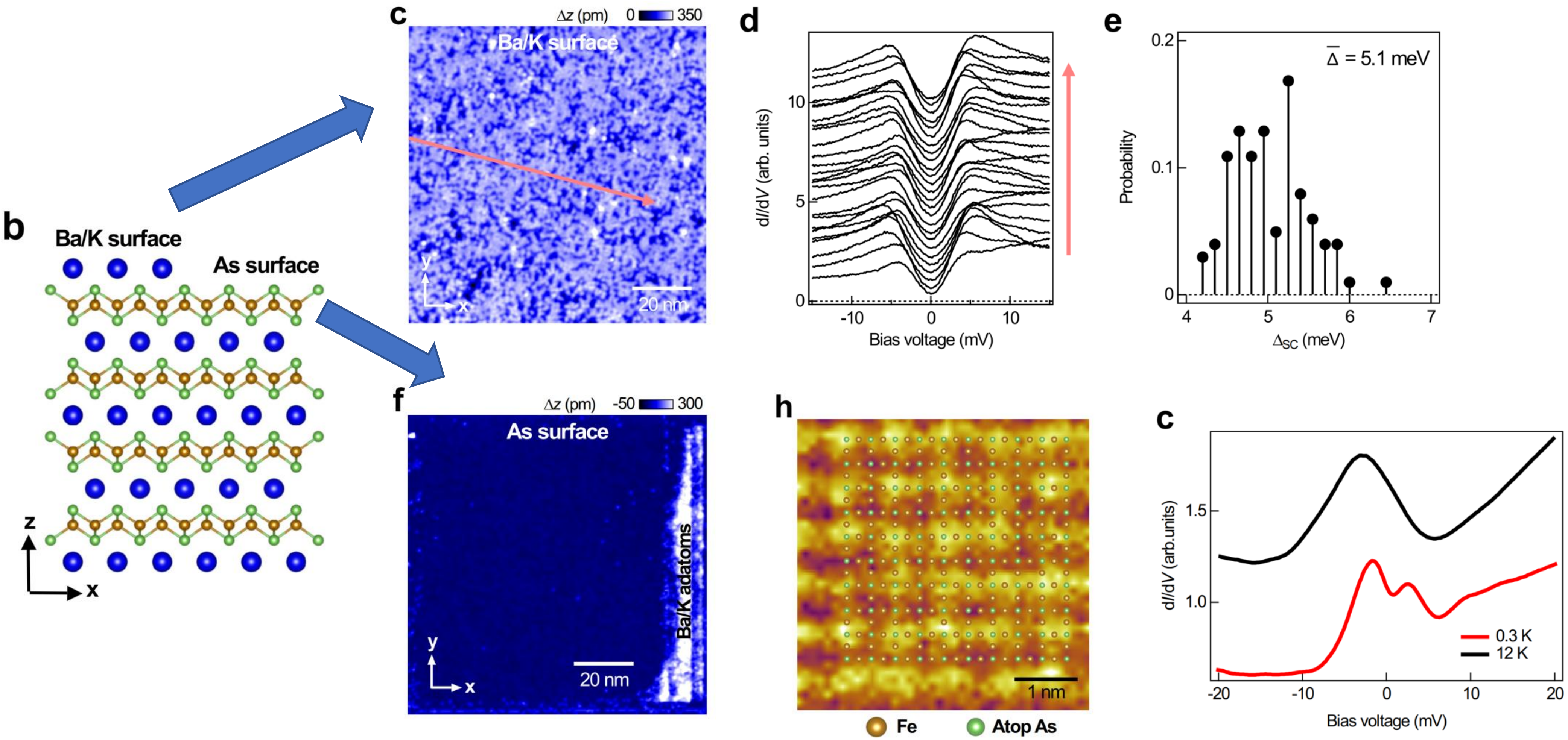
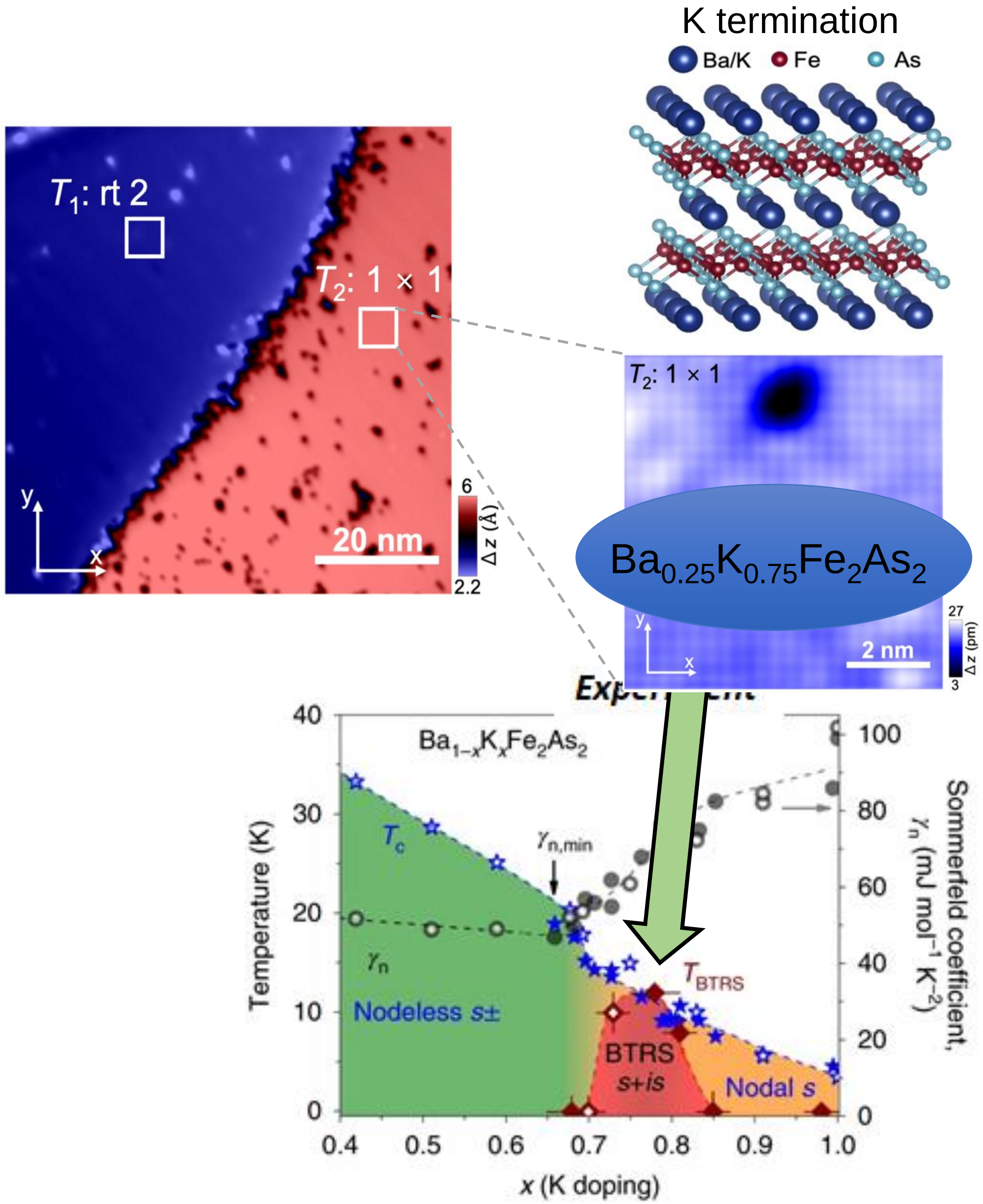


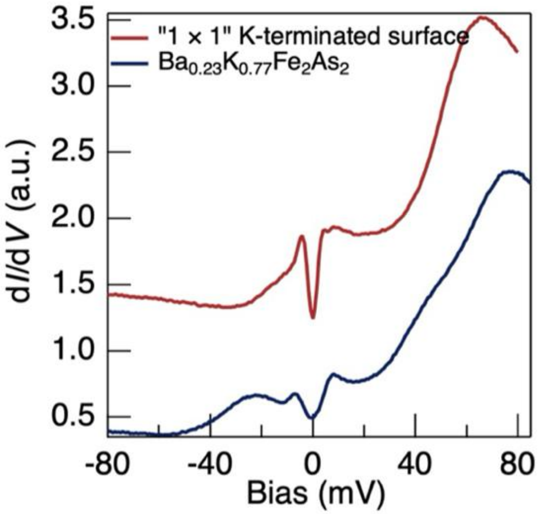
Quanxin Hu

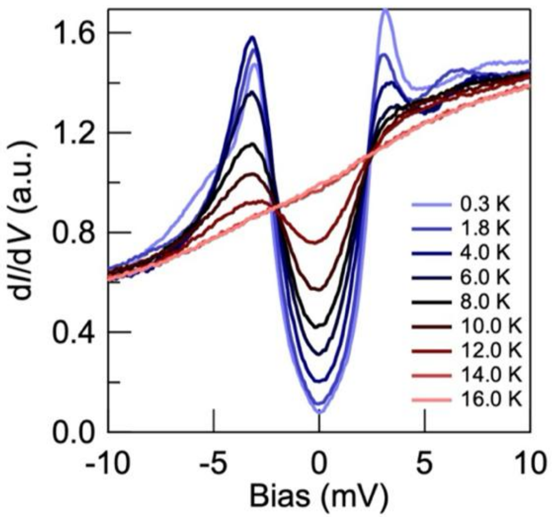
- Initially, we searched for fractional vortices in the same sample where they were observed in scanning SQUID experiments.
- The vortices was not resolved on the disordered Ba/K surface.
- As surface is non-superconducting – exabits a charge density wave (CDW) state.

I. Shipulin *et al*., *Nat Commun* **14**, 6734 (2023).
Q. Hu, Y. Zheng et al. Nat Commun 16, 253 (2025).

Surface doping on K-terminated surface of KFe_2As_2







- ✓ 1x1K surface has nearly the same characteristic spectra as $Ba_{0.23}K_{0.77}Fe_2As_2$ sample.
- ✓ 1x1K surface has a large gap which close at ~16 K. Bulk T_c of the KFe_2As_2 is 3.4 K only!

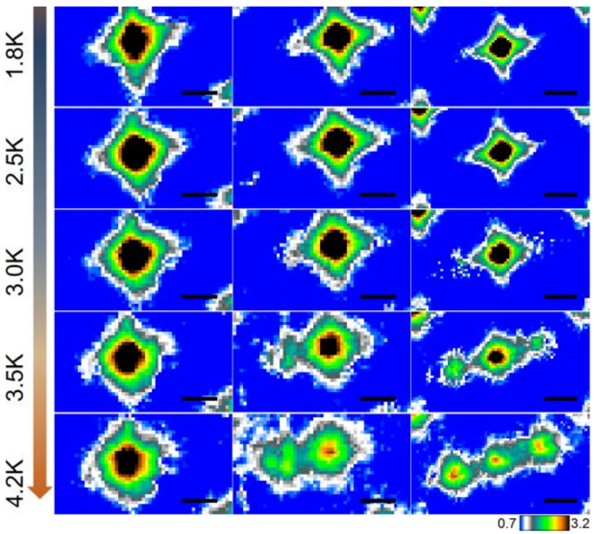
We concluded that extra electron doping resides within a single unit cell layer.

Yu Zheng, Quanxin Hu *et al.*, arXiv:2407.18610; Science (2026)

STM observation of vortex core fractionalization at K-terminated surface of KFe_2As_2

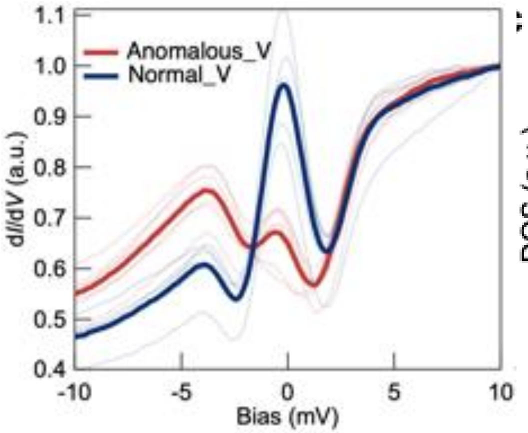
The key observations:

1. Vortex core fractionalization.

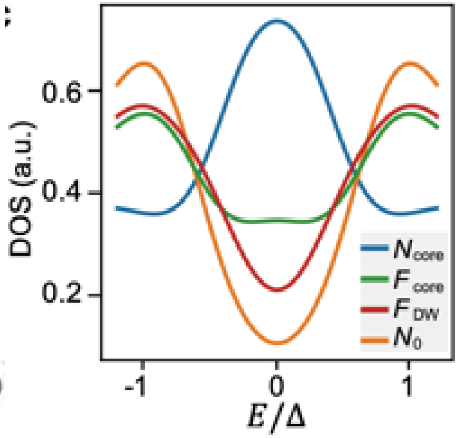


2. Vortex core of fractional vortices is spectroscopically different from the integer vortices.

Experiment



Theory



Quanxin Hu

Yu Zheng

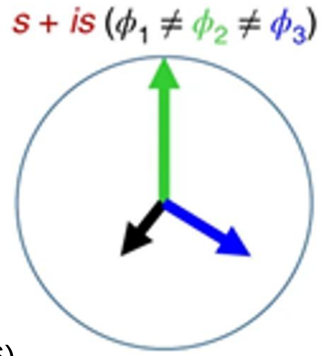
Hong Ding

Chi Ming Yim

✓ **One quanta (conventional) vortices spits into 2 or 3 fractions**!

✓ Counting the number of vortices indicates a significant increase in the number of cores with the temperature increase at the fixed applied field.

