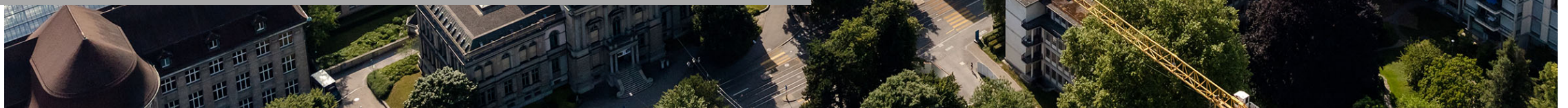


Different flavours of fermion
quadrupling condensates in
magic-angle twisted bilayer graphene

Dr. Ilaria Maccari
SNSF Postdoctoral Fellow, ETH Zürich

ERG 2024 - Les Diablares



Acknowledgements



Today's talk

Theory



Egor Babaev
(KTH Stockholm)



Julien Garaud
(University of Tours)



Johan Carlström
(Stockholm University)



Daniel Weston
(KTH Stockholm)

μ SR/NQR

Vadim Grinenko, Rajib Sarkar,
Hans-Henning Klauss,
TU Dresden, Germany/Shanghai
Jiao Tong University, China

$Ba_{1-x}K_xFe_2As_2$ single crystals Kunihiro Kihou and Chul-Ho Lee

AIST, Tsukuba, Japan

AC specific heat and ultrasound

Tino Gottschall, Gorbunov,
Denis, Sergei Zherlitsyn, Jochen
Wosnitza
HZDR, Dresden-Rossendorf,
Germany

AC microcalorimetry

Andreas Rydh,
Stockholm University, Sweden

STM Measurements

Quanxin Hu, Yu Zheng, Fashi
Yang, Yongwei Li, Chi-Ming
Yim
Shanghai Jiao Tong University,
China

X-Ray measurements

Ilya Shipulin, Yongwei Li
TU Dresden, Germany /
Shanghai Jiao Tong University,
China

Thermoelectrical transport

Federico Cagliaris, Ilya
Shipulin, Nadia Stegani,
Christoph Wuttke, Christian
Heß
IFW Dresden, Germany / CNR-
Spin Genova, Italy

Experiments

Beyond the pairing paradigm: fermion quadrupling condensates

Cooper's pair



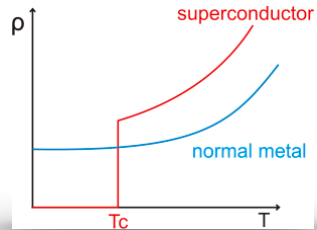
Ginzburg-Landau theory

Complex order parameter:

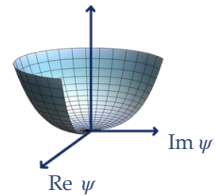
Cooper pairs density

$$\psi(x) = |\psi(x)| e^{i\phi(x)}$$

$$f = \frac{1}{2} \left| \left(\vec{\nabla} - ie\vec{A} \right) \psi \right|^2 + \alpha |\psi|^2 + \frac{\beta}{2} |\psi|^4 + \frac{\mathbf{B}^2}{2}$$

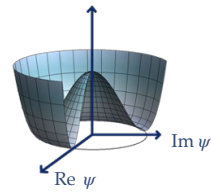
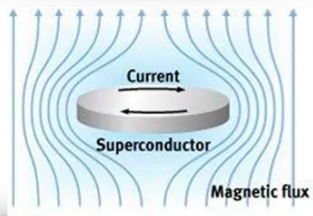


Zero Resistance



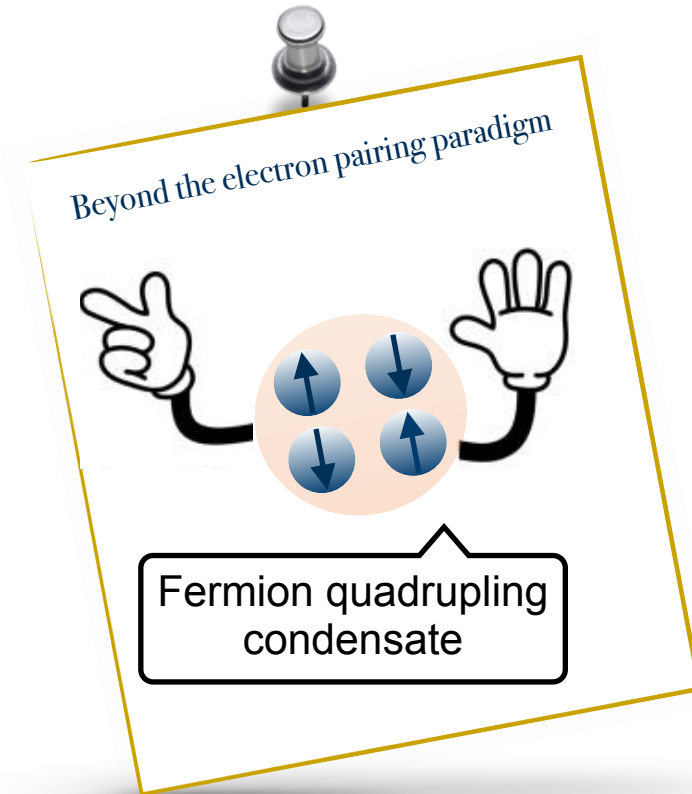
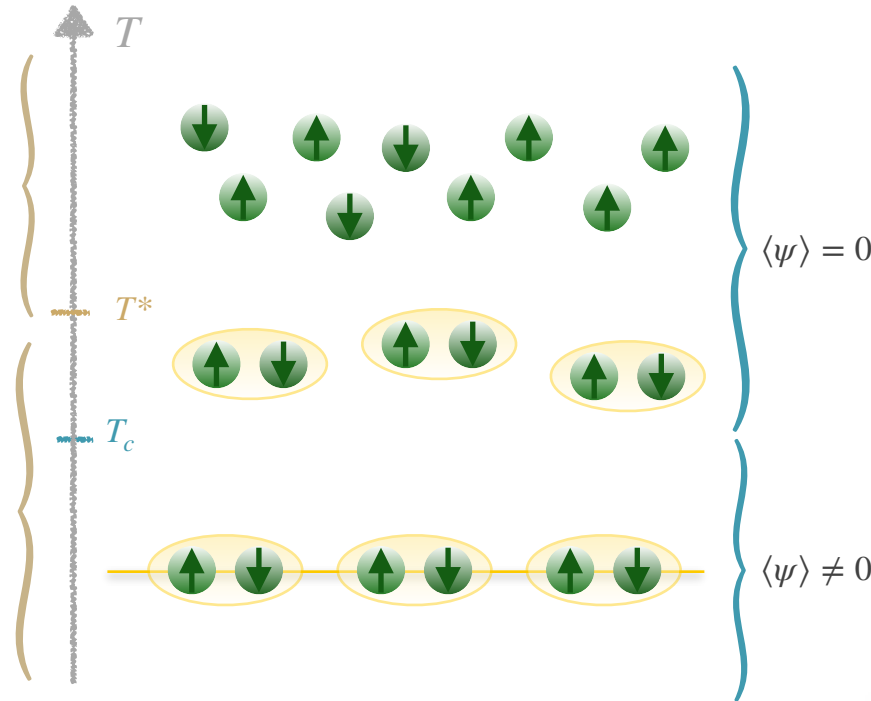
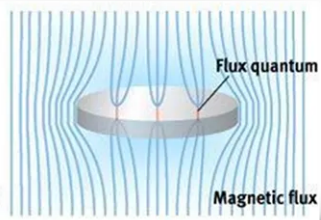
$$|\psi| = 0$$

Meissner effect
(low magnetic fields)



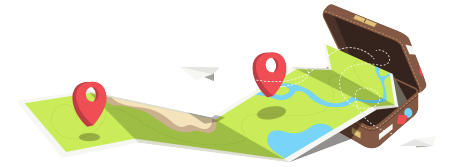
$$|\psi| \neq 0$$

Magnetic flux
quantisation
(type-II SCs, higher
magnetic fields)

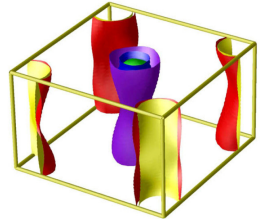


Spontaneous breaking of the U(1) gauge symmetry

How can we go beyond the pairing paradigm?

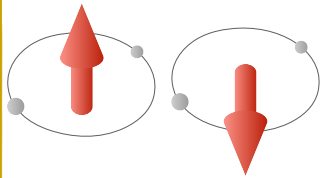


/ Multicomponent Superconductors

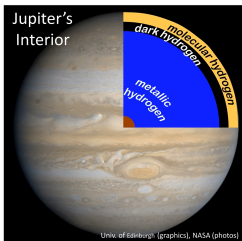


Multi-band SCs

I.I. Mazin, J. Schmalian,
Physica C 469 (2009) 614.



Single-band SCs
unconventional pairing
p-wave chiral SC:
 $\Delta(\vec{k}) = \Delta_x \sin(k_x a) + \Delta_y \sin(k_y a)$

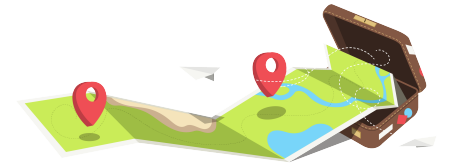


Mixture of charged condensates
(e.g. liquid metallic hydrogen)

/ $\psi_{i=1,2}(x) = |\psi_i(x)| e^{i\phi_i(x)}$ Two components: 

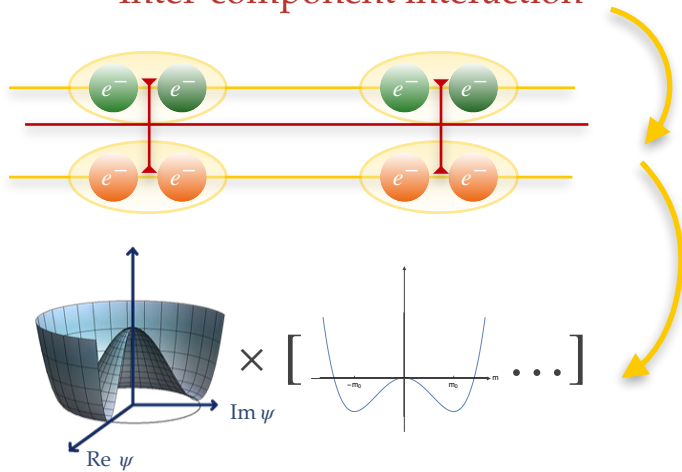
Component label \rightarrow

How can we go beyond the pairing paradigm?