✓ **The result is consistent with 3-component order parameter!**

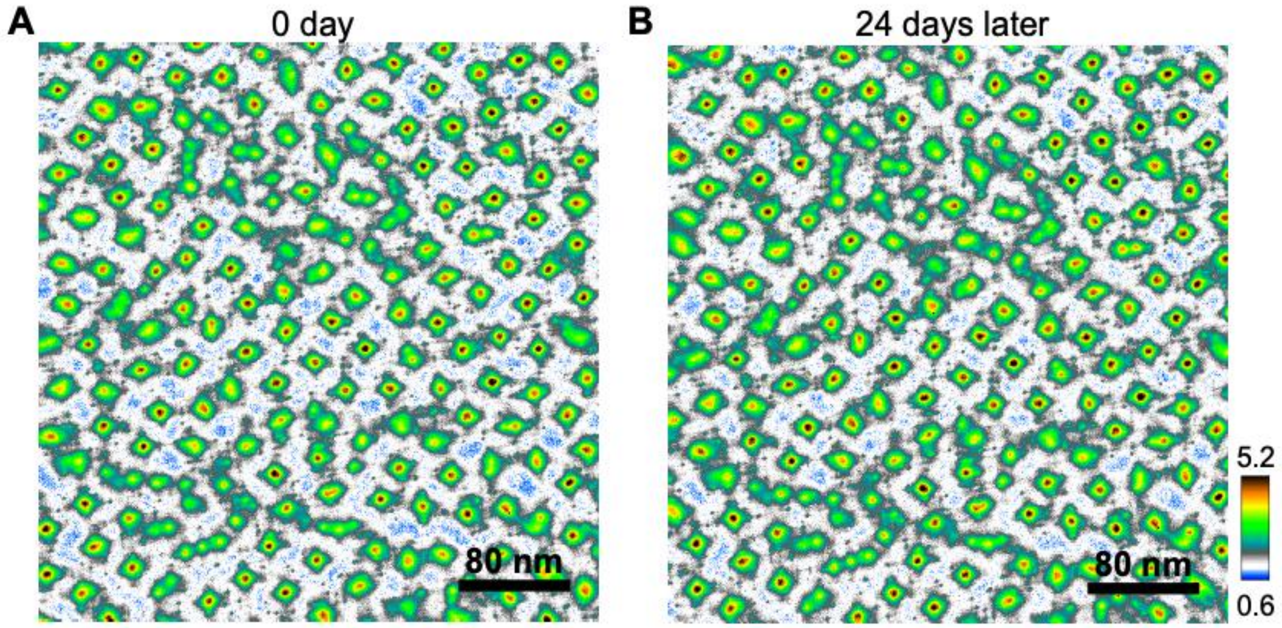


Yu Zheng, Quanxin Hu *et al.*, arXiv:2407.18610; Science (2026)

Unquantized vortices are not a consequence of integer vortex fluctuations

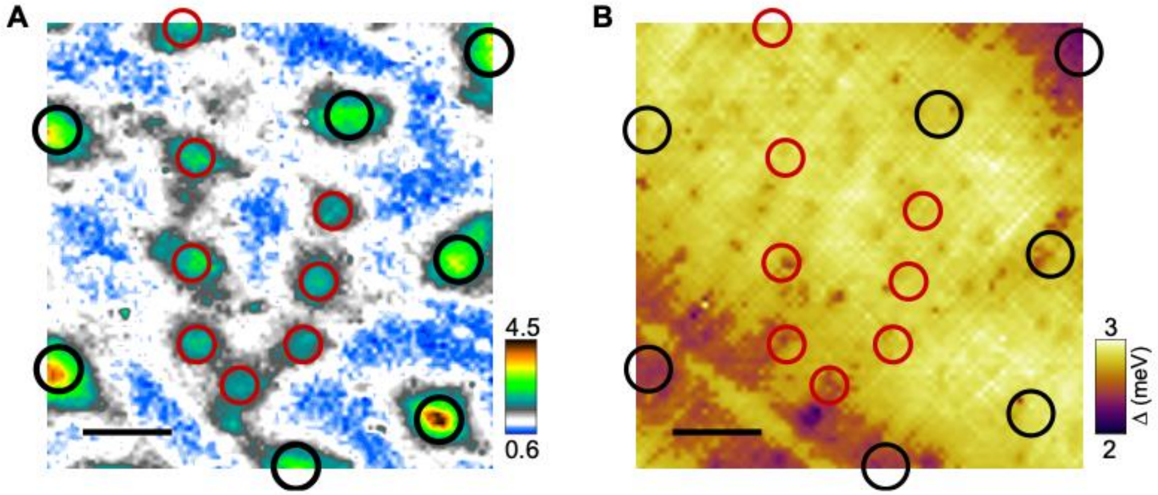
李政道研究所
TSUNG-DAO LEE INSTITUTE

T = 4.2 K the same surface at different experimental cycles at 2T



Vortices

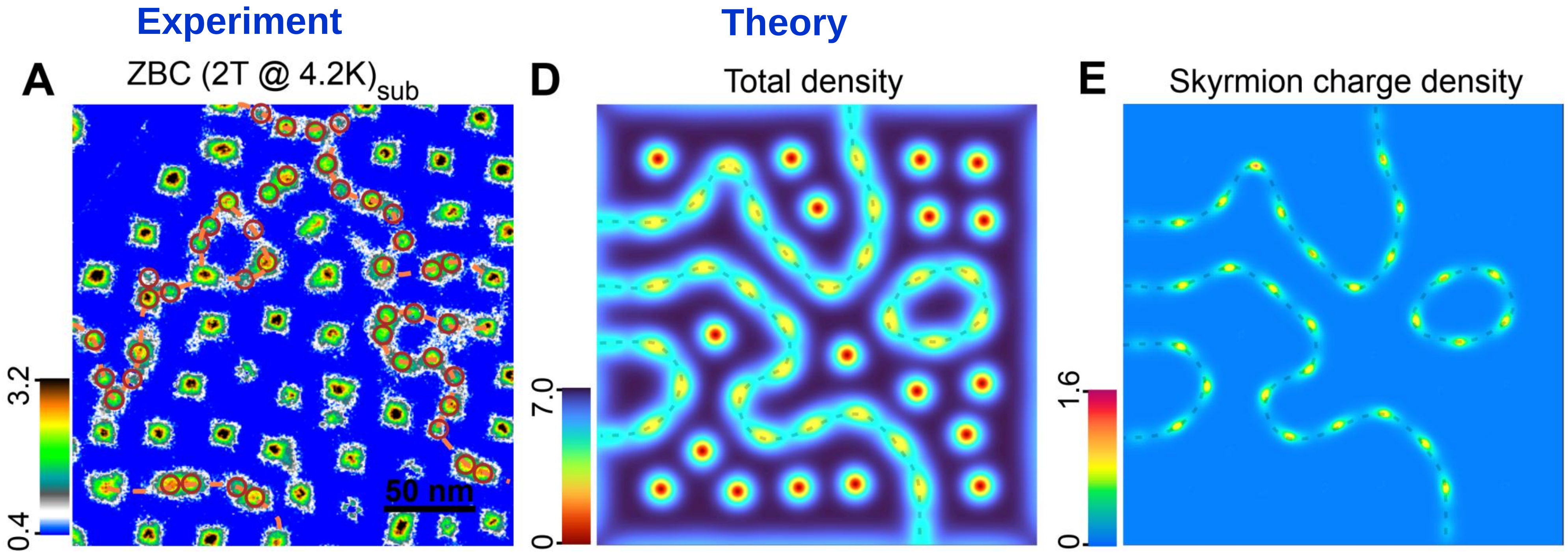
Superconducting-gap map in zero field



- ✓ Unquantized vortices are very stable at constant temperature → not a simple fluctuation effect.
- ✓ The positions of unquantized vortices are unrelated to the locations of inhomogeneities → the vortex positions are stabilised by the pinning landscape and interactions with other vortices.

Yu Zheng, Quanxin Hu *et al.*, arXiv:2407.18610; Science (2026)

STM observation of vortex chains at K-terminated surface of KFe_2As_2



$$B_{sk} = \frac{i}{e\varrho^4}[\varrho^2 \nabla\Psi^\dagger \times \nabla\Psi + (\Psi^\dagger\nabla\Psi) \times (\nabla\Psi^\dagger\Psi)].$$

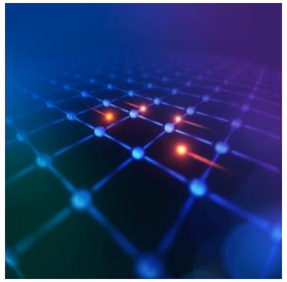
$$Q_{sk} = \int_S \frac{eB_{sk}}{2\pi} \cdot dS.$$

- ✓ The unquantized vortex chins are new type of topological defects in superconductors
- ✓ They carry a topological charge of CP2 Skyrmions.

Unquantized vortex chains align along the superconducting domain walls.

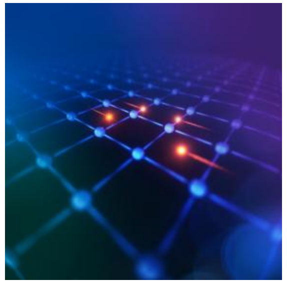
Electron-Quadrupling condensates

BCS paradigm: electrons can form only pairs.



Could there be electron-quadrupling condensates?

Electron-Quadrupling condensates



BTRS-SC U(1)xZ_2

$< \Delta_i > \neq 0$

$< \Delta_i \Delta_j^* > \neq 0$

Quadrupling phase Z_2

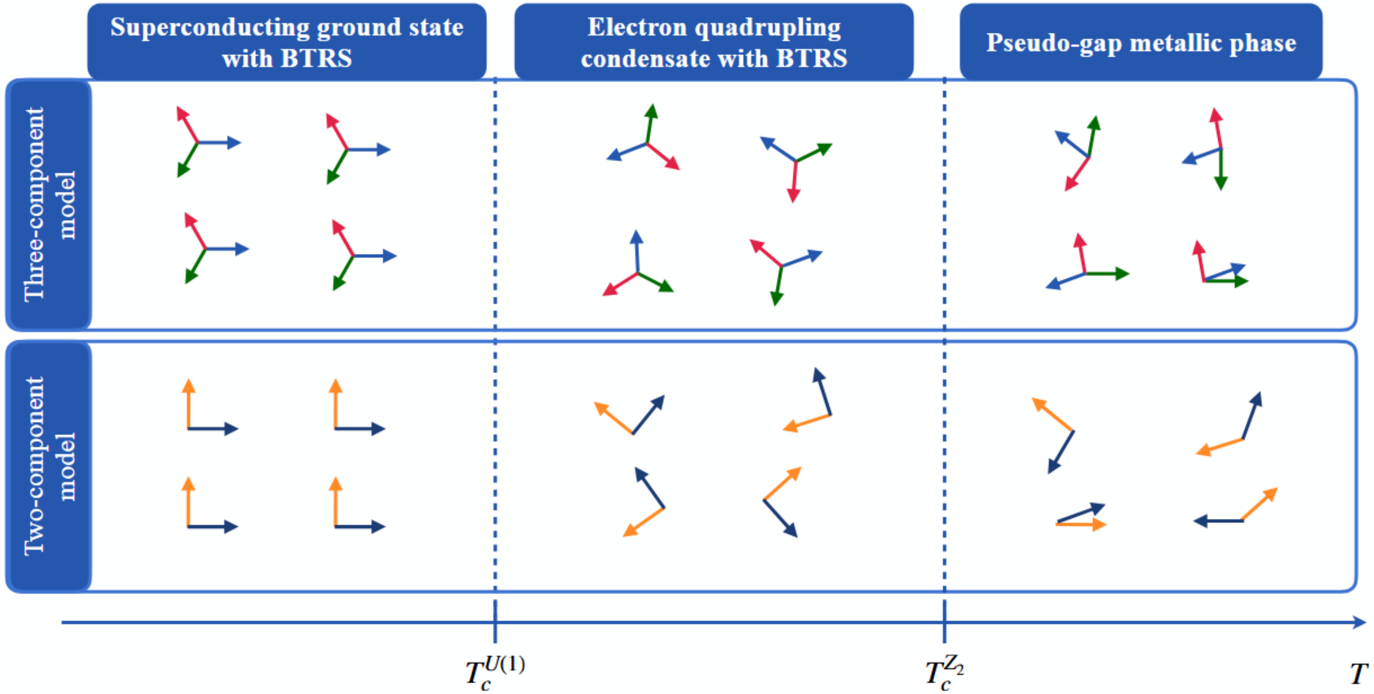
$< \Delta_i > = 0$

$< \Delta_i \Delta_j^* > \neq 0$

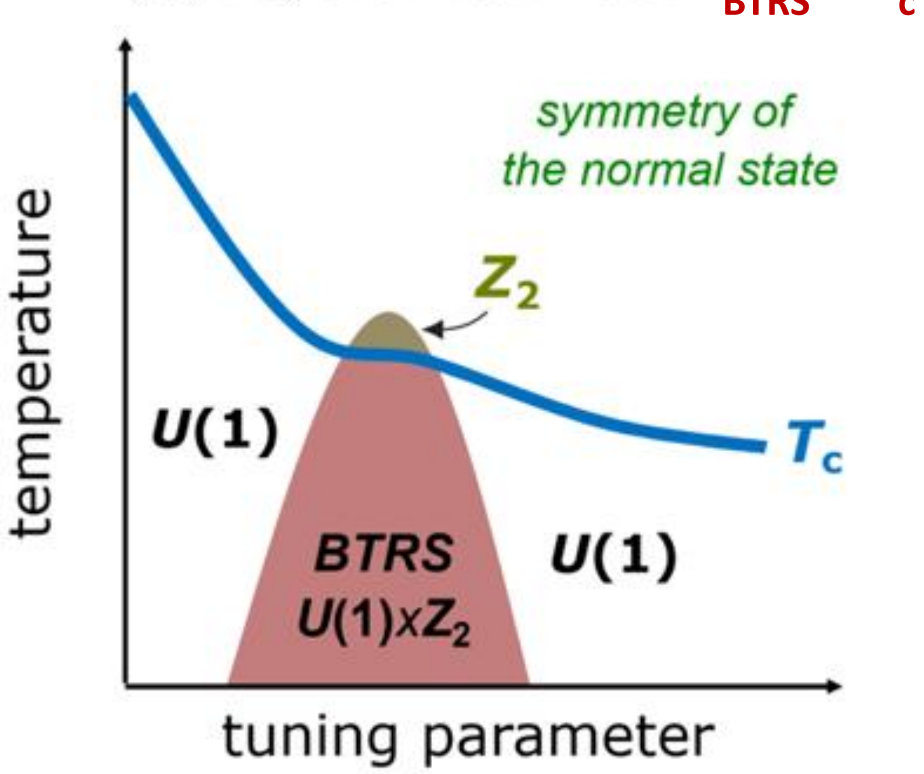
Pairing fluctuations

$< \Delta_i > = 0$

$< \Delta_i \Delta_j^* > = 0$

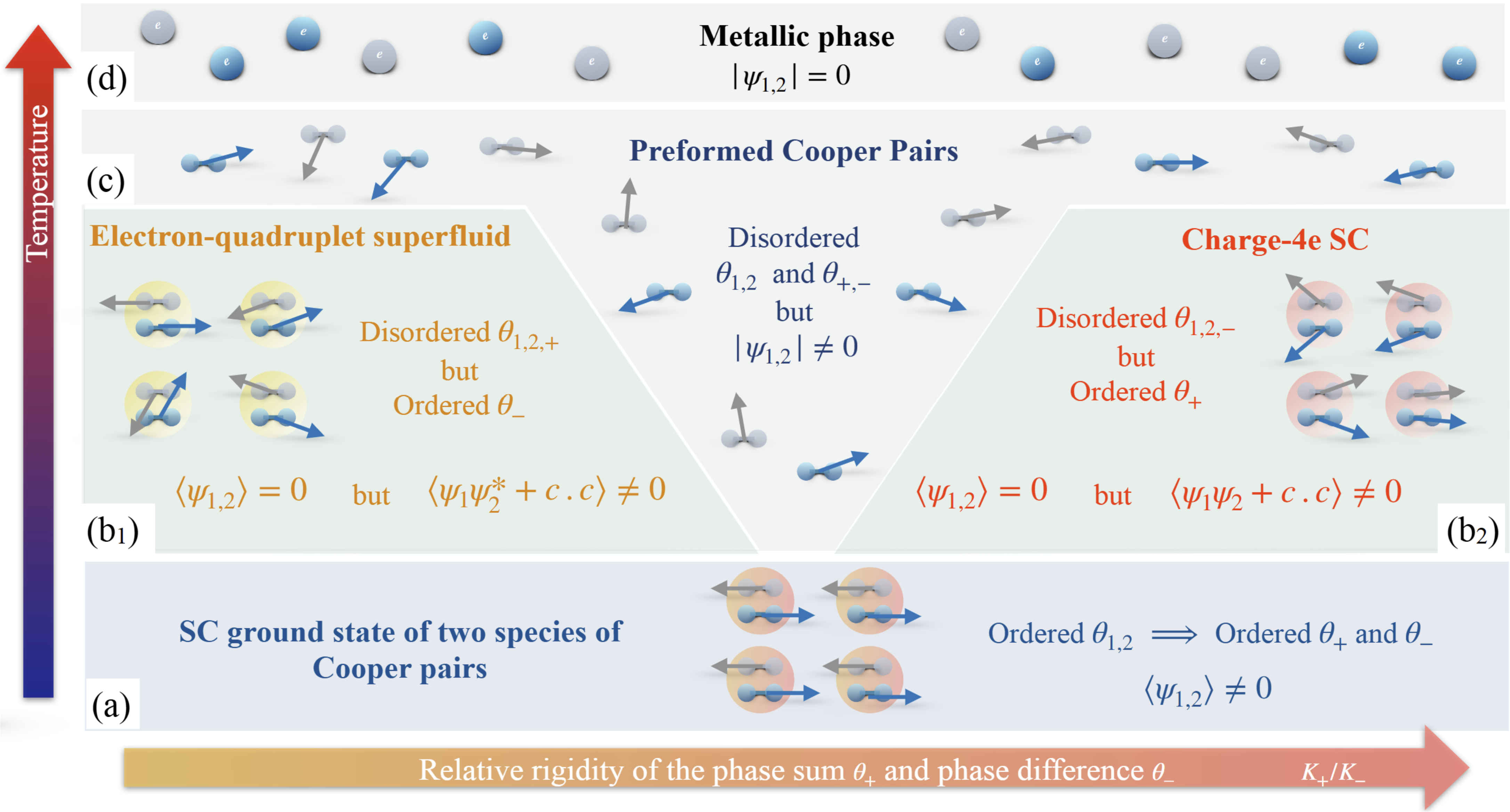


(b) Beyond Mean-field $T_{BTRS} > T_c$



V. Grinenko *et al.*, *Nat. Phys.* **17,** 1254–1259 (2021).

Types of fermion quadrupling in metals



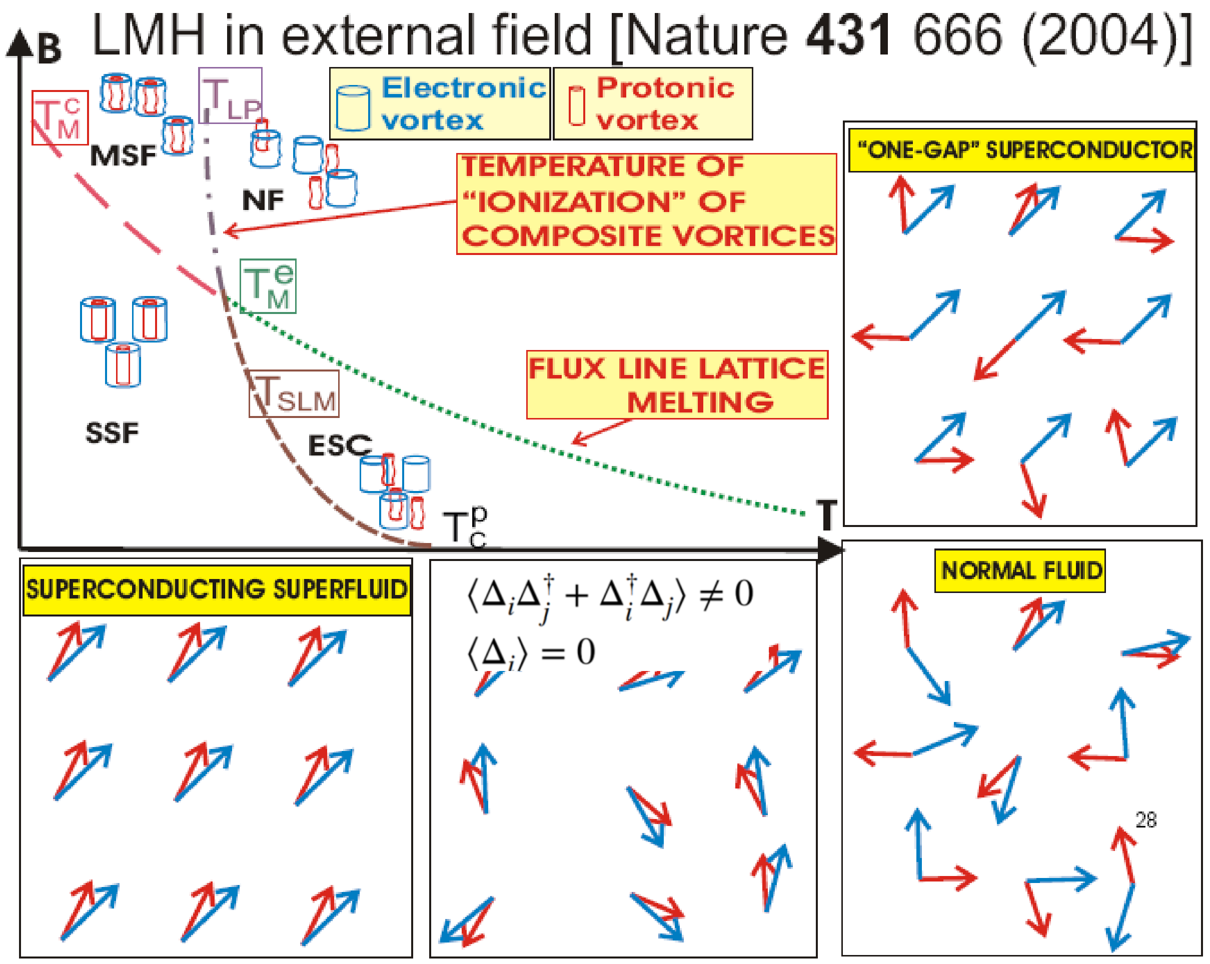
Predicted phase diagram of Liquid metallic hydrogen (LMH)

In an external magnetic field LMH may undergo phase transitions governed by vortex matter between phases with all types of “super” properties:

- Superconducting Superfluid
- Superfluid
- Superconductor

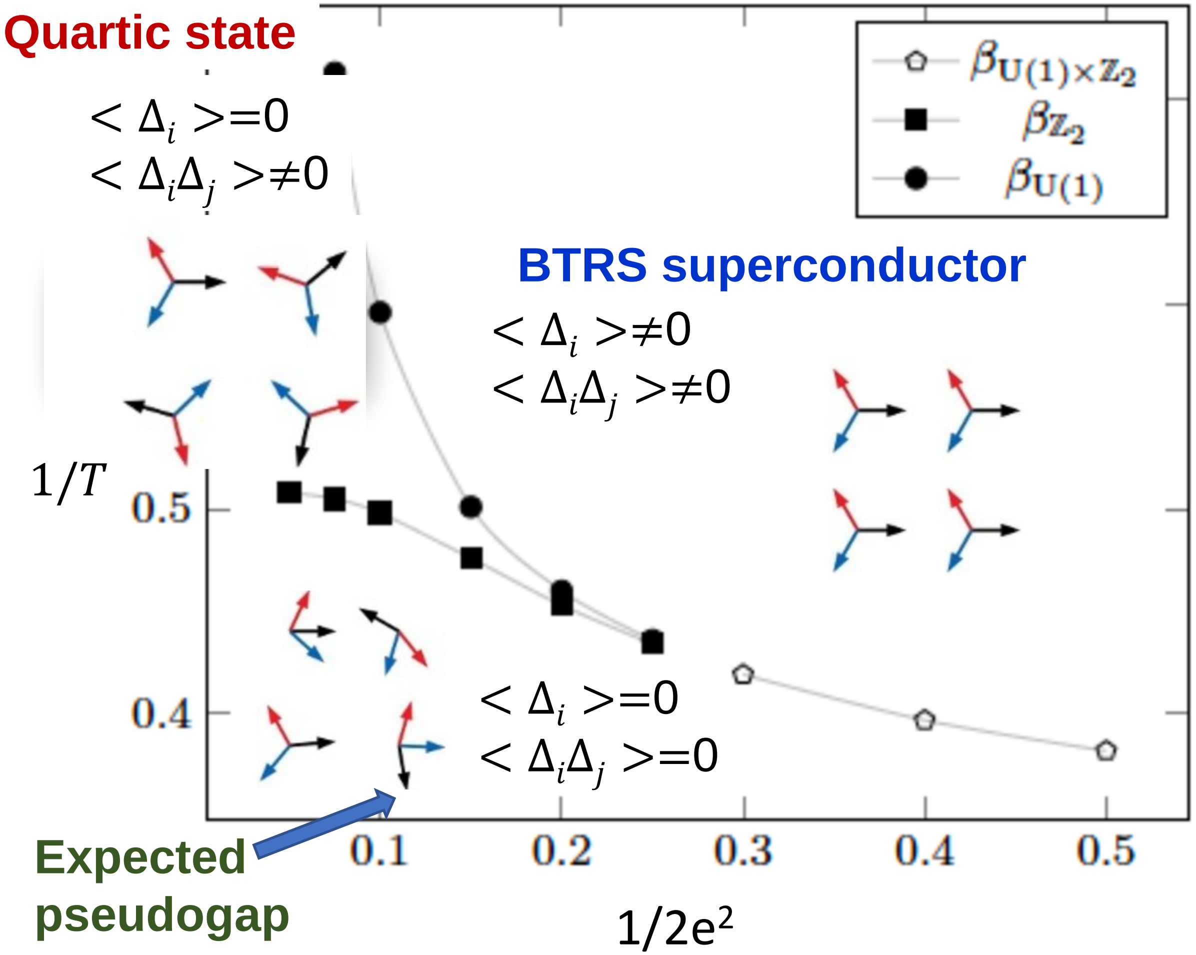
The system is characterized by 2-component order parameter with U(1)xU(1) symmetry

$$\Psi_\alpha = |\Psi_\alpha| e^{i\theta^{(\alpha)}} \quad (\alpha = 1, 2)$$

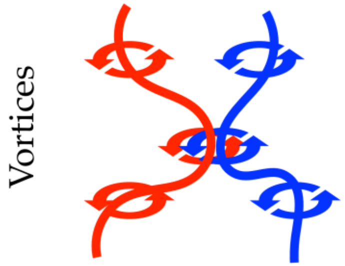


Egor Babaev, arXiv cond-mat/0201547 (2002) E.Babaev., *et al* Nature 431 666 (2004)

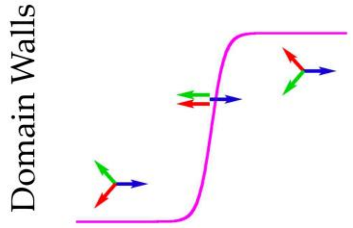
Fluctuations in *s+is* and *s+id* systems: quartic phase



A Bojesen, E Babaev, A Sudbø PRB 88 (22), 220511 (2013); PRB 89 (10), 104509 (2014)
I. Maccari and E. Babaev PRB 105, 214520 (2022)
Most detailed recent Monte-Carlo simulations: I. Maccari, E. Babaev PRB **105**, 214520 (2022)



U(1) symmetry restoration is driven by the proliferation of composite vortices.



Z_2 (BTRS) symmetry restoration is driven by the proliferation of unquantized vortices and domain walls;

Spontaneous fields are induced by current counterflow term

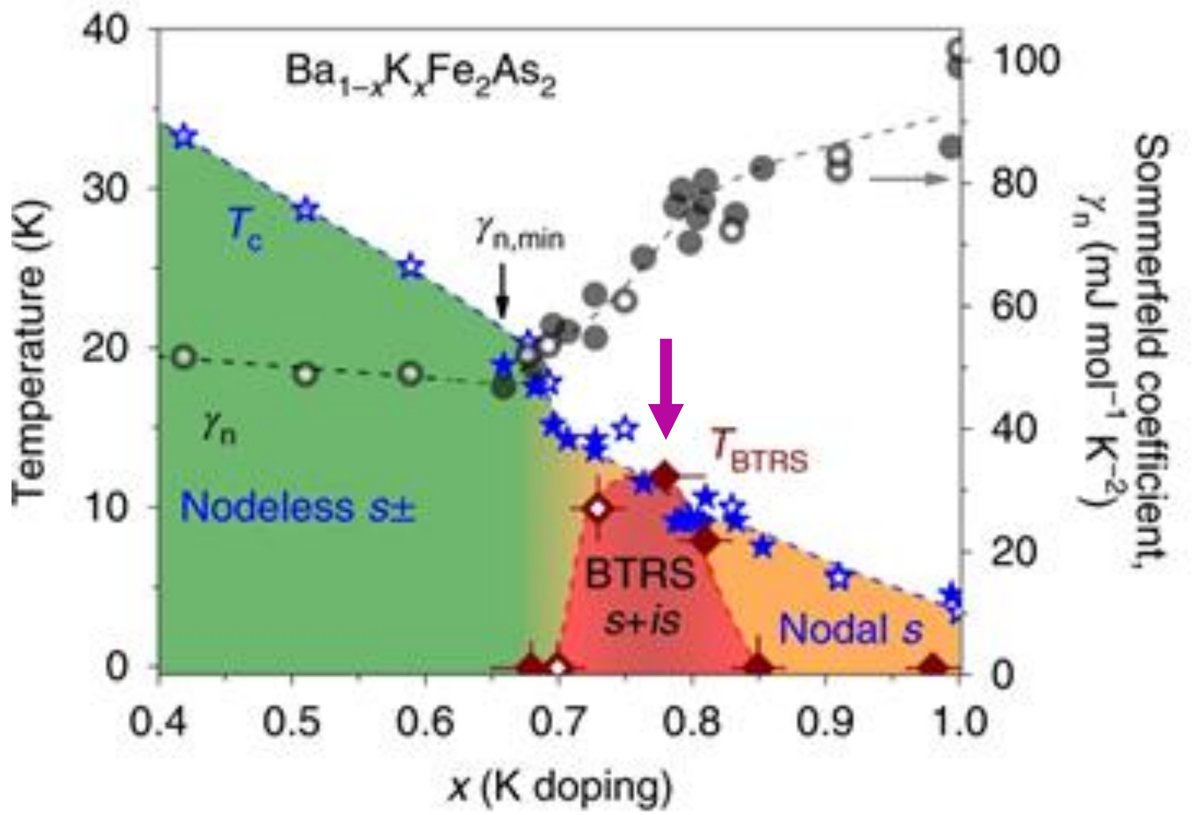
$$B_k = \partial_l A_m - \partial_m A_l = -\epsilon_{lm}\partial_l\left(\frac{J_m}{e|\Psi|^2}\right) - \frac{i\epsilon_{lm}}{e^2|\Psi|^4}\left[|\Psi|^2\partial_l\Psi^\dagger\partial_m\Psi + \Psi^\dagger\partial_l\Psi\partial_m\Psi^\dagger\Psi\right]$$