1. Multicomponent Superconductors
2. Multiple broken symmetries

Inter-component interaction



SC state breaking multiple symmetries in the GS

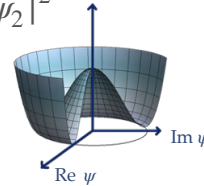
1. $\psi_{i=1,2}(x) = |\psi_i(x)| e^{i\phi_i(x)}$ Two components:
 Component label

2. Simplest case: two components interacting only via the EM field

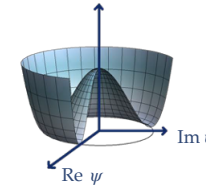
$$f = \frac{1}{2} \left| (\vec{\nabla} - ie\vec{A}) \psi_1 \right|^2 + \frac{1}{2} \left| (\vec{\nabla} - ie\vec{A}) \psi_2 \right|^2 + \sum_{i=1,2} \left[\alpha_i |\psi_i|^2 + \frac{\beta_i}{2} |\psi_i|^4 \right] + \frac{\mathbf{B}^2}{2}$$

$$f = \frac{|\psi_1|^2 |\psi_2|^2}{2\rho^2} \left[\vec{\nabla}(\phi_1 - \phi_2) \right]^2 + \frac{1}{2\rho^2} \left[|\psi_1|^2 \vec{\nabla} \phi_1 + |\psi_2|^2 \vec{\nabla} \phi_2 - e\rho^2 \vec{A} \right]^2 + \sum_{i=1,2} \left[\alpha_i |\psi_i|^2 + \frac{\beta_i}{2} |\psi_i|^4 \right] + \frac{\mathbf{B}^2}{2}$$

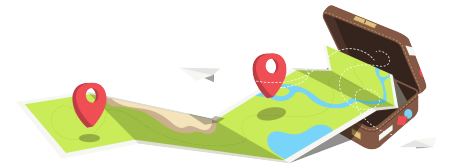
$$\rho^2 = |\psi_1|^2 + |\psi_2|^2$$



$U(1)$ Neutral mode \times $U(1)$ charged mode



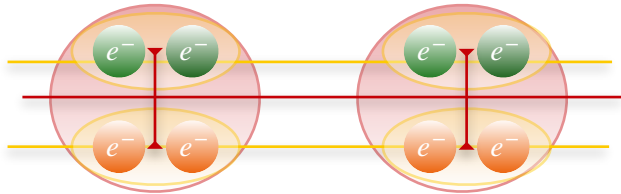
How can we go beyond the pairing paradigm?



1. Multicomponent Superconductors

2. Multiple broken symmetries

3. "Something" destroying coherence between Cooper pairs while preserving coherence between pairs of Cooper pairs



Composite vortices

$$(Q_1, Q_2) \text{ with: } Q_i = \frac{1}{2\pi} \oint d\vec{l} \cdot \vec{\nabla} \phi_i$$



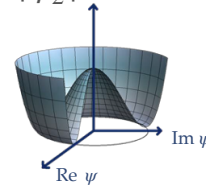
(0, 1) — (1, 0) — (1, 1) — (1, -1)

1. $\psi_{i=1,2}(x) = |\psi_i(x)| e^{i\phi_i(x)}$ Two components: Component label

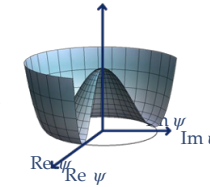
2. Simplest case: two components interacting only via the EM field

$$f = \frac{|\psi_1|^2 |\psi_2|^2}{2\rho^2} \left[\vec{\nabla}(\phi_1 - \phi_2) \right]^2 + \frac{1}{2\rho^2} \left[|\psi_1|^2 \vec{\nabla} \phi_1 + |\psi_2|^2 \vec{\nabla} \phi_2 - e\rho^2 \vec{A} \right]^2 + \sum_{i=1,2} \left[\alpha_i |\psi_i|^2 + \frac{\beta_i}{2} |\psi_i|^4 \right] + \frac{\mathbf{B}^2}{2}$$

$$\rho^2 = |\psi_1|^2 + |\psi_2|^2$$



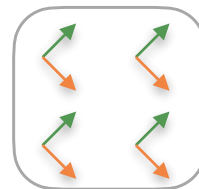
$U(1)$ Neutral mode \times $U(1)$ charged mode



3.

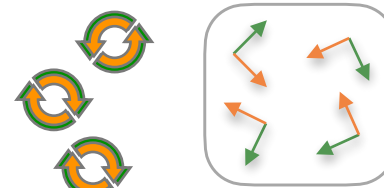
Rigid phase difference

$$\phi_1 - \phi_2$$



Superconducting Superfluid

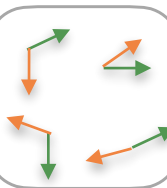
$$\langle \psi_{1,2} \rangle \neq 0$$



Fermion quadrupling condensate

$$\langle \psi_{1,2} \rangle = 0$$

$$\langle \psi_1 \psi_2^* + \psi_1^* \psi_2 \rangle \neq 0$$

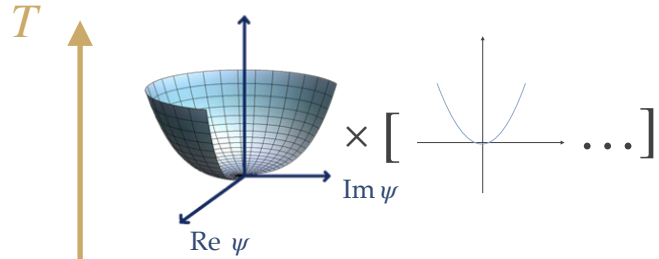


Normal Metal

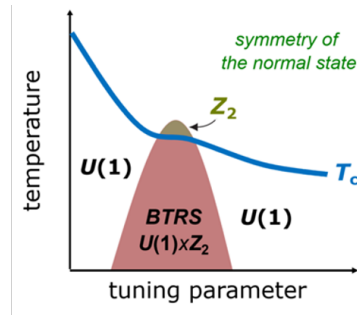
$$\langle \psi_{1,2} \rangle = 0$$

$$\langle \psi_1 \psi_2^* + \psi_1^* \psi_2 \rangle = 0$$

Beyond the pairing paradigm: fermion quadrupling condensates

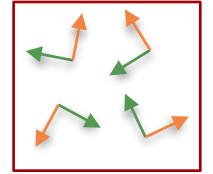


non-SC fermion quadruplets

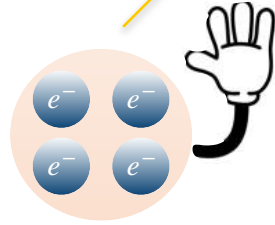


Typically associated with a phase difference rigidity

$$\langle \psi_i \psi_j^* + \psi_i^* \psi_j \rangle \neq 0$$

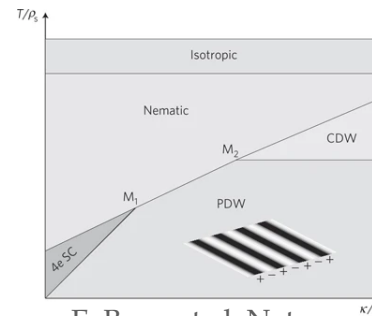


Fermion quadrupling condensates as vestigial phases



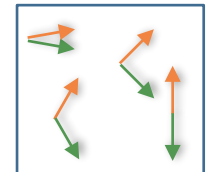
V. Grinenko, D. Weston, F. Caglieris, C. Wuttke, C. Hess, T. Gottschall, I. Maccari, et al., *Nat. Physics* 17, 1254–1259 (2021)

Charge-4e SC



Typically associated with a phase sum rigidity

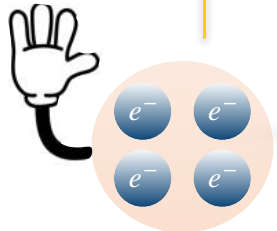
$$\langle \psi_i^* \psi_j^* + \psi_i \psi_j \rangle \neq 0$$



Multiple broken symmetries

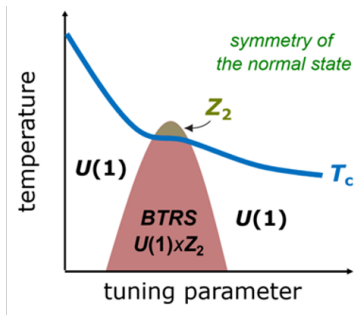
E. Berg, et al. *Nature Physics* 5, 830 (2009)