$\Psi^\dagger = (\psi_1^*, \psi_2^*, \psi_3^*)$

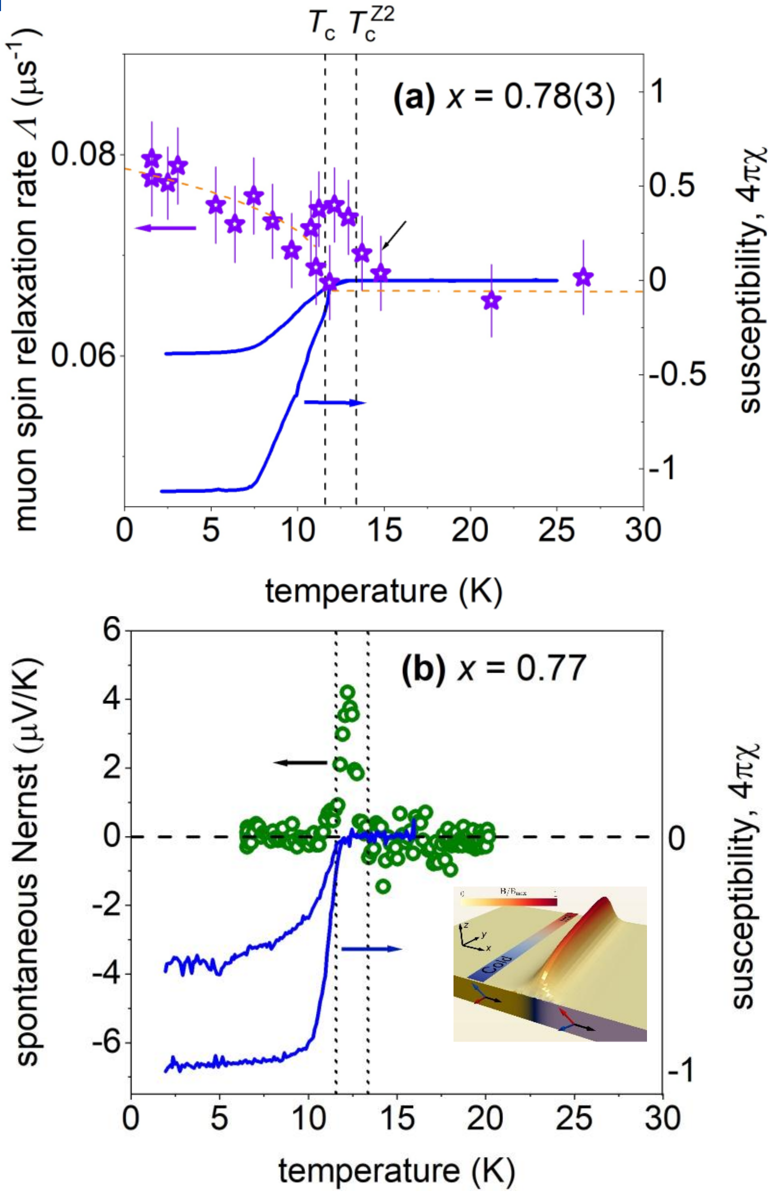
In the quartic phase the current counterflow term is non-zero!

Are there BTRS and pseudogap state above T_c in $Ba_{1-x}K_xFe_2As_2$?

Evidence of BTRS state above T_c



V. Grinenko et al., *Nat. Phys.* 16, 789–794 (2020).



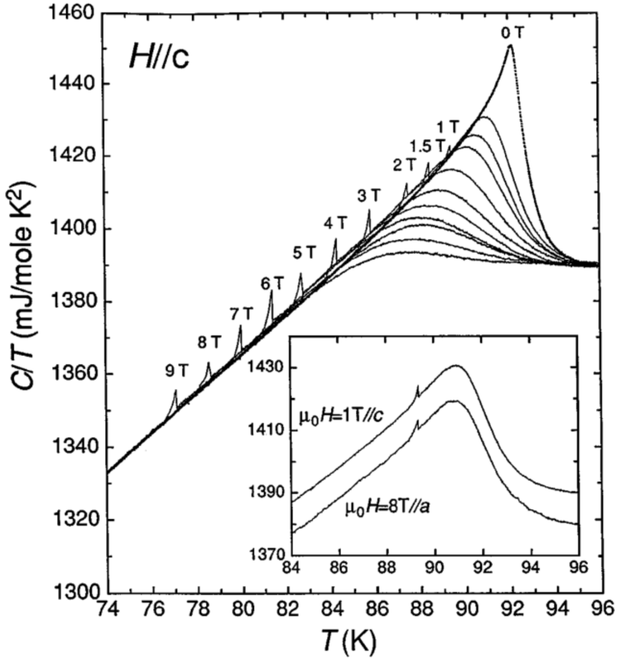
Evidence for the Z_2 transition above T_c from μSR, note also spontaneous Nernst effect.

Is there a thermodynamic evidence for the phase transition above T_c?

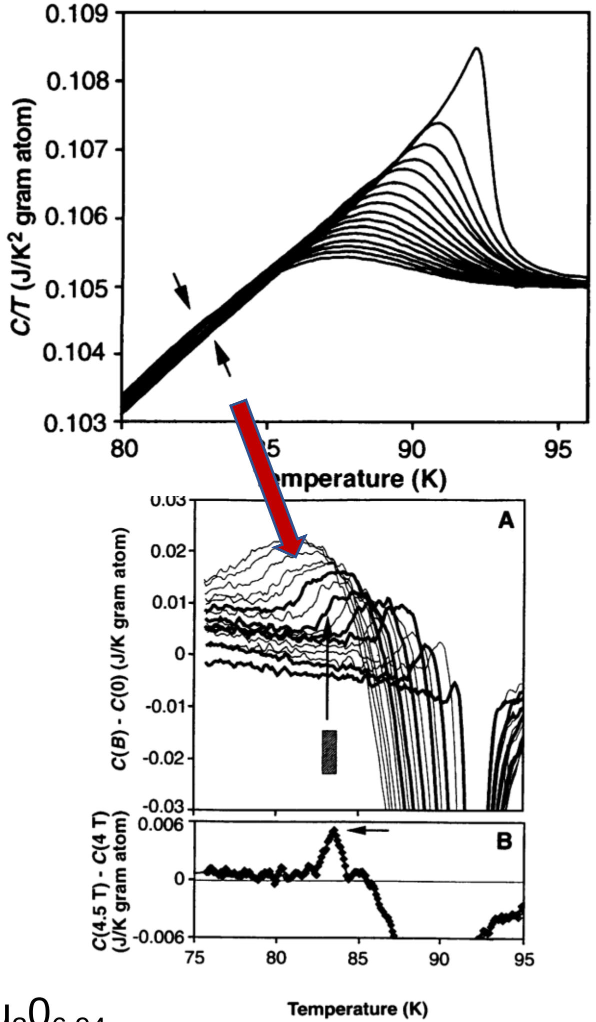
V. Grinenko *et al.*, *Nat. Phys.* **17,** 1254–1259 (2021)

What is the expected size of an anomaly in the specific heat at the BTRS phase transition?

Vortex melting transition in HTS



Total specific heat of an untwined $YBa_2Cu_3O_{7-d}$ single crystal for $H||c$.
A. Schilling et al., PRL 78, 4833 (1997).

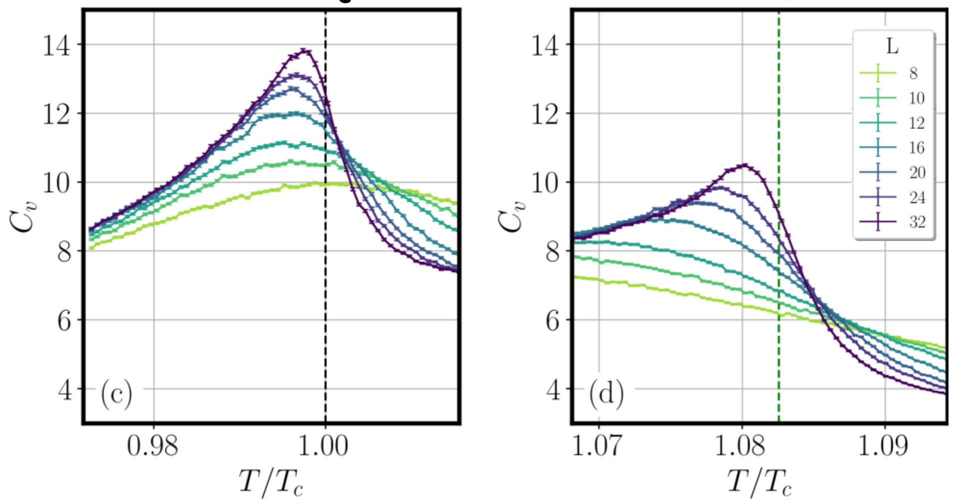


Total specific heat C/T of $YBa_2Cu_30_{6.94}$ as a function of T, *Science* **273** (5279), 1210-1212 (1996).

Ilaria Maccari, *ETH*

Monte-Carlo (MC) simulations of two component BTRS model with phase fluctuations.

Vortex melting, T_c Domain walls proliferation, T_c^{Z2}

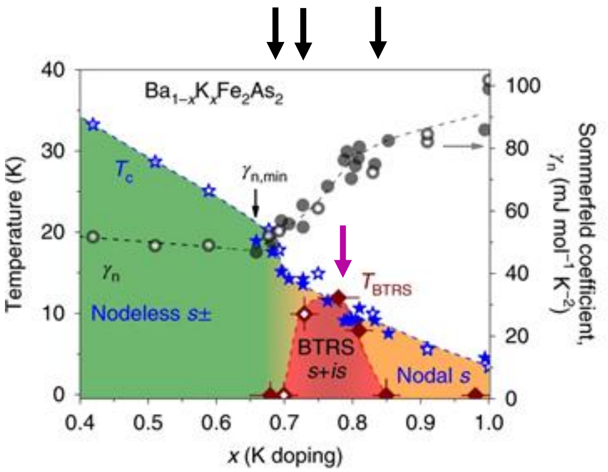


I. Shipulin *et al.*, *Nat Commun* **14**, 6734 (2023).

The simulations predicted two different anomalies in the specific heat.

Is there a phase transition above T_c?

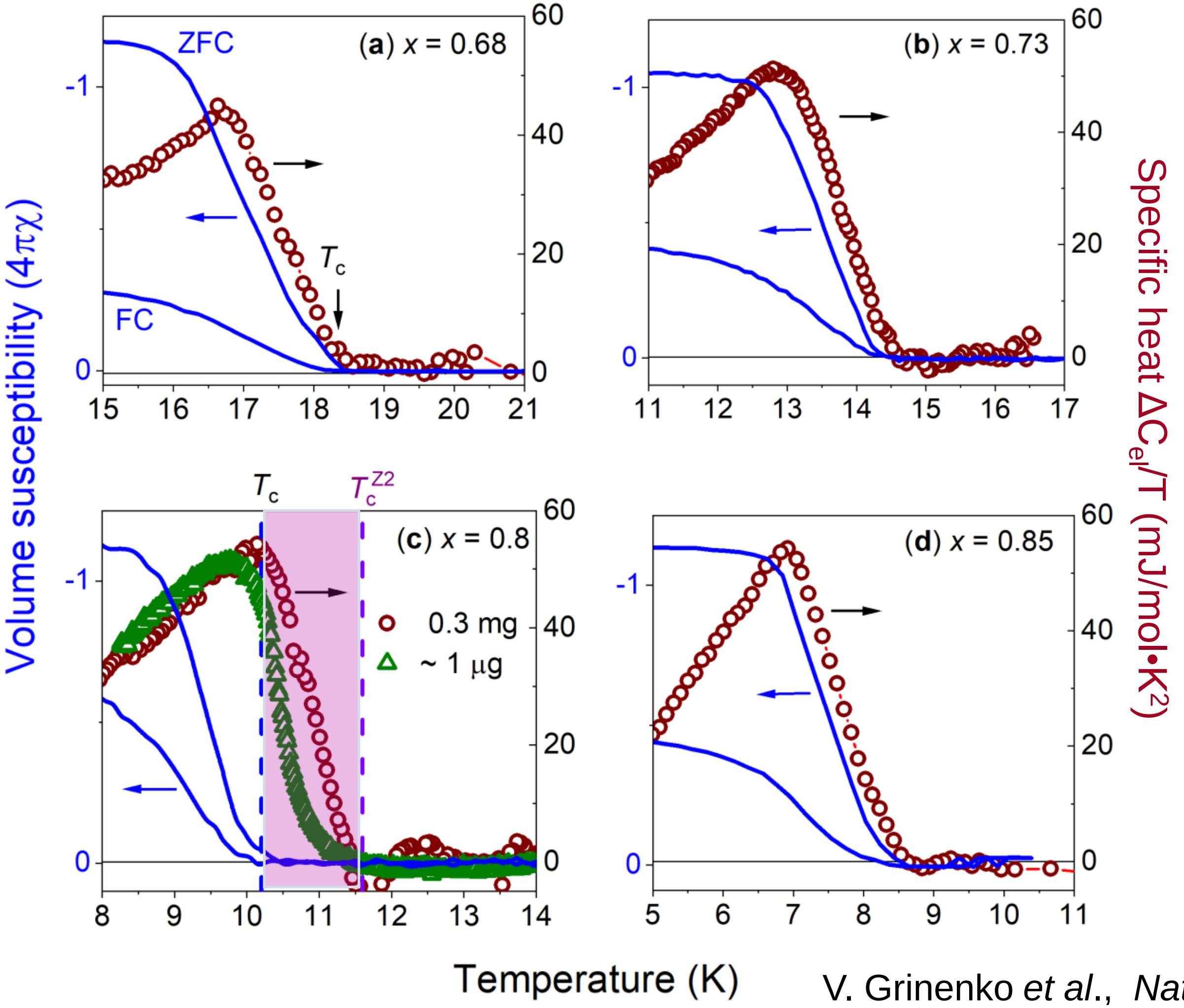
Tsung-Dao Lee Institute

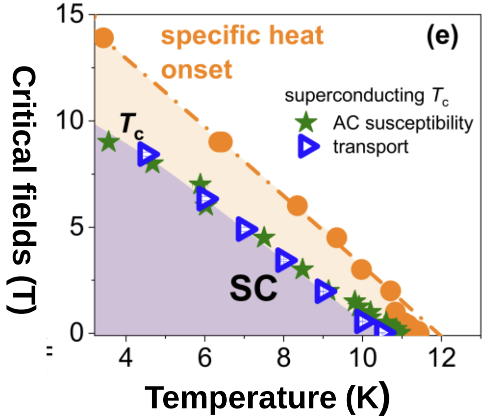


V. Grinenko et al., *Nat. Phys.* 16, 789–794 (2020).

Is there specific heat anomaly at T_{BTRS} ?

What is the nature of the transition above T_c?





V. Grinenko *et al*., *Nat. Phys.* **17,** 1254–1259 (2021).

The splitting increases in applied magnetic field.

The splitting between transitions seen by the specific heat and susceptibility is observed for the samples **with the "*strongest*" BTRS phase** found in the μSR experiments.