Beyond the pairing paradigm: fermion quadrupling condensates



Above T_c

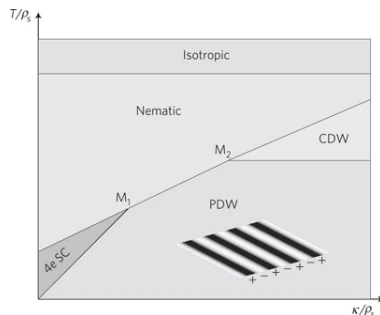
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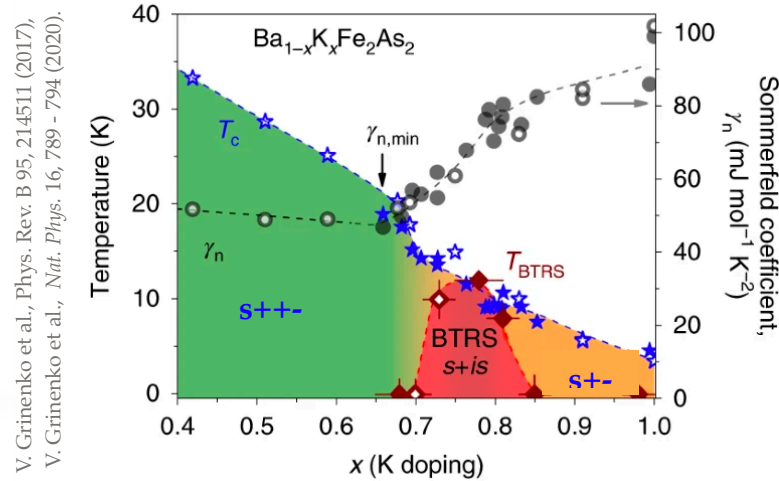
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First experimental observation in a multi band iron-based SC

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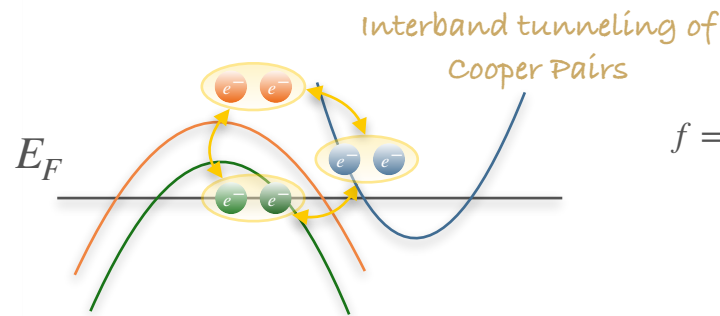
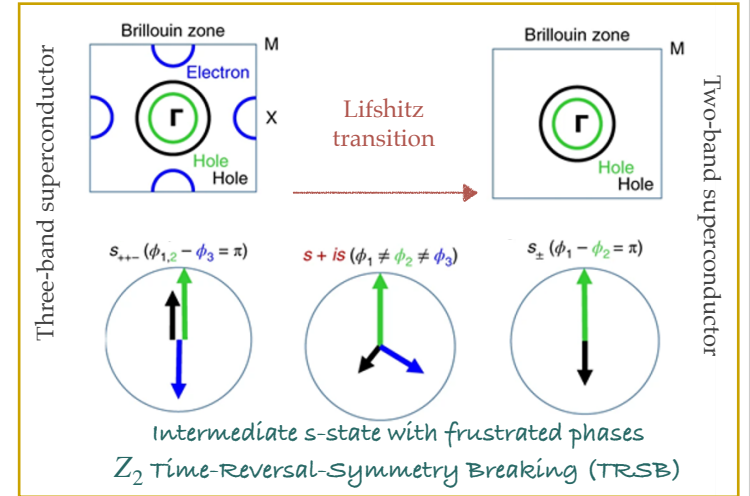


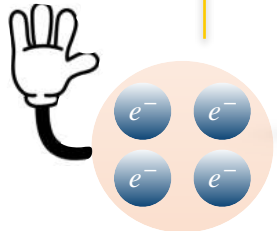
Illustration of a three-band SC

$$f = \frac{1}{2} \sum_i \left| \left(\vec{\nabla} - ie\vec{A} \right) \psi_i \right|^2 + \sum_i \left[\alpha_i |\psi_i|^2 + \frac{\beta_i}{2} |\psi_i|^4 \right] + \sum_{i>j} \eta_{ij} |\psi_i| |\psi_j| \cos(\phi_i - \phi_j) + \frac{B^2}{2};$$

Josephson intercomponent coupling

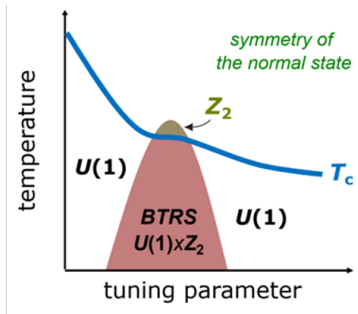
$$\propto \frac{1}{2} [\psi_i \psi_j^* + \psi_i^* \psi_j]$$

Beyond the pairing paradigm: fermion quadrupling condensates



Above T_c

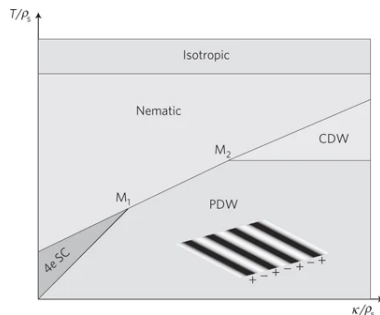
non-SC fermion quadruplets



V. Grinenko, D. Weston, F. Caglieris, C. Wuttke, C. Hess, T. Gottschall, I. Maccari, et al., *Nat. Physics* 17, 1254–1259 (2021)

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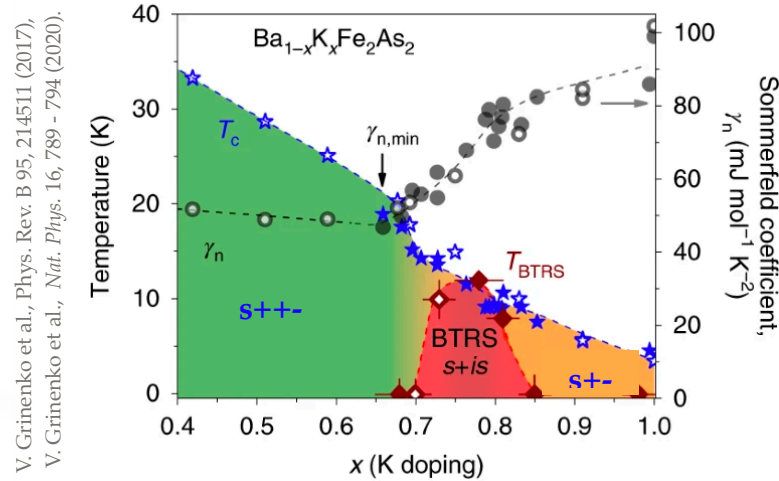
Charge-4e SC



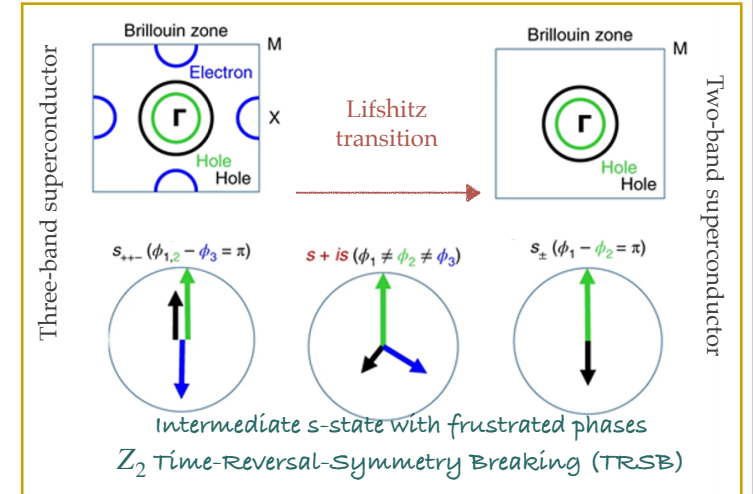
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V. Grinenko et al., *Nat. Phys.* 16, 789 - 794 (2020).



Interband tunneling of Cooper Pairs

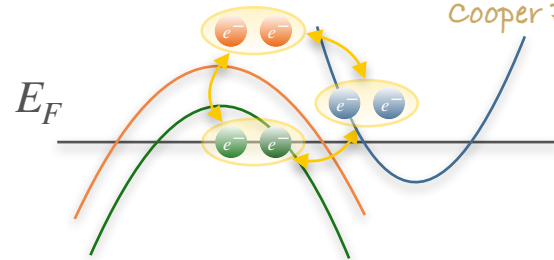
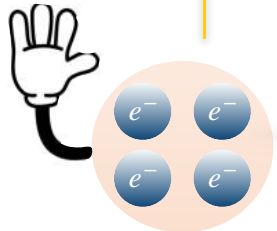


Illustration of a three-band SC

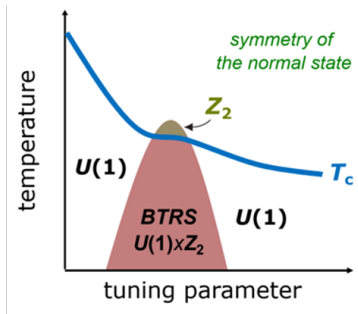
$$f = \frac{1}{2\rho^2} \left[\left(\sum_{i=1,2,3} |\psi_i|^2 \vec{\nabla} \phi_i \right) - e\rho^2 \vec{A} \right]^2 + \sum_{i>j} \frac{|\psi_i|^2 |\psi_j|^2}{2\rho^2} \left[\vec{\nabla} (\phi_i - \phi_j) \right]^2 + \sum_{i>j} \eta_{ij} |\psi_i| |\psi_j| \cos(\phi_i - \phi_j) + \sum_i V(|\psi_i|^2,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