Calorimetric evidence for phase transition above T_c in zero magnetic field

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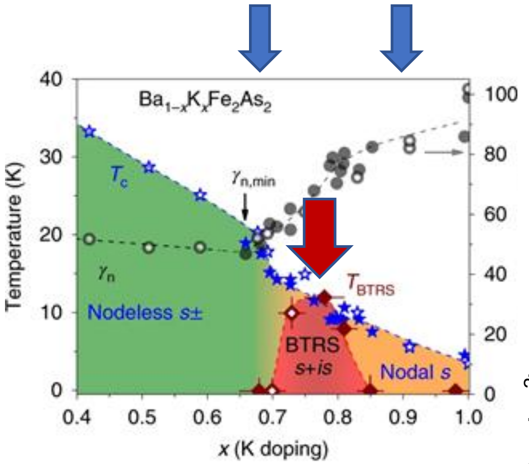
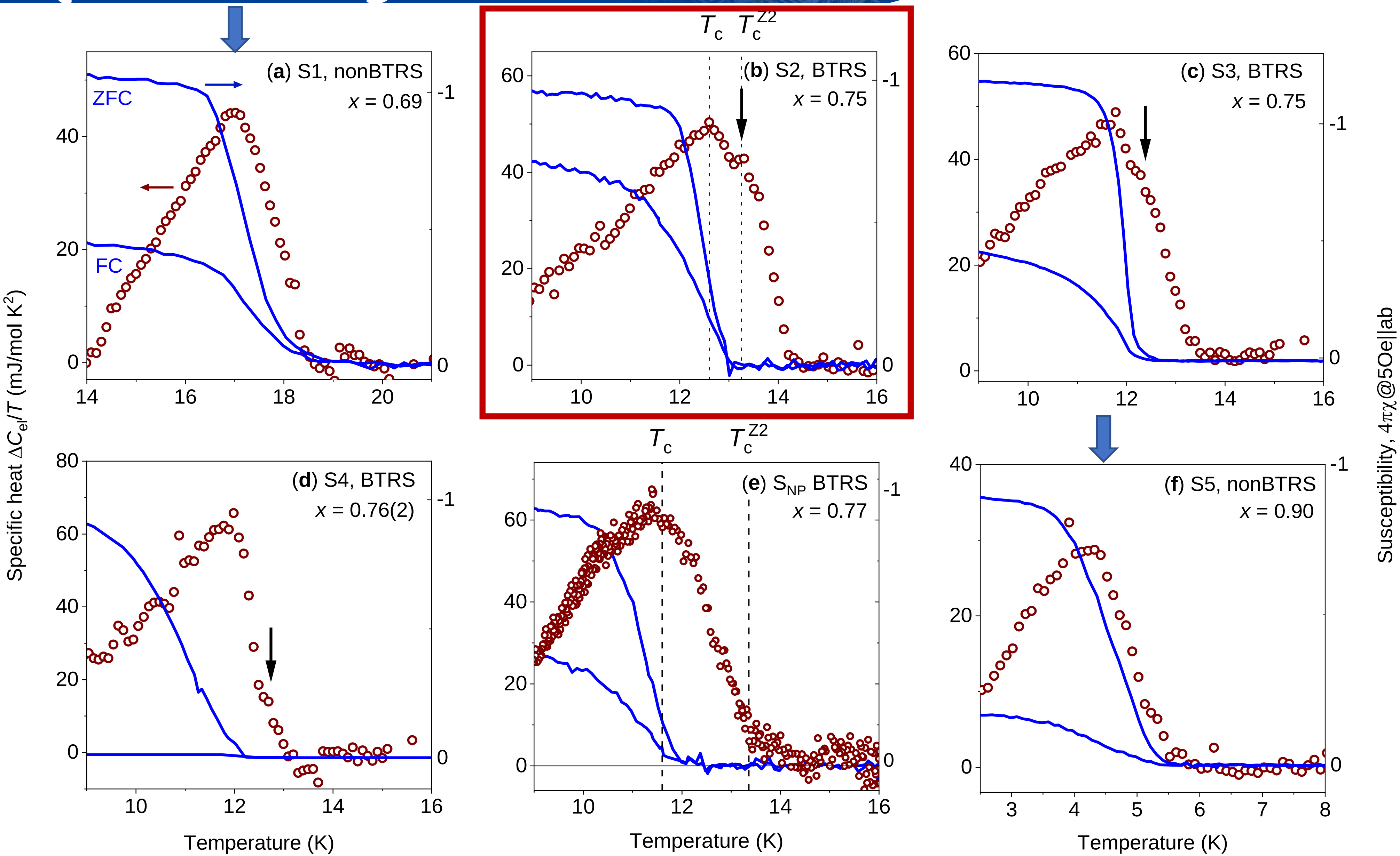


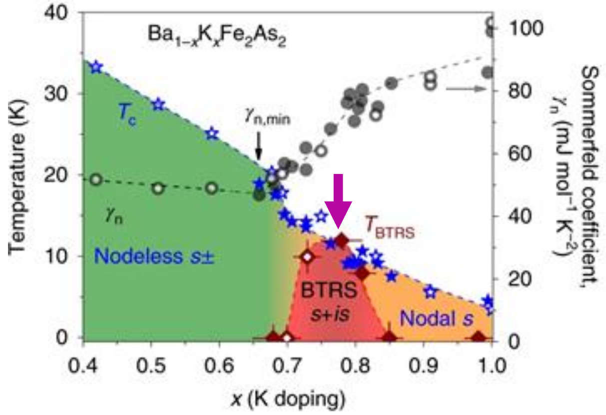
I. Shipulin *et al.*, *Nat Commun* **14**, 6734 (2023).

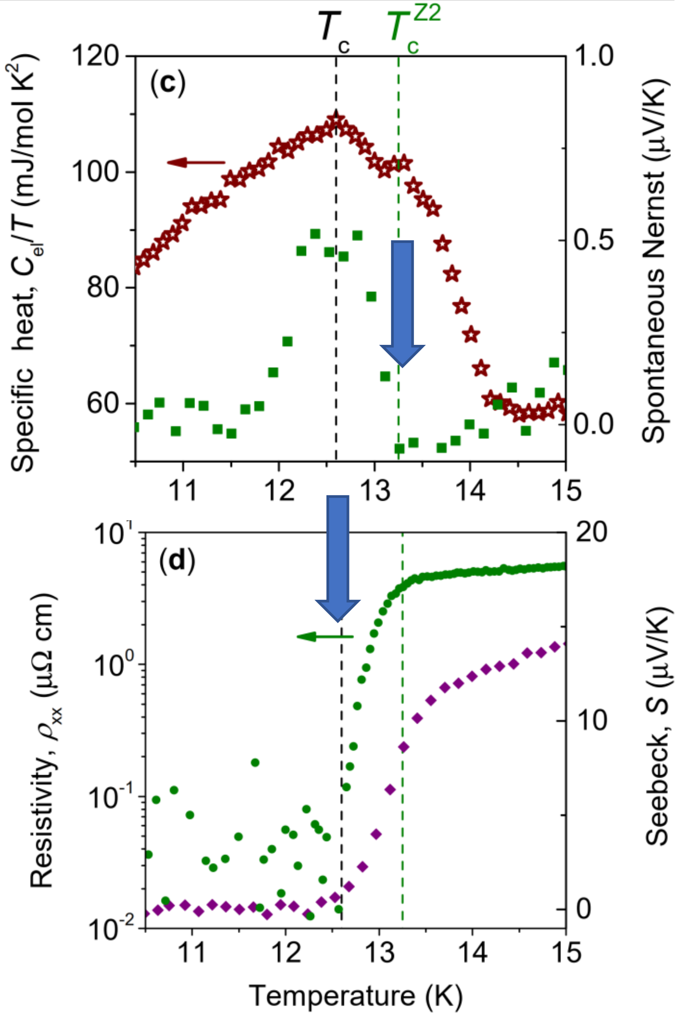
➢**The shoulder-like anomaly above T_c in a narrow doping range around x ~ 0.75.**

Can the double anomaly interpreted as evidence two phase transitions?



Calorimetric evidence for BTRS phase transition above T_c in zero magnetic field



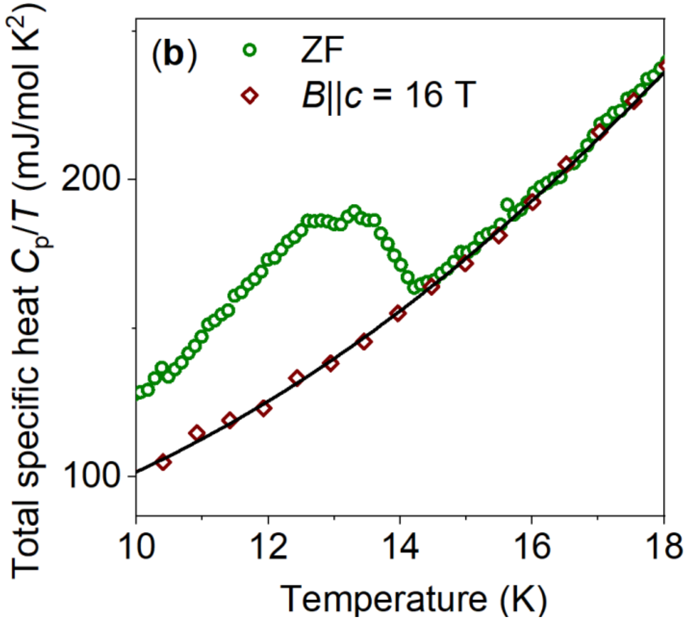


- ✓ **The spontaneous Nernst signal corelates with upper transition.**
- ✓ **Zero resistivity corresponds to lower transition in the specific heat.**

Ilya Shipulin, *IFW Dresden, Germany*

I. Shipulin *et al*., *Nat Commun* **14**, 6734 (2023).

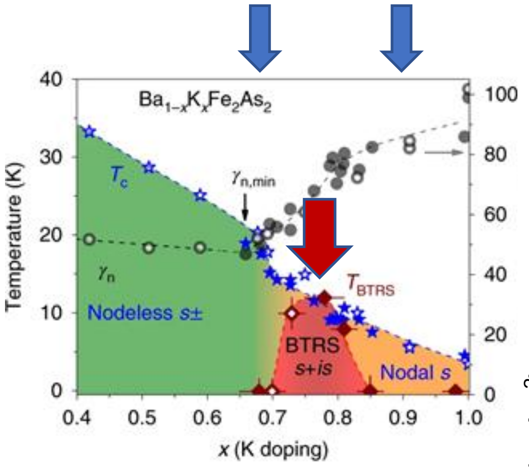
A doble anomaly is seen in the raw data but the scattering of the data is large.



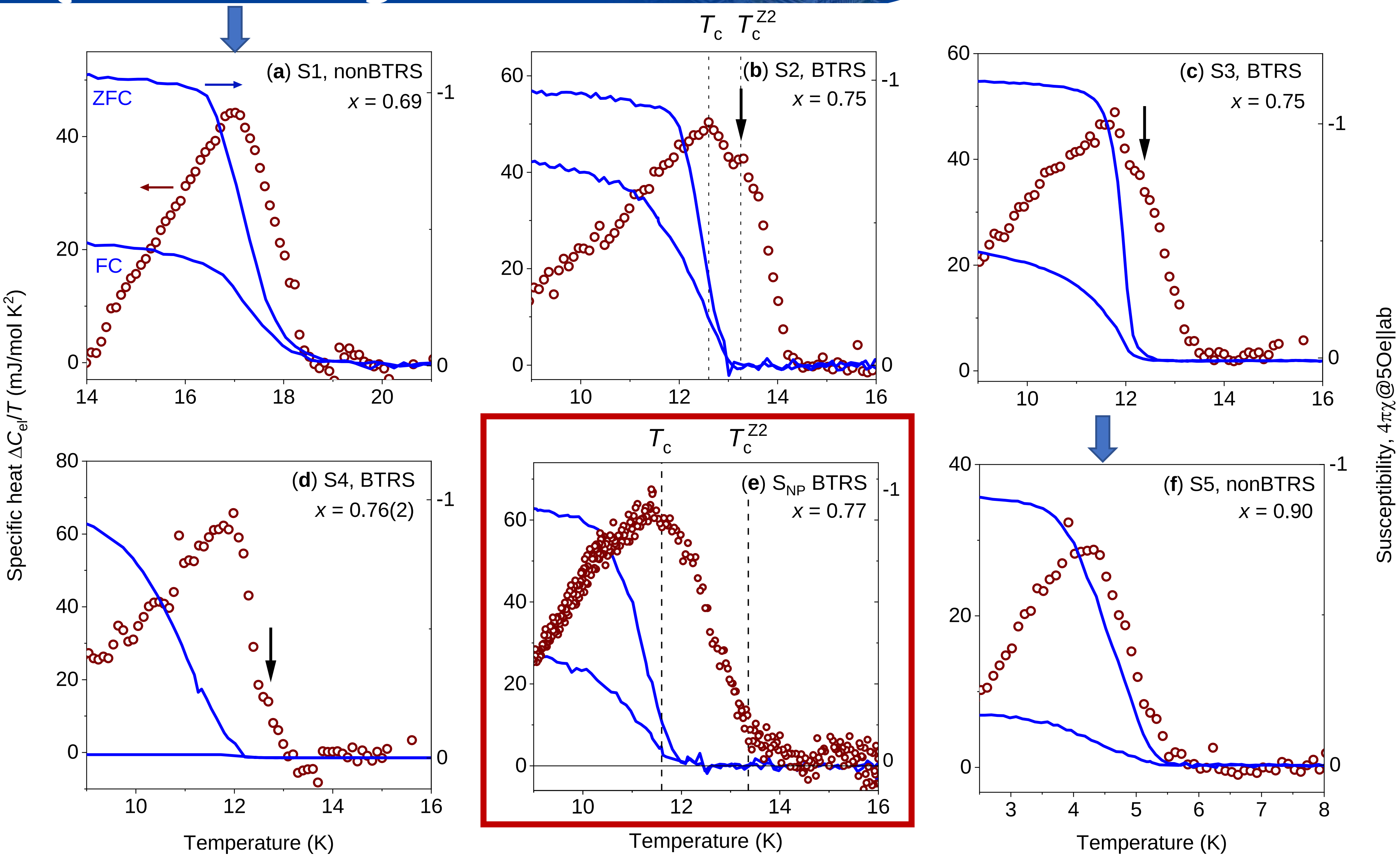
Can we get the data with a higher resolution?

Calorimetric evidence for phase transition above T_c in zero magnetic field

李政道研究所 TSUNG-DAO LEE INSTITUTE



I. Shipulin *et al.*, *Nat Commun* **14**, 6734 (2023).



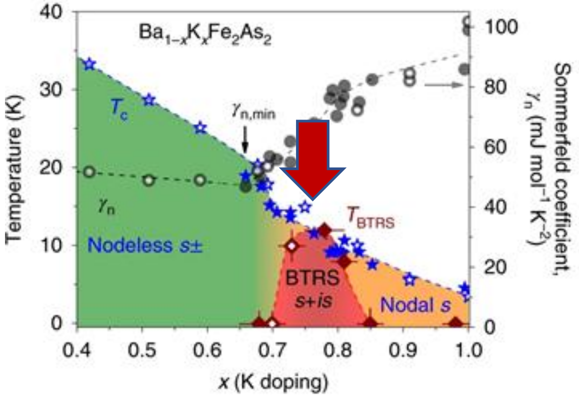
➢ **The shoulder-like anomaly above T_c in a narrow doping range around x ~ 0.75.**

Can the double anomaly be interpreted as evidence of two phase transitions?