Beyond the pairing paradigm: fermion quadrupling condensates



Above T_c

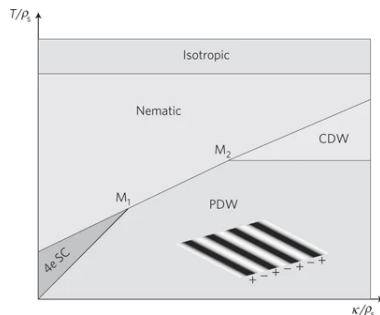
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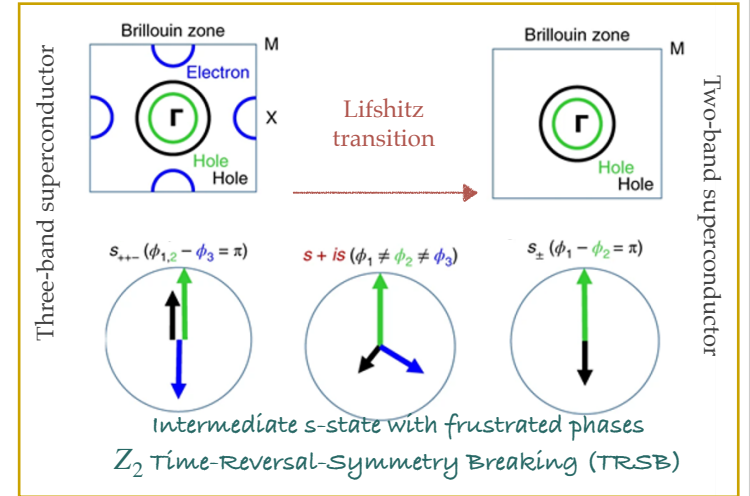
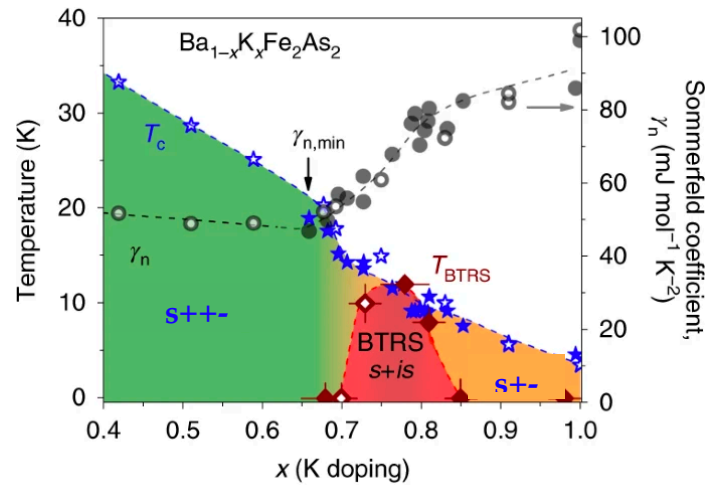


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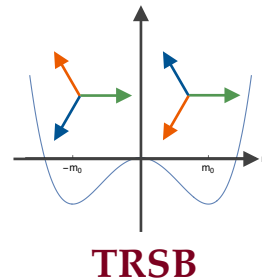
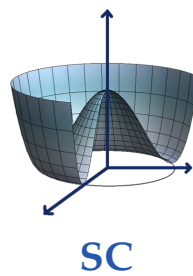
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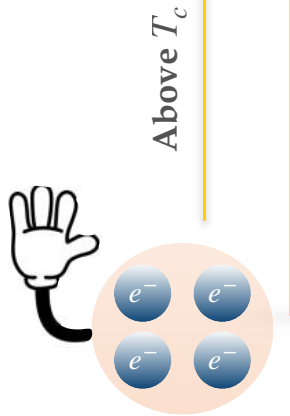


The ground state breaks a total $U(1) \times Z_2$ symmetry



$$f = \frac{1}{2\rho^2} \left[\left(\sum_{i=1,2,3} |\psi_i|^2 \vec{\nabla} \phi_i \right) - e\rho^2 \vec{A} \right]^2 + \sum_{i>j} \frac{|\psi_i|^2 |\psi_j|^2}{2\rho^2} \left[\vec{\nabla} (\phi_i - \phi_j) \right]^2 + \sum_{i>j} \eta_{ij} |\psi_i| |\psi_j| \cos(\phi_i - \phi_j) + \sum_i V(|\psi_i|^2, |\psi_i|^4) + \frac{B^2}{2}$$

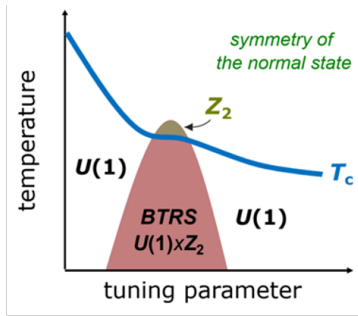
Beyond the pairing paradigm: fermion quadrupling condensates



Above T_c

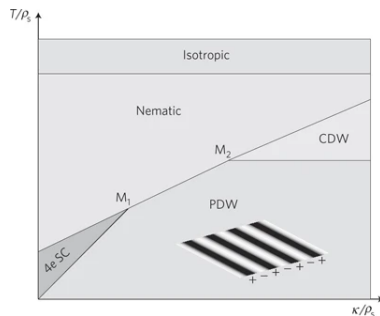
Below T_c

non-SC fermion quadruplets



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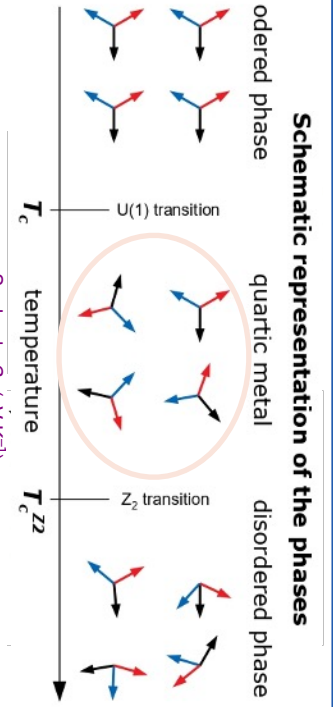
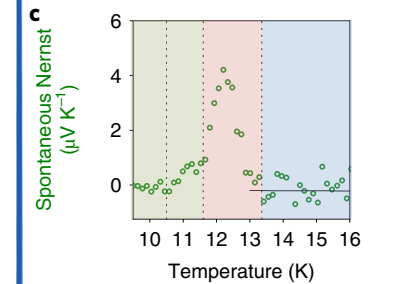
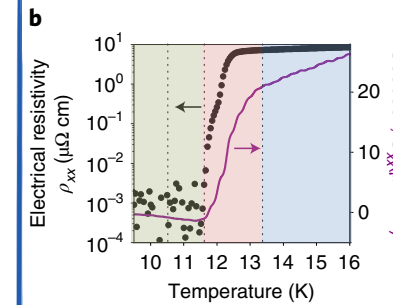
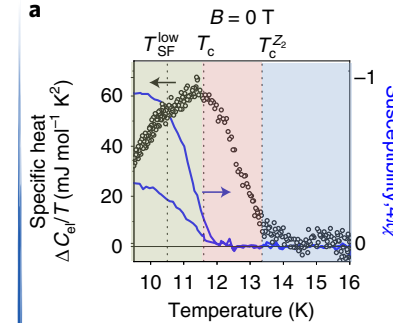
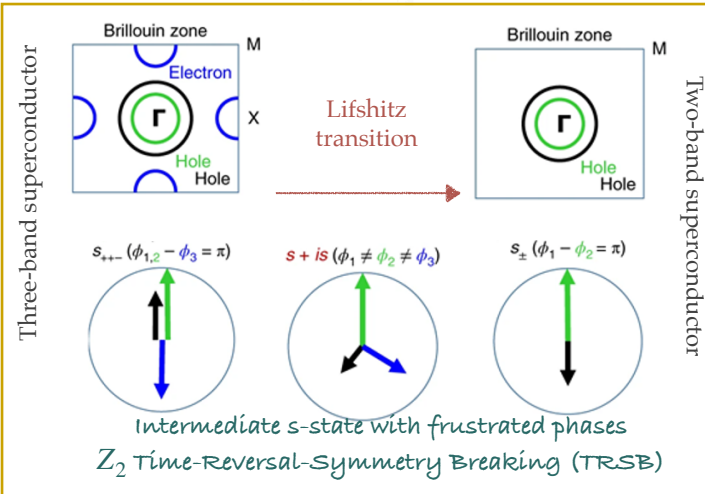
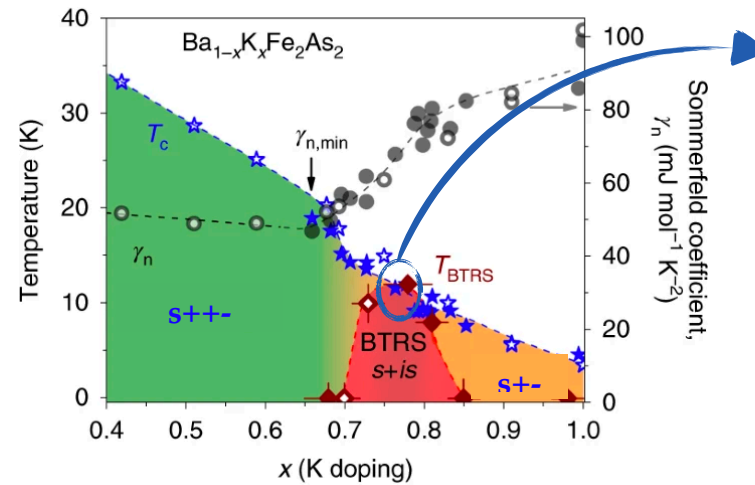
Charge-4e SC



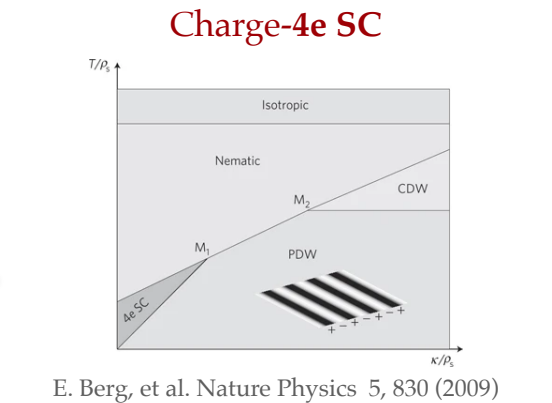
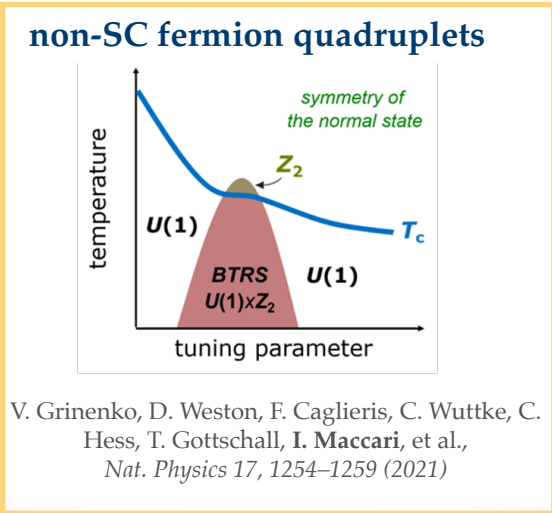
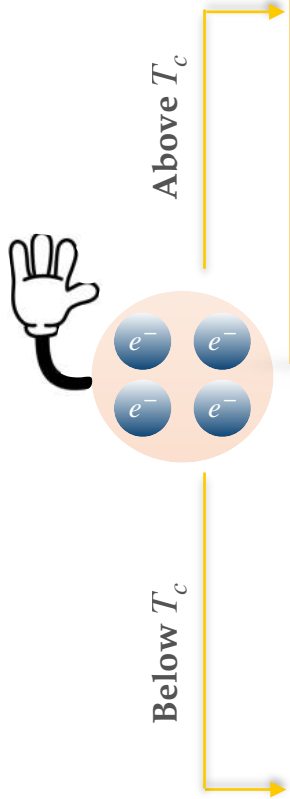
E. Berg, et al. *Nature Physics* 5, 830 (2009)

First experimental observation in a multi band iron-based SC

V. Grinenko, D. Weston, F. Caglieris, C. Wuttke, C. Hess, T. Gottschall, I. Maccari, et al., *Nature Physics* 17, 1254–1259 (2021)



Beyond the pairing paradigm: fermion quadrupling condensates



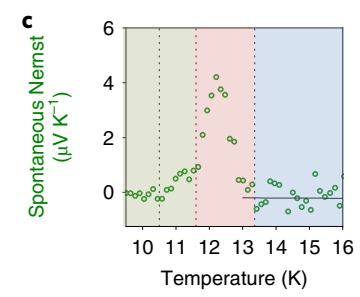
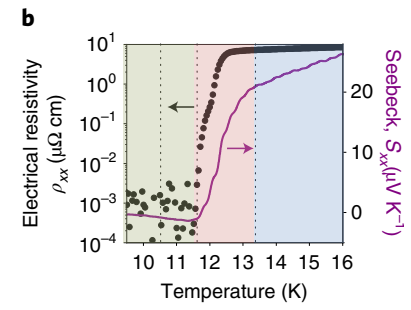
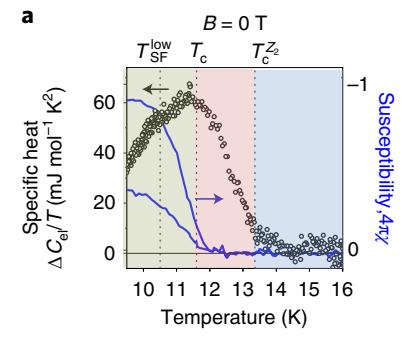
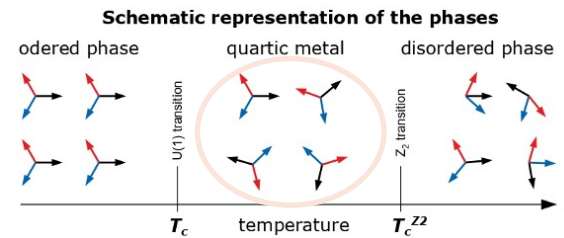
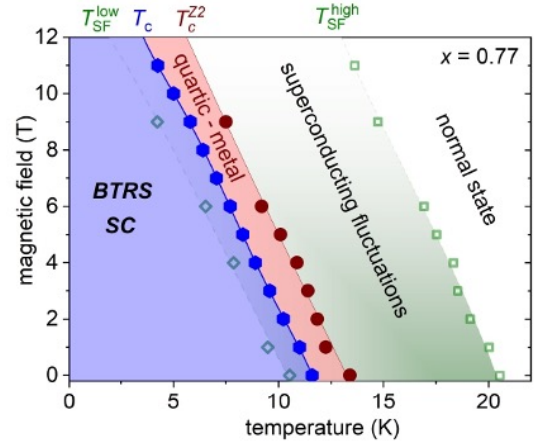
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Main experimental evidences

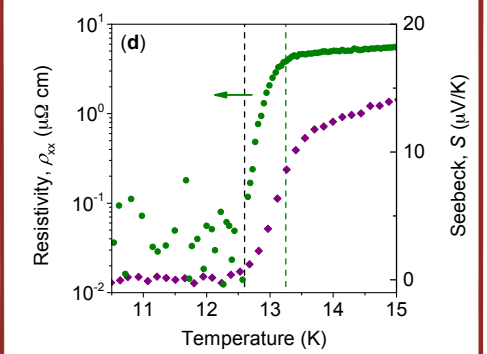
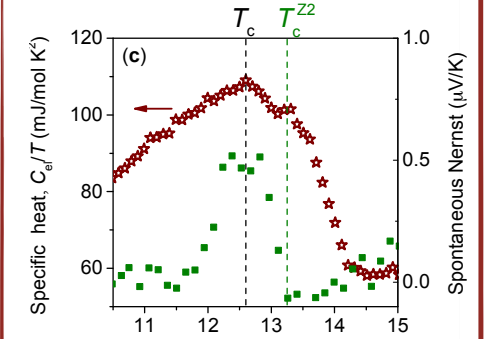
- * Muon-spin relaxation measurements
- * Spontaneous Nernst effect
- * Ultrasound measurements
- * **Two peaks in the specific heat**

Experimental phase diagram of Ba_{1-x}K_xFe₂As₂



Calorimetric signatures

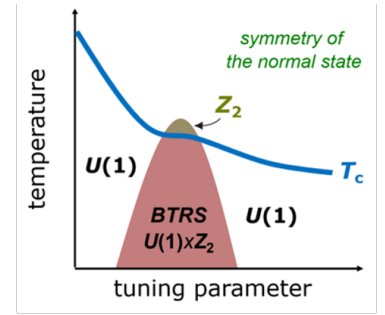
I. Shipulin, N. Stegani, I. Maccari, et al., *Nature Communications* 14, 6734 (2023)



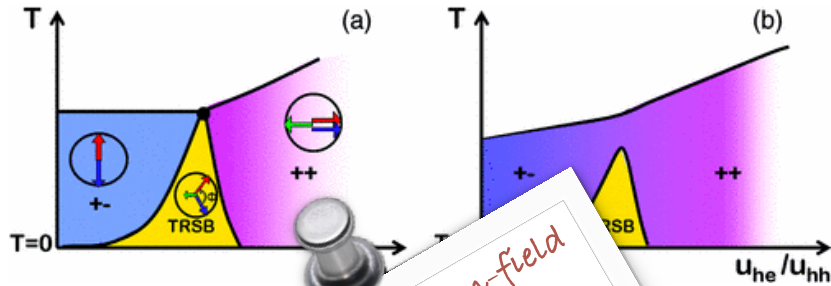
Monte Carlo results on an effective 3D multicomponent model

The role of different intercomponent interactions

I. Maccari and E. Babaev Phys. Rev. B 105, 214520 (2022)

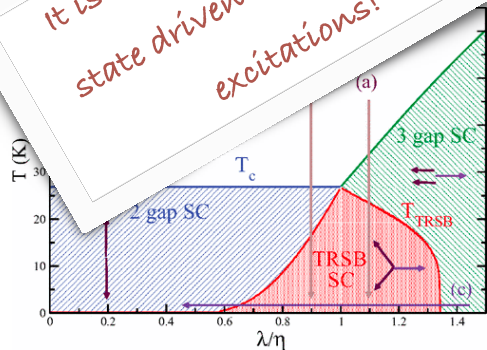


Mean-field models



S. Maiti and A. V. ...

It is a beyond-mean-field state driven by topological excitations!



M. Marciani et al, Phys. Rev. B 88, 214508 (2013)

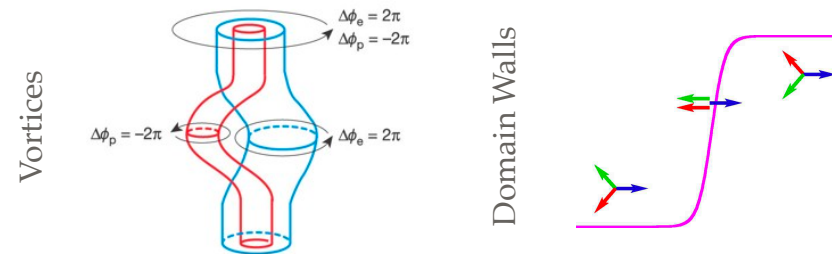
Increasing $x =$ hole doping

Renormalization group approach

- ✓ It accounts for the proliferation of topological phase excitations.
- ◆ You need to properly account for the coupling between the different topological defects.

Monte Carlo numerical simulations

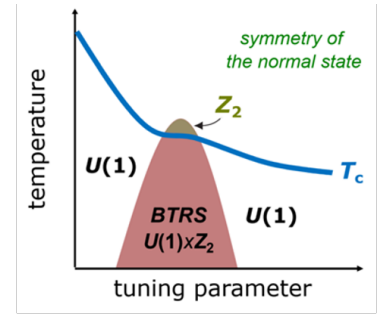
- ✓ They allow for a systematic and unbiased study of the proliferation of different topological defects and their interactions.