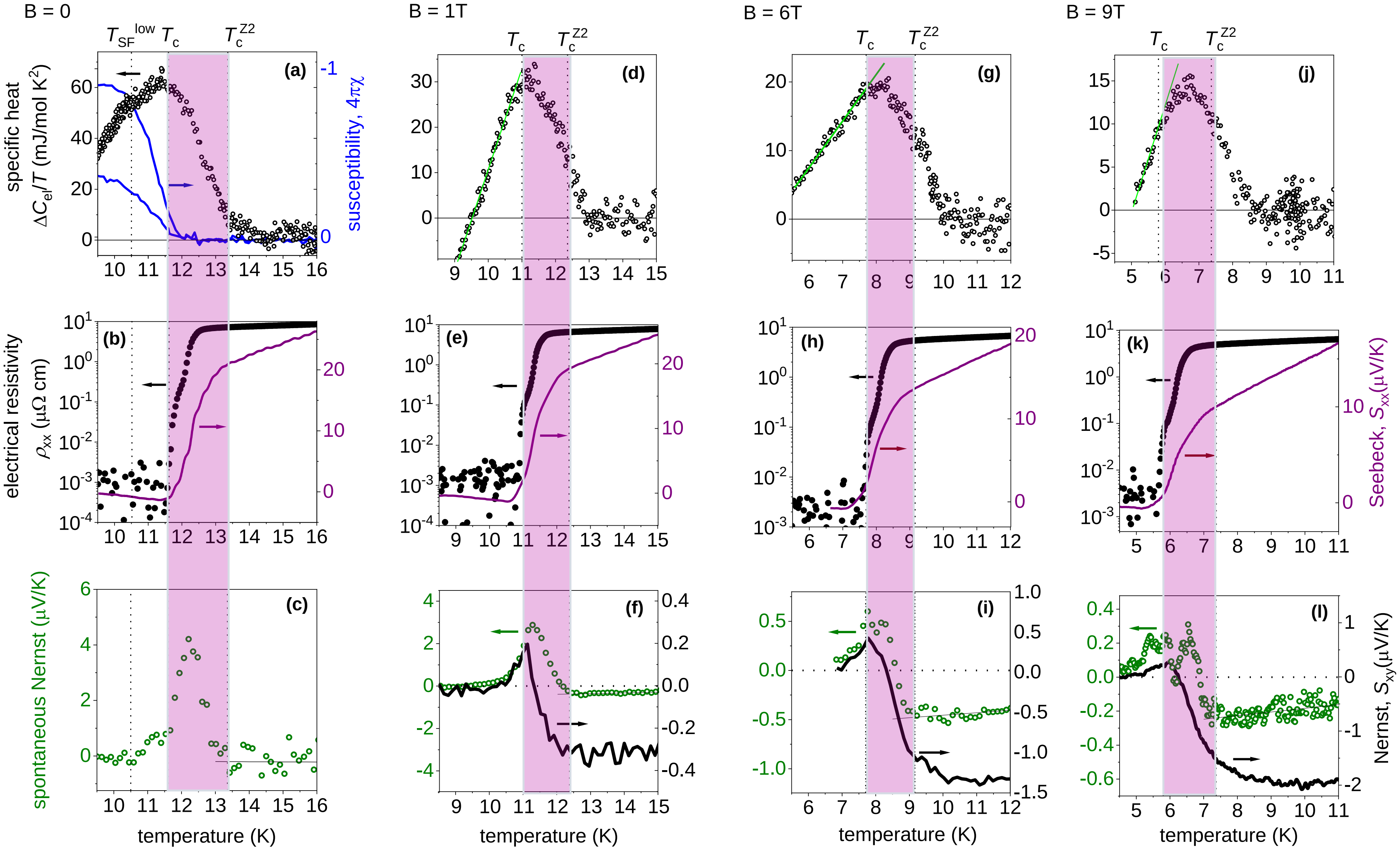
Calorimetric evidence for phase transition above T_c

Federico Caglieris
CNR-SPIN Genova

Tino Gottschall
HZDR, Dresden-Rossendorf

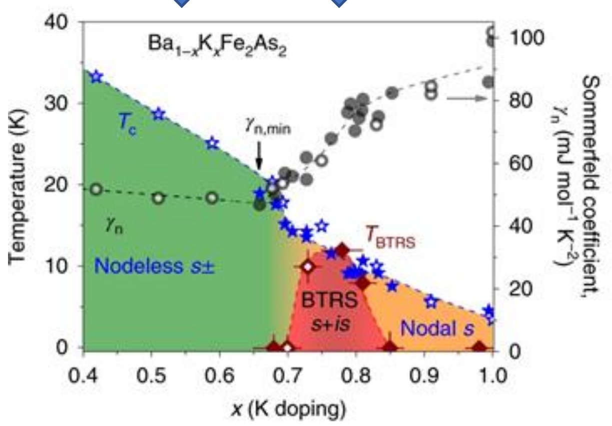


V. Grinenko et al., *Nat. Phys.* 16, 789–794 (2020).



The small size of anomalies at T_c and T_c^{Z2} are consistent with theory predictions.

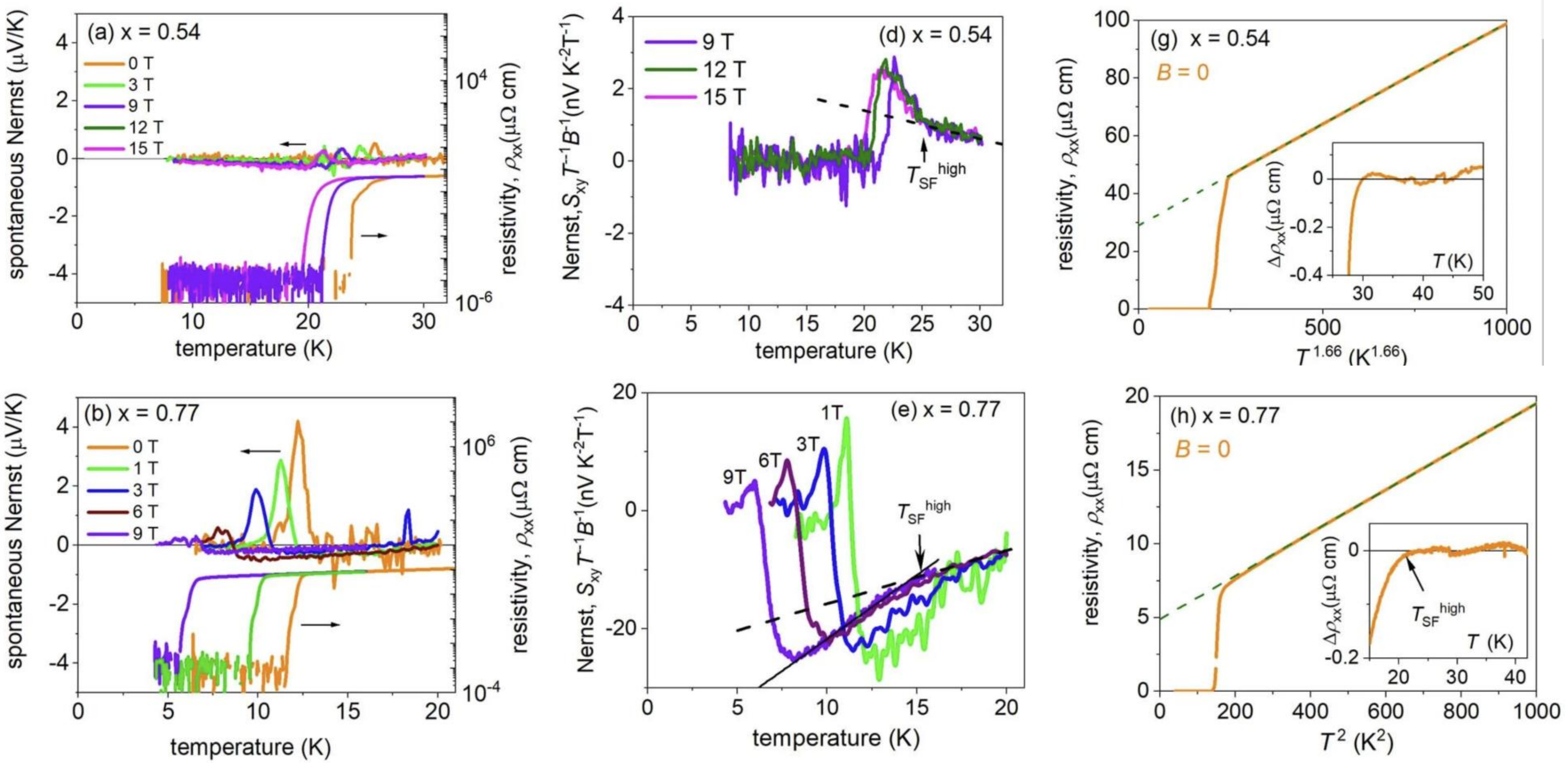
Spontaneous Nernst and superconducting fluctuations.



Federico Caglieris, *CNR-SPIN Genova*

Superconducting fluctuations (preformed pairs) in the samples with BTRS state at $T^* \sim 2T_c$

Spontaneous (even in the field) Nernst | Odd in the field Nernst | Electrical resistivity



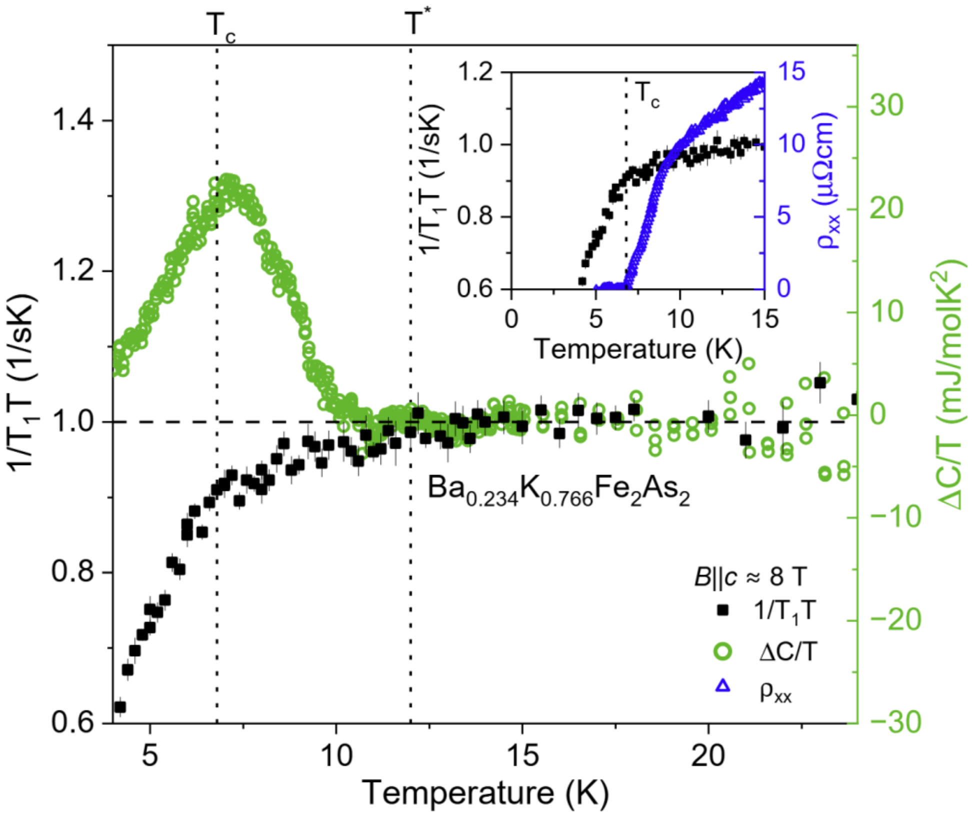
V. Grinenko *et al.*, *Nat. Phys.* **17,** 1254–1259 (2021).

- ✓ In BKFA without BTRS state spontaneous Nernst signal is absent and superconducting fluctuations are weak.

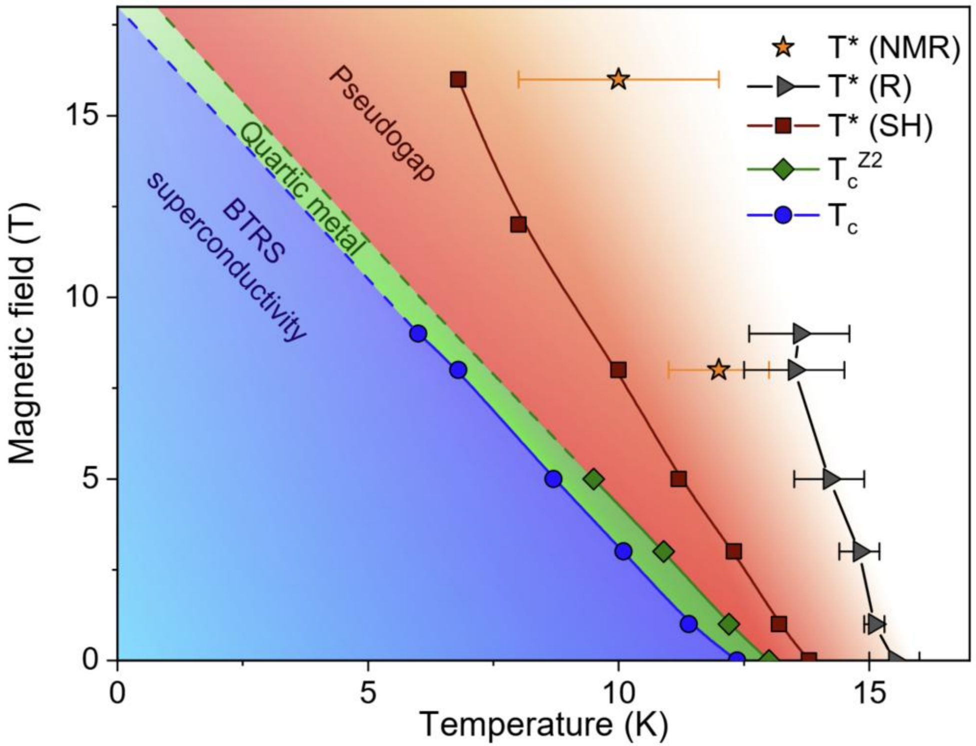
NMR and μSR evidence for pseudogap behavior in $Ba_{1-x}K_xFe_2As_2$ with a quartic state: F. Bärtl et al., arXiv:2501.11936 (2025)

TSUNG-DAO LEE INSTITUTE

NMR spin-lattice relaxation rate: $x = 0.77$



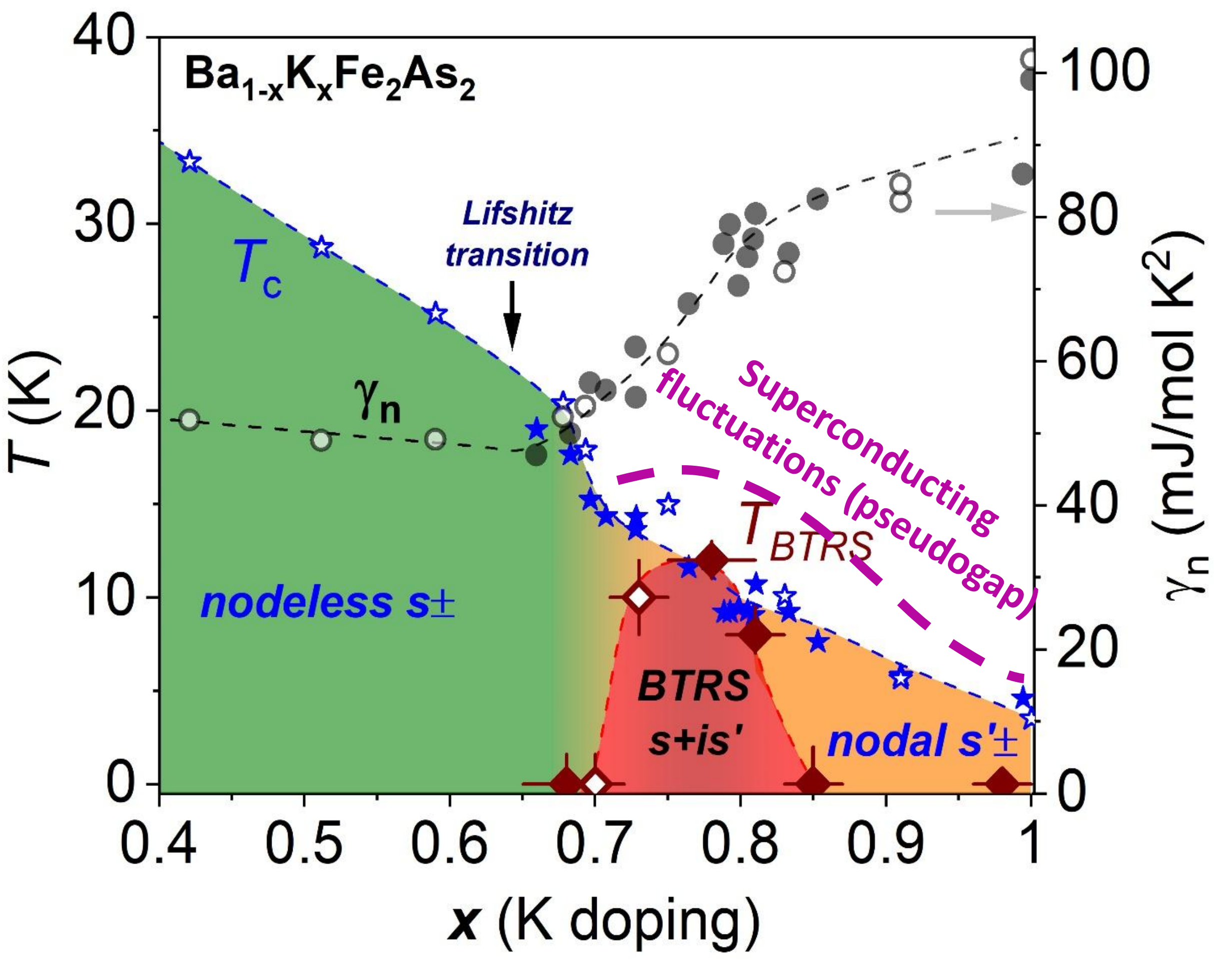
Phase diagram: $x = 0.77$

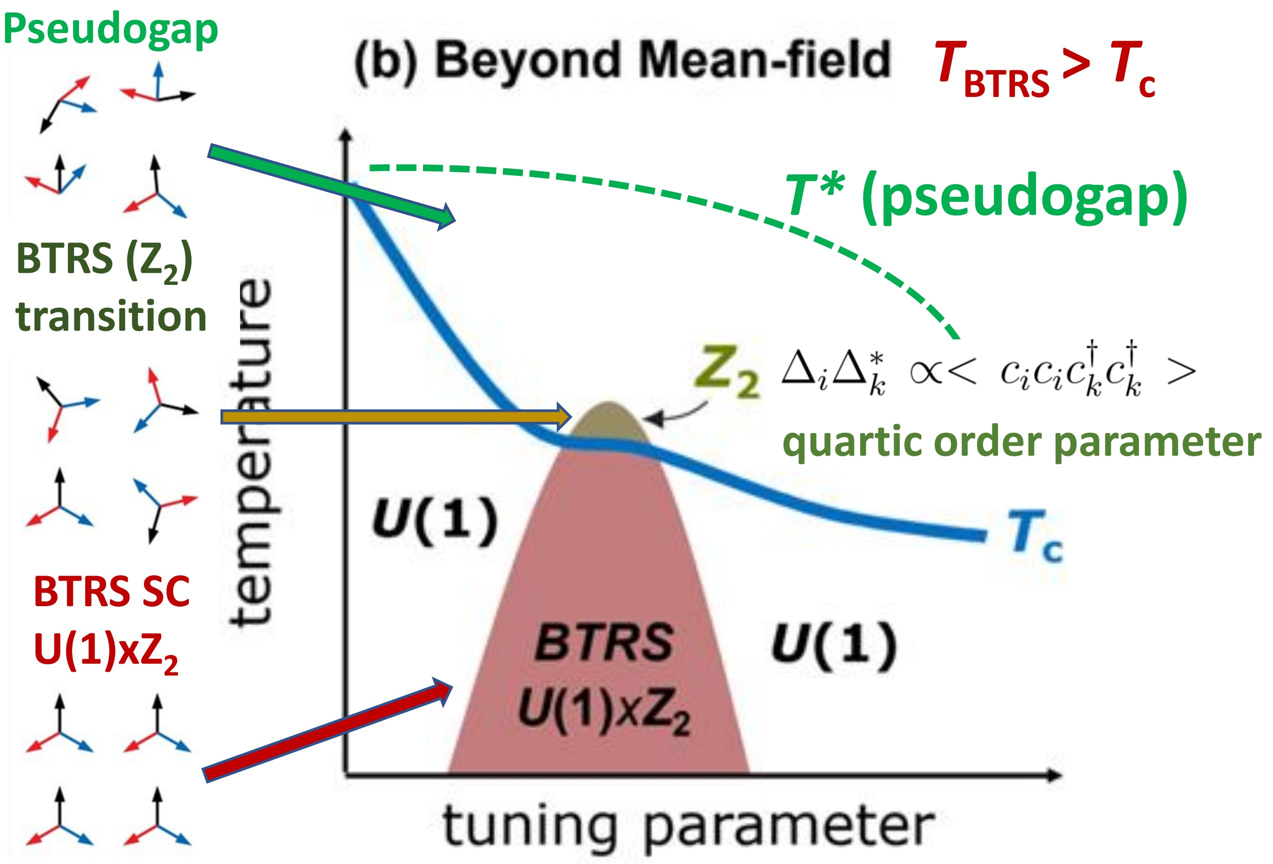



Hannes Kühne, **Florian Bärtl**
HZDR, Dresden-Rossendorf, Germany

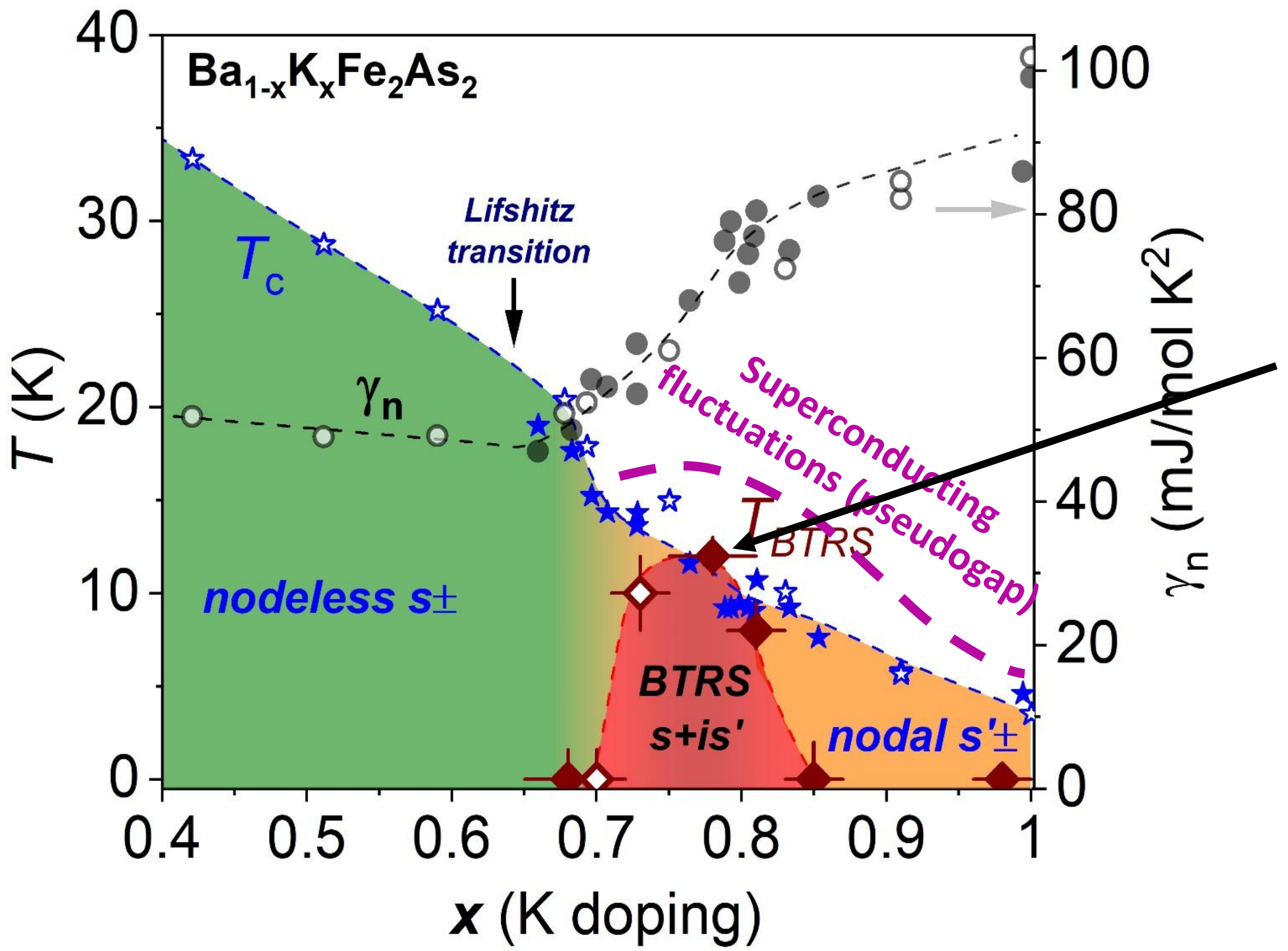
- $1/T_1T \propto \sum_q \chi''(q,\omega_0)$ (spin-lattice relaxation rate) is flat with temperature in the normal state no evidence for proximity to magnetism;
- $1/T_1T$ reduces below T^* -> pseudo gap like behavior;
- Close to T^* we observed onset of the anomaly in the specific heat → consistent with preformed Cooper pairs scenario.

Superconducting fluctuations and pseudogap



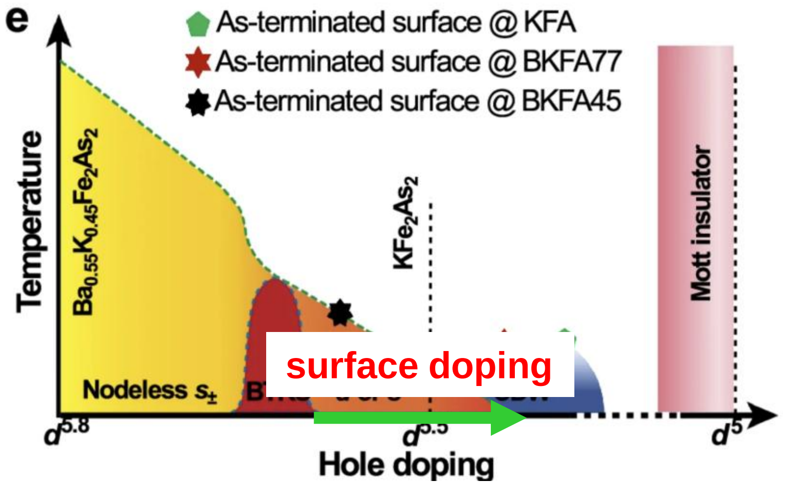


Evidence for domain walls above T_c

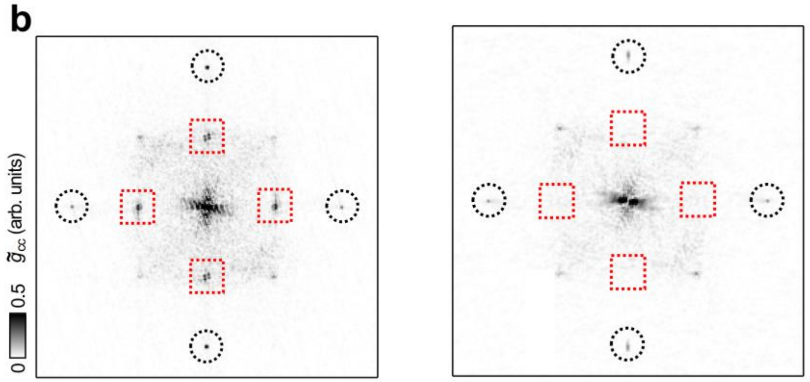


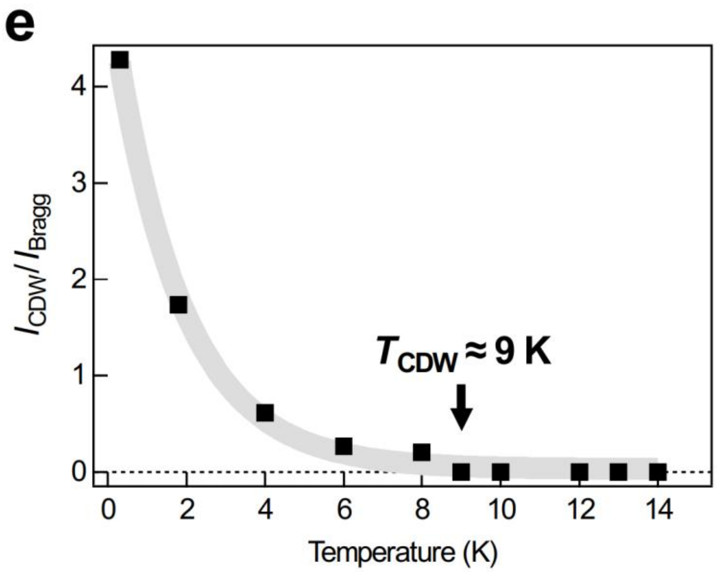
Domain walls?