Monte Carlo results on an effective 3D multicomponent model

The role of different intercomponent interactions

I. Maccari and E. Babaev *Phys. Rev. B* 105, 214520 (2022)



Starting point:

Three-component GL model with phase frustration

$$f = \frac{1}{2}(\nabla \times \mathbf{A})^2 + \sum_i \frac{1}{2} |(\nabla + ie\mathbf{A})\psi_i|^2 + a_i |\psi_i|^2 + \frac{b_i}{2} |\psi_i|^4 + \sum_{i<j} \eta_{ij} |\psi_i| |\psi_j| \cos(\phi_i - \phi_j)$$



At zero external magnetic field: any BTRS quadrupling fermionic condensate in the type-II regime ($e \rightarrow 0$)

What are we missing?

+Andreev-Bashkin interaction

A. F. Andreev and E. P. Bashkin, *Sov. Phys. JETP* 42, 164 (1975);
A. J. Leggett, *Rev. Mod. Phys.* 47, 331 (1975).

$$f = \frac{1}{2}(\nabla \times \mathbf{A})^2 + \sum_i \frac{1}{2} |(\nabla + ie\mathbf{A})\psi_i|^2 + a_i |\psi_i|^2 + \frac{b_i}{2} |\psi_i|^4 + \sum_{i<j} \eta_{ij} |\psi_i| |\psi_j| \cos(\phi_i - \phi_j) - \nu \sum_{j>i} \vec{J}_j \cdot \vec{J}_i$$

Mapping to a two-component London effective model

J. Garaud, M. Silaev, and E. Babaev, *Physica C* 533, 63 (2017)

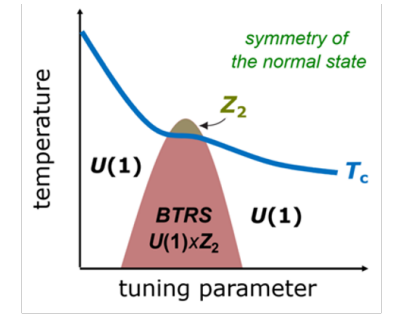
$$f = \frac{1}{2} (\nabla \times \mathbf{A})^2 + \sum_{i=1,2} \frac{\rho_i}{2} (\nabla \phi_i - e\mathbf{A})^2 - \nu (\nabla \phi_1 - e\mathbf{A}) \cdot (\nabla \phi_2 - e\mathbf{A}) + \eta_2 \cos[2(\phi_1 - \phi_2)]$$

Phase frustration via a biquadratic Josephson coupling

Monte Carlo results on an effective 3D multicomponent model

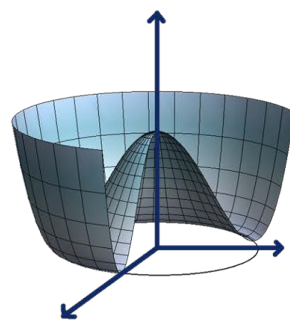
The role of different intercomponent interactions

I. Maccari and E. Babaev *Phys. Rev. B* 105, 214520 (2022)

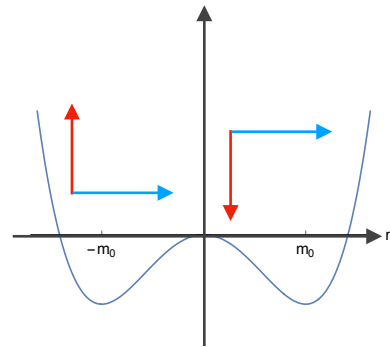


$$f = \frac{1}{2} (\nabla \times \mathbf{A})^2 + \sum_{i=1,2} \frac{\rho_i}{2} (\nabla \phi_i - e\mathbf{A})^2 - \nu (\nabla \phi_1 - e\mathbf{A}) \cdot (\nabla \phi_2 - e\mathbf{A}) + \eta_2 \cos[2(\phi_1 - \phi_2)]$$

Phase frustration via a biquadratic Josephson coupling

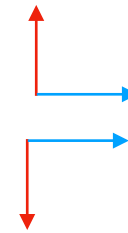


U(1)



Z₂

Two possible chiralities:



$$(\rho_1, \rho_2 e^{i\frac{\pi}{2}})$$

$$(\rho_1, \rho_2 e^{-i\frac{\pi}{2}})$$

$$\rho_1 = \rho_2 = 1 \quad f = \frac{1-\nu}{4} (\nabla \phi_1 + \nabla \phi_2 - 2e\mathbf{A})^2 + \frac{1+\nu}{4} (\nabla \phi_1 - \nabla \phi_2)^2 + \eta_2 \cos[2(\phi_1 - \phi_2)] + \frac{1}{2} (\nabla \times \mathbf{A})^2$$

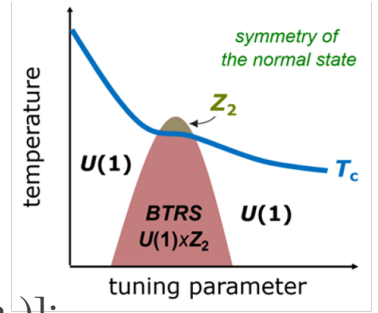
Key intercomponent interactions:

- ◆ e : coupling with the EM field → increasing e decreases the cost of (1,1) vortices
- ◆ η_2 : biquadratic Josephson coupling → increasing η_2 increases the cost of domain walls
- ◆ ν : current-current interaction → increasing ν decreases the relative cost of phase sum fluctuations

Monte Carlo results on an effective 3D multicomponent model

The role of different intercomponent interactions

I. Maccari and E. Babaev *Phys. Rev. B* 105, 214520 (2022)



Extreme type-II limit: $\lambda \rightarrow \infty \rightarrow e = 0$

$$f = \frac{1-\nu}{4} (\nabla\phi_1 + \nabla\phi_2)^2 + \frac{1+\nu}{4} (\nabla\phi_1 - \nabla\phi_2)^2 + \eta_2 \cos[2(\phi_1 - \phi_2)];$$

Helicity modulus sum
(superfluid stiffness)

$$\Upsilon_+^\mu = \frac{1}{L^2} \left. \frac{\partial^2 F(\{\phi_i'\})}{\partial \delta_\mu^2} \right|_{\delta_\mu=0}$$

$$\begin{pmatrix} \phi_1'(\mathbf{r}) \\ \phi_2'(\mathbf{r}) \end{pmatrix} = \begin{pmatrix} \phi_1(\mathbf{r}) + \delta \cdot \mathbf{r}_\mu \\ \phi_2(\mathbf{r}) + \delta \cdot \mathbf{r}_\mu \end{pmatrix}.$$

Fermion quadrupling
condensate with TRSB

$$T_c \quad \text{---} \quad T_c^{Z_2}$$

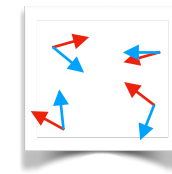
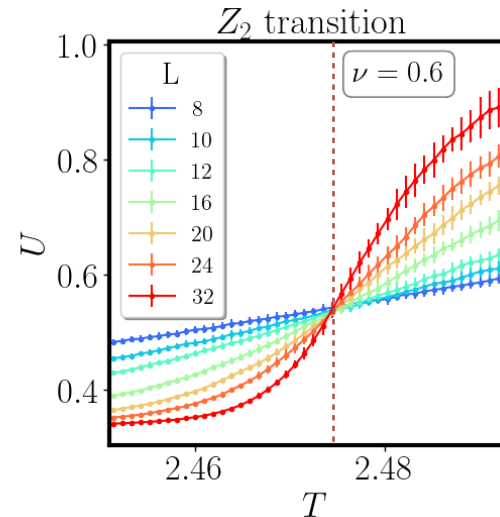
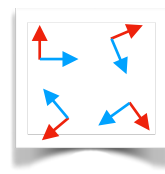
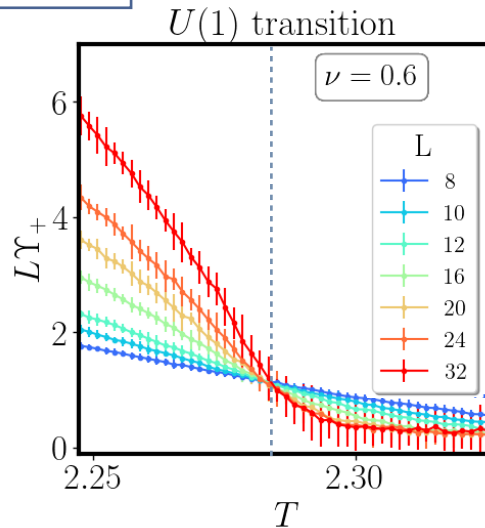
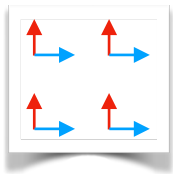
Z_2 Ising parameter

$m=+1$ $m=-1$

$[\phi_1 - \phi_2]_{-\pi,\pi} > 0$ $[\phi_1 - \phi_2]_{-\pi,\pi} < 0$

Binder cumulant

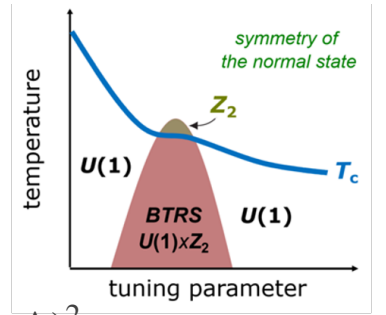
$$U = \frac{\langle m^4 \rangle}{3\langle m^2 \rangle^2}$$



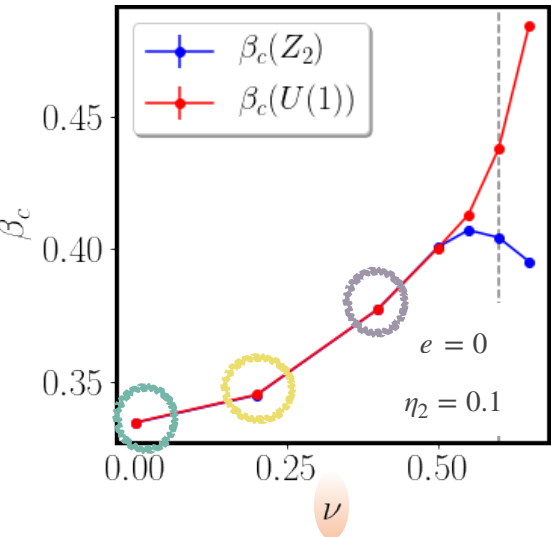
Monte Carlo results on an effective 3D multicomponent model

The role of different intercomponent interactions

I. Maccari and E. Babaev *Phys. Rev. B* 105, 214520 (2022)

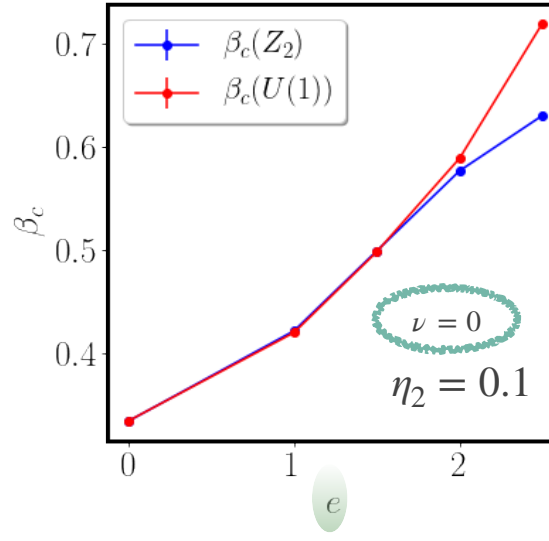
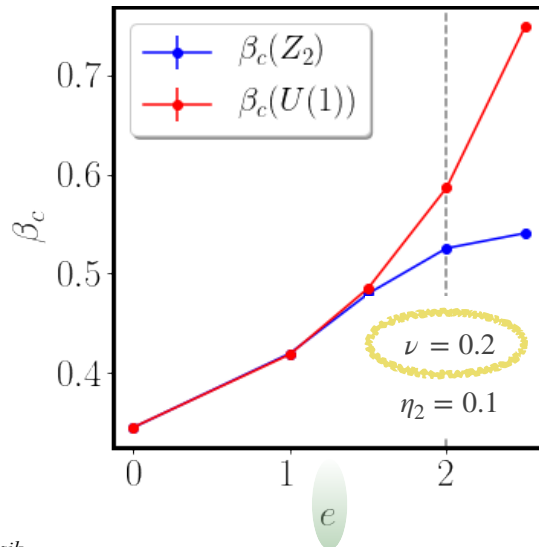


$$\rho_1 = \rho_2 = 1 \quad f = \frac{1-\nu}{4} (\nabla\phi_1 + \nabla\phi_2 - 2e\mathbf{A})^2 + \frac{1+\nu}{4} (\nabla\phi_1 - \nabla\phi_2)^2 + \eta_2 \cos[2(\phi_1 - \phi_2)] + \frac{1}{2} (\nabla \times \mathbf{A})^2$$

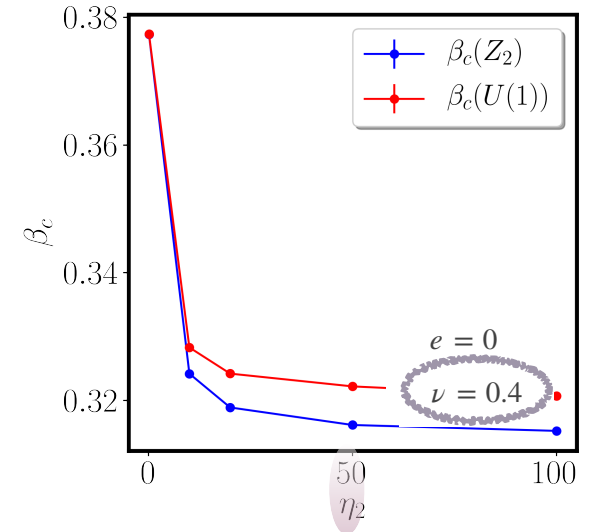


It decreases the relative cost of phase sum fluctuations

It decreases the cost of (1,1) vortices

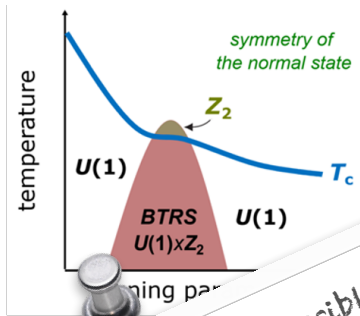


It increases the cost of domain walls



Beyond the pairing paradigm: fermion quadrupling condensates

non-SC fermion quadruplets



Above T_c

V. Grinenko

Bosonic counterparts are also possible, where $\langle b_i \rangle = 0$ but $\langle b_i^\dagger b_j \rangle \neq 0$

Observation of counterflow superfluidity in a two-component Mott insulator

Yong-Guang Zheng^{1,2,3,*}, An Luo^{1,2,*}, Ying-Chao Shen^{1,2}, Ming-Gen He^{1,2}, Zi-Hang Zhu^{1,2}, Ying Liu^{1,2}, Wei-Yong Zhang^{1,2}, Hui Sun^{1,2}, Youjin Deng^{1,2,3}, Zhen-Sheng Yuan^{1,2,3} and Jian-Wei Pan^{1,2,3}

¹Hefei National Research Center for Physical Sciences at the Microscale and School of Physical Sciences, University of Science and Technology of China, Hefei 230026, China

²CAS Center for Excellence in Quantum Information and Quantum Physics, University of Science and Technology of China, Hefei 230026, China

³Hefei National Laboratory, University of Science and Technology of China, Hefei 230088, China

(Dated: March 7, 2024)

E. Berg, et al. Nature Physics 5, 830 (2009)

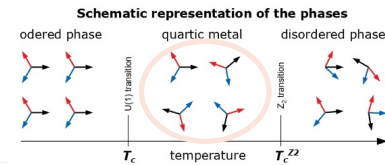
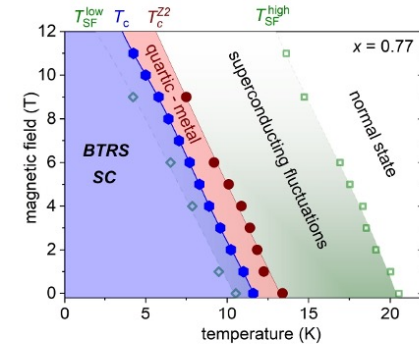
First experimental observation in a multi band iron-based SC

V. Grinenko, D. Weston, F. Caglieris, C. Wuttke, C. Hess, T. Gottschall, I. Maccari, et al.,
Nat. Physics 17, 1254–1259 (2021)

Main experimental evidences

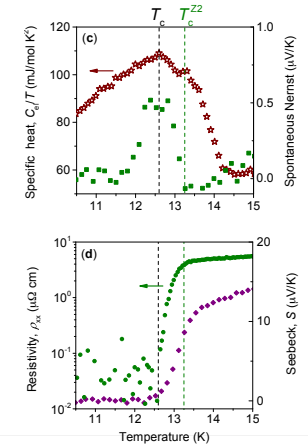
- * Muon-spin relaxation measurements
- Spontaneous Nernst effect
- Ultrasound measurements
- peaks in the specific heat

Experimental phase diagram of $Ba_{1-x}K_xFe_2As_2$

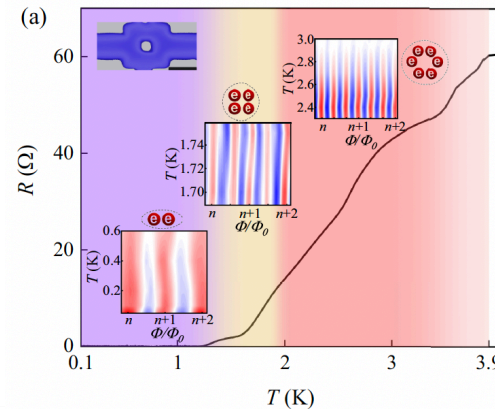
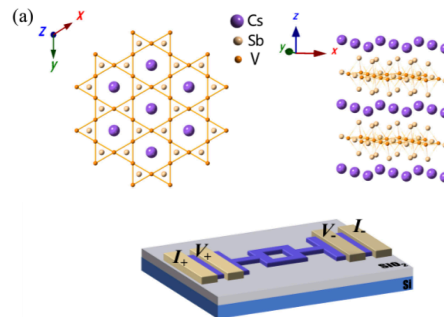


Calorimetric signatures

I. Shipulin, N. Stegani, I. Maccari, et al.,
Nature Communications 14, 6734 (2023)



First experimental signatures in a Kagome SC ring device



Main experimental evidence:

- * $h/4e$ and $h/6e$ quantization of magnetic flux in magnetoresistance oscillations.

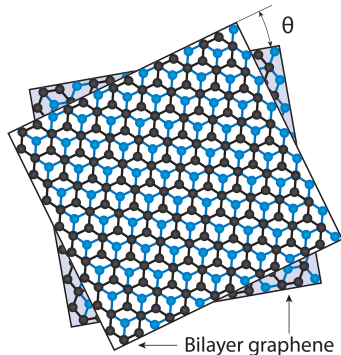
J. Ge et al., Phys. Rev. X 14, 021025 (2024)

Hunting fermion quadruplets: the case of twisted bilayer graphene

I. Maccari, J. Carlström and E. Babaev, *Phys. Rev. B* 107 (6), 064501 (2023)

MATBG promising candidate to stabilize fermion quadrupling condensates

1. 2D \rightarrow more phase fluctuations
2. Single-band SC unconventional ground state (multicomponent)



Picture from:
X. Liu et al., *Nature* 583, 221 (2020)

Proposed pairing ad $n = -2$:
 $d \pm id \rightarrow (\Delta_{E_1}, \Delta_{E_2})$

Key inter-component potential term

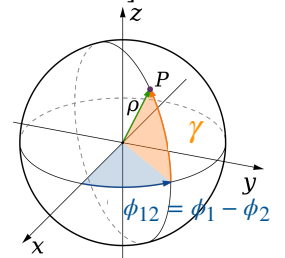
$$V(\Delta_{E_1}, \Delta_{E_2}) = 2K |\Delta_1|^2 |\Delta_2|^2 [\cos(2(\phi_1 - \phi_2)) - 1]$$

[D. V. Chichinadze, L. Classen, A. Chubokov, *PRB* 101, 224513 (2020)]

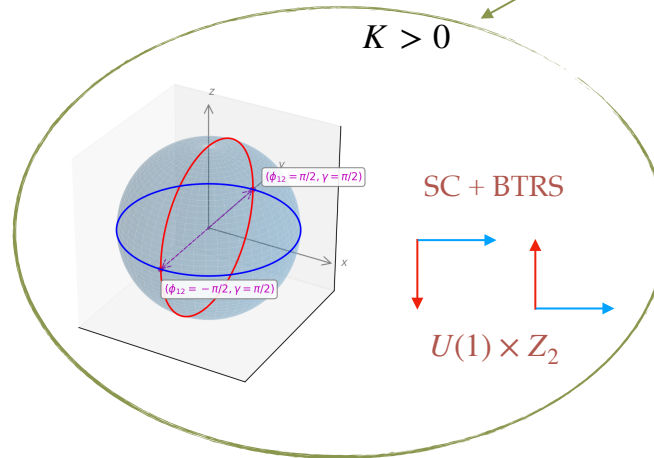
$$f = \frac{1}{2}(\vec{\nabla} \times \vec{A})^2 + \sum_{i=1,2} \left[\frac{1}{2} |(\vec{\nabla} - ie\vec{A})\Delta_i|^2 \right] + V(\Delta_1, \Delta_2); \quad V(\Delta_1, \Delta_2) = 2K |\Delta_1|^2 |\Delta_2|^2 [\cos(2(\phi_1 - \phi_2)) - 1]$$

$$\begin{cases} \Delta_1 = |\Delta_1| e^{i\phi_1}; \\ \Delta_2 = |\Delta_2| e^{i\phi_2}; \\ |\Delta_1|^2 + |\Delta_2|^2 = \rho^2 = 1. \end{cases} \rightarrow \begin{cases} \Delta_1 = |\cos(\frac{\gamma}{2})| e^{i\phi_1}; \\ \Delta_2 = |\sin(\frac{\gamma}{2})| e^{i\phi_2}; \end{cases}$$

$$V(\Delta_1, \Delta_2) = \frac{K}{2} \sin^2(\gamma) [\cos(2(\phi_1 - \phi_2)) - 1]$$

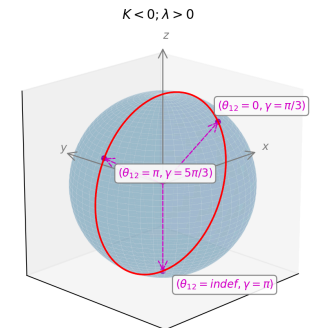


$K > 0$



$K < 0$

Nematic SC
 $U(1) \times Z_3$



$$V_6(\Delta_1, \Delta_2) = \frac{\lambda}{2} [(\Delta_1 - i\Delta_2)^3 (\Delta_1^* - i\Delta_2^*)^3 + c.c.]$$

To be added to lift the accidental degeneracy

Two complementary regions of parameter space describing a chiral ($K > 0$) and a nematic ($K < 0$) SC

Hunting fermion quadruplets: the case of twisted bilayer graphene

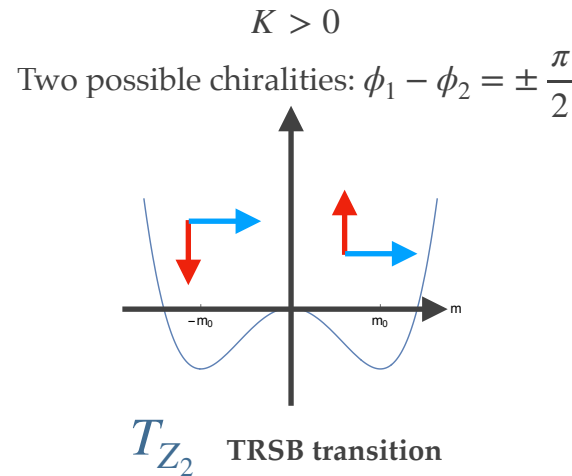
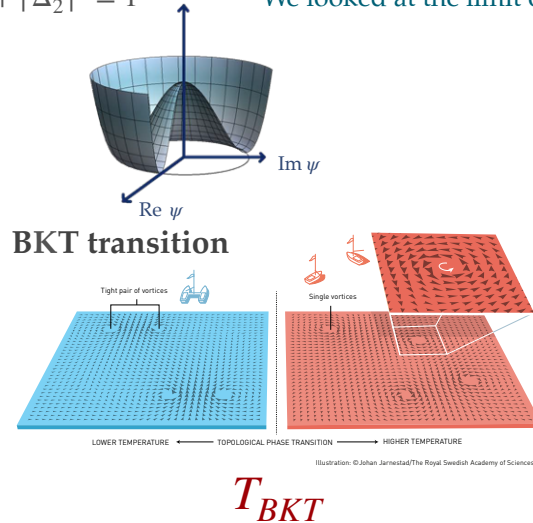
I. Maccari, J. Carlström and E. Babaev, *Phys. Rev. B* 107 (6), 064501 (2023)

Large-scale Monte Carlo simulations $K > 0$ $e = 0$

$$f = \frac{1}{2\rho^2} \left[|\Delta_1|^2 \nabla \phi_1 + |\Delta_2|^2 \nabla \phi_2 \right]^2 + \frac{|\Delta_1|^2 |\Delta_2|^2}{2\rho^2} \left[\nabla(\phi_1 - \phi_2) \right]^2 + \frac{1}{2} \left[(\nabla |\Delta_1|)^2 + (\nabla |\Delta_2|)^2 \right] + 2K |\Delta_1|^2 |\Delta_2|^2 [\cos(2(\phi_1 - \phi_2)) - 1]$$

$$\rho^2 = |\Delta_1|^2 + |\Delta_2|^2 = 1$$

*We looked at the limit of very large penetration length, so we neglect the coupling to the vector potential



$T_{BKT} < T_c^{Z_2}?$ SC vortices Trigger BKT transition	$T_{BKT} = T_c^{Z_2}?$ Fractional vortices + DW string Trigger BKT and Z_2 transition	$T_c^{Z_2} < T_{BKT}?$ Z_2 DW Trigger Z_2 transition
---	---	--

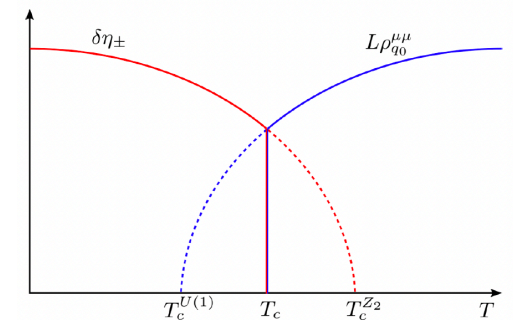
Stabilising fermion quadruplets is not easy...

Chiral and nematic SC in 3D

* "Absence of Ginzburg-Landau mechanism for vestigial order in the normal phase above a two-component superconductor"
 P. T. How et al., *Phys. Rev. B* 107, 104514 (2023)

* "First order SC phase transition in a chiral p+ip system"
 H. H. Haugen et al., *Phys. Rev. B* 104, 104515 (2021).

Preemptive first-order phase transition



Hunting fermion quadruplets: the case of twisted bilayer graphene

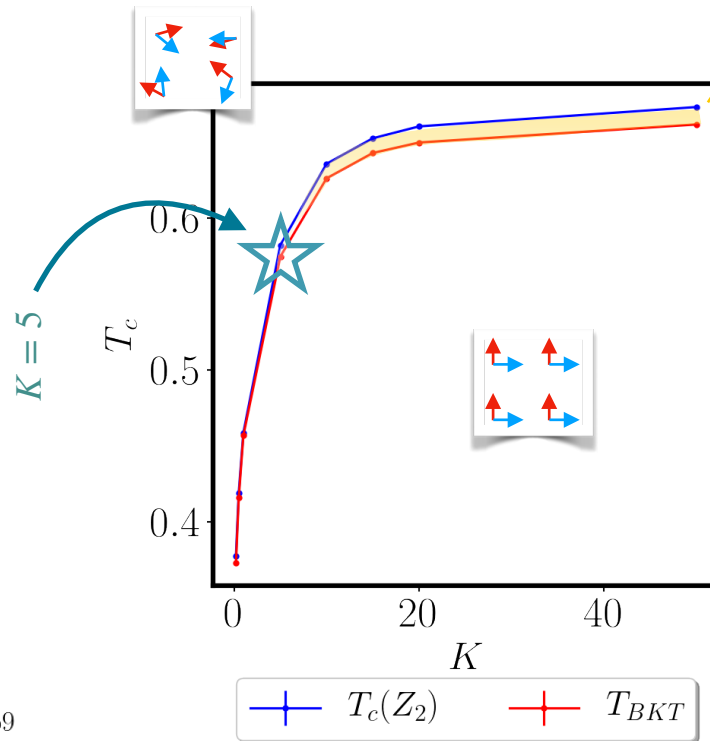