Coupling of superconducting domain walls to charge density waves in the BTRS state

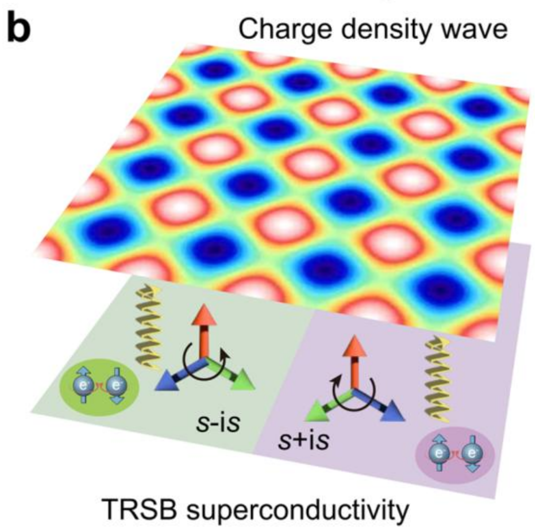


Surface CDW phase below ~ 9 K





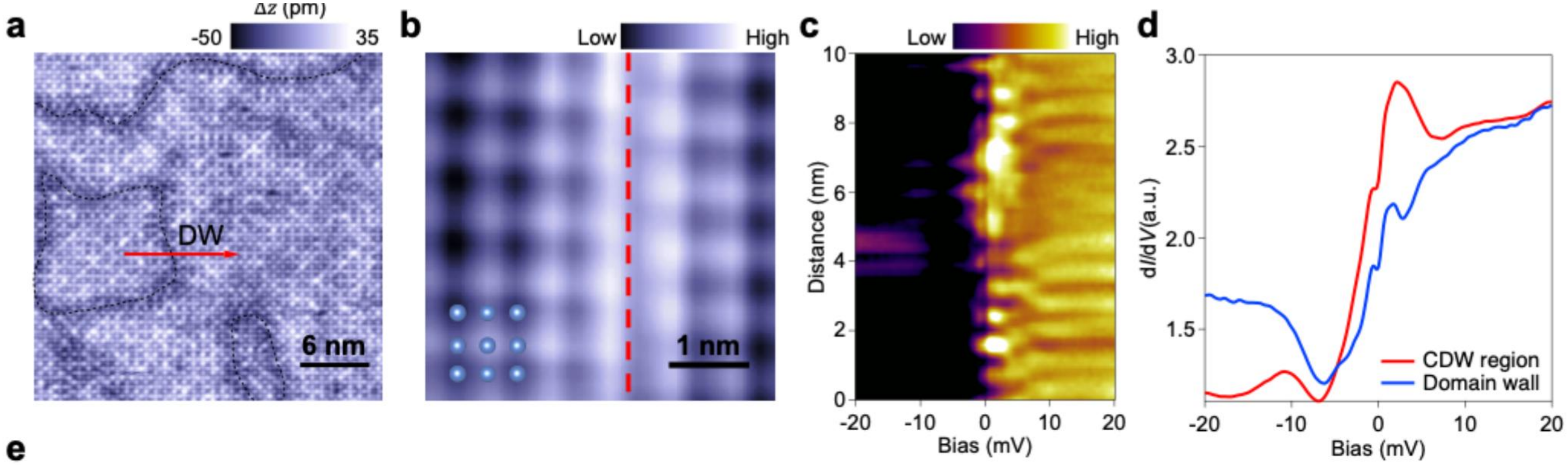
Q. Hu et al., Nat Commun 16, 253 (2025)



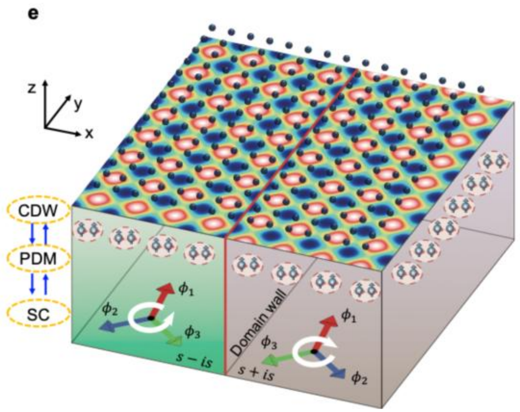
- The temperature dependence of the surface CDW order parameter is unusual.
- Is there a coupling between surface CDW and superconductivity and BTRS ?

Coupling of domain walls to charge density waves

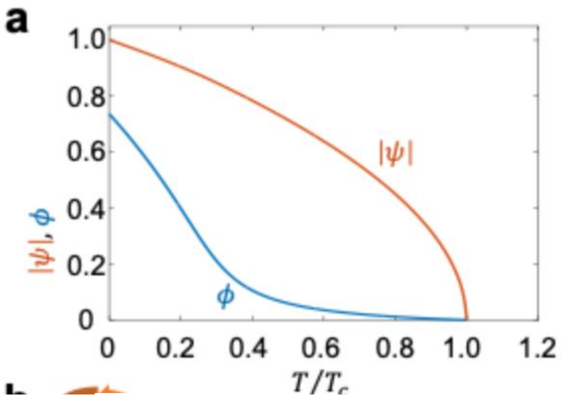
Experimental signature for domain walls (DW)



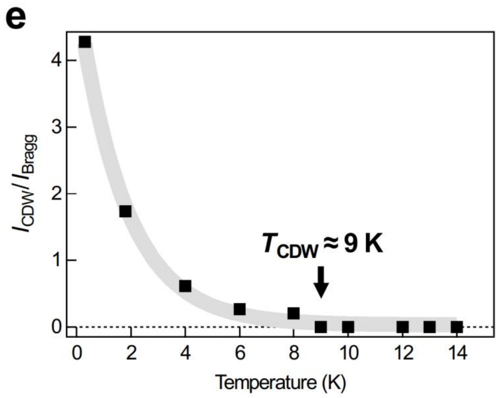
Coupling of the CDW order parameter with superconductivity results in $\pi/2$ - CDW domain walls.



Theory



Experiment

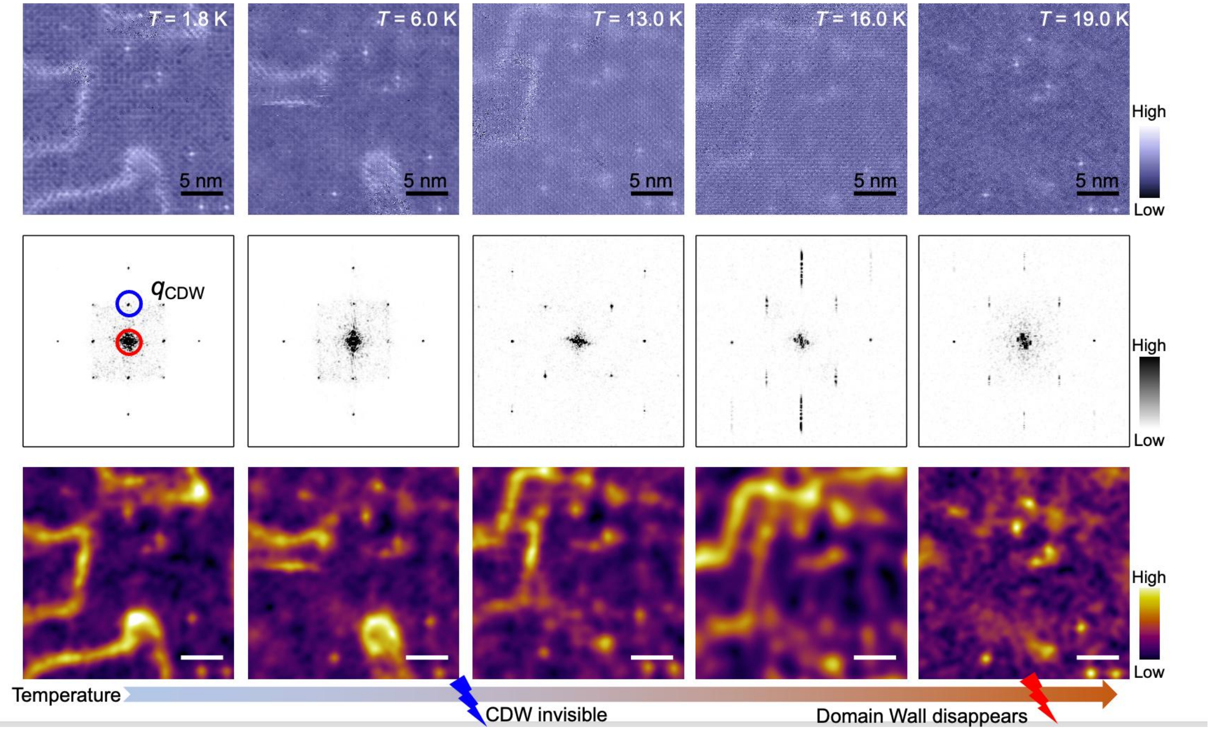


The CDW domain walls are an image of superconducting domain walls!

Quanxin Hu et al., arXiv:2512.07272

Properties of domain walls

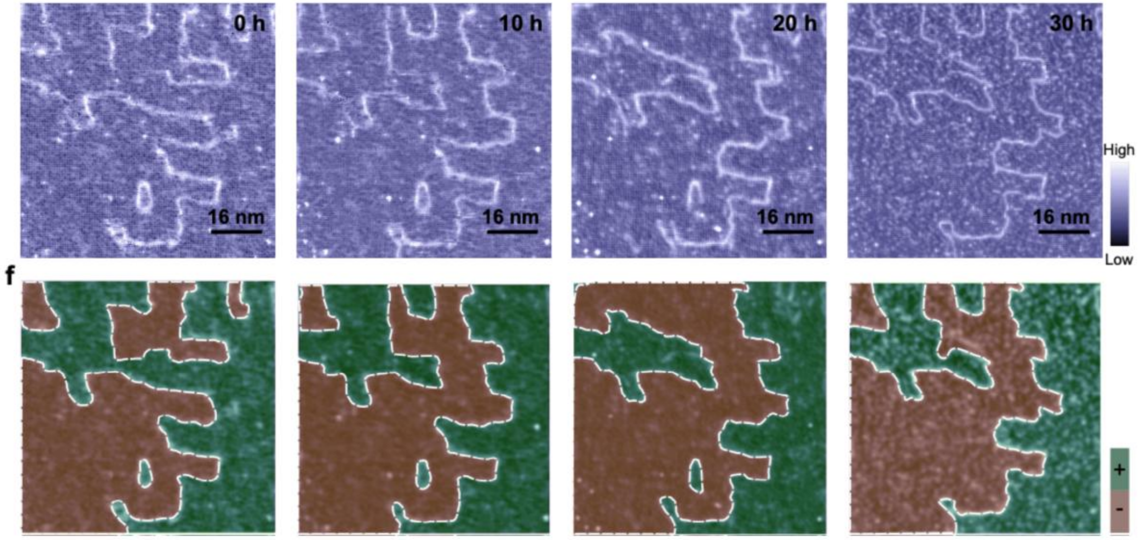
On the nanoscale domain walls exist up to $T^* > T_c \sim 11$ K



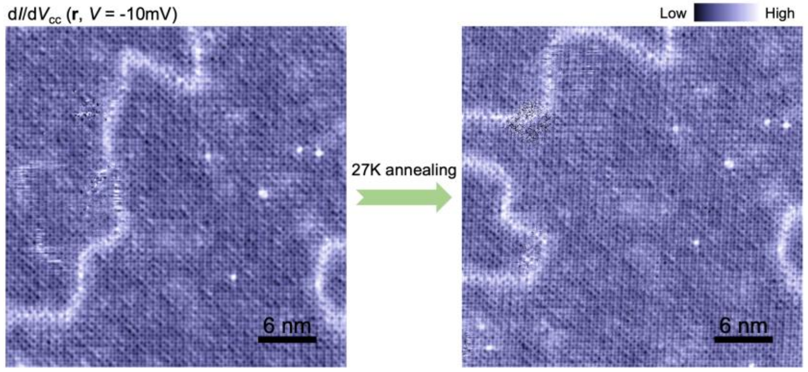
✓ Domain walls change the shape with temperature, time and magnetic field.

➢ Domain walls are metastable objects.

Domain walls change with time



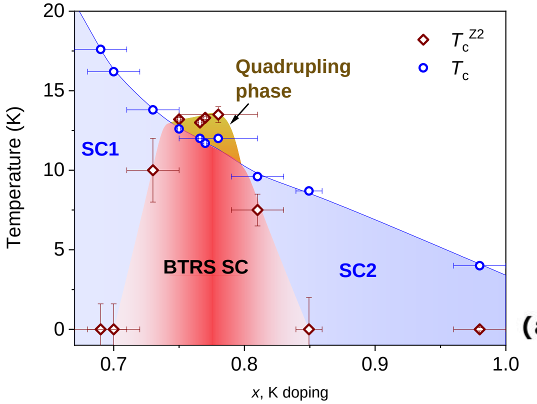
Thermocycling above T^*



Quanxin Hu et al., arXiv:2512.07272

Conclusions:

- The first singlet superconductor with broken time reversal symmetry discovered by μSR.
- Discovery of unquantized vortices.
- Evidence for a new state of matter – electron quadrupling condensate.

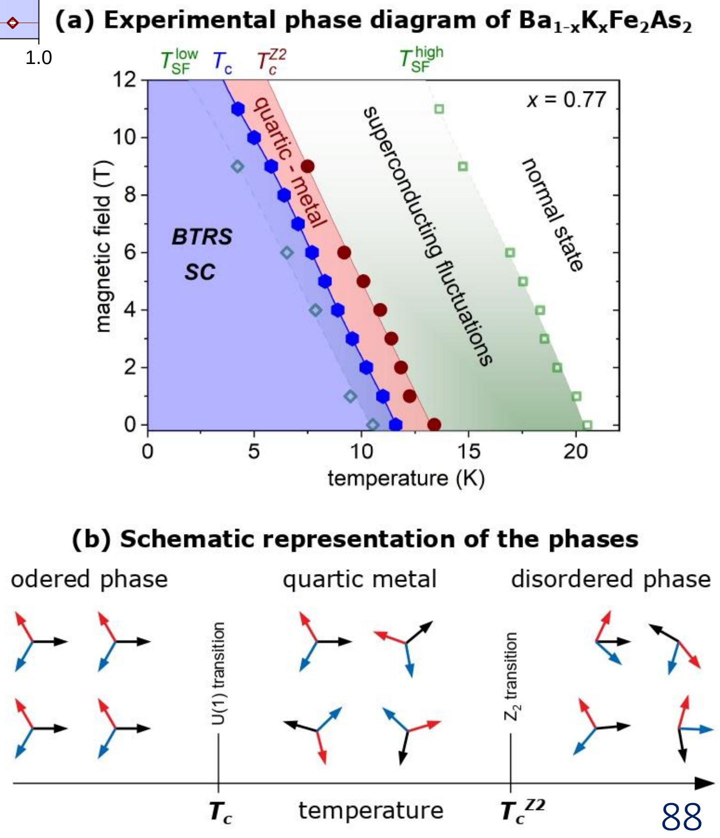


Looking for postdocs and PhD students!

vadim.grinenko@sjtu.edu.cn

V. Grinenko *et al.*, *Phys. Rev. B* **95**, 214511 (2017).
V. Grinenko *et al.*, *Nat. Phys.* **16**, 789 - 794 (2020).
V. Grinenko *et al.*, *Nat. Phys.* **17,** 1254–1259 (2021).
Y. Iguchi *et al. Science* **380**,1244-1247(2023).
I. Shipulin *et al.*, *Nat Commun* **14**, 6734 (2023).

Q. Hu, Y. Zheng et al. Nat Commun 16, 253 (2025).
F. Bärtl et al., arXiv:2501.11936.
Q. Z. Zhou et al., arXiv:2408.05902.
Y. Zheng, Q. Hu et al., *Sceince* (2026).
C. Halcrow et al., npj Quantum Materials **10**, 107 (2025).
Elena Corbae *et al*., arXiv:2510.06435.
Franz Eckelt et al., arXiv:2510.09151.



***Microscopic theory of electron quadrupling*: Albert Samoilenka and Egor Babaev**
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Implications/applications: fractional vortices are anyons, new platform for fractional Fluxonics, counterparts were suggested to composite topological order, symmetric mass generation in high energy physics model, composite orders in cold atoms.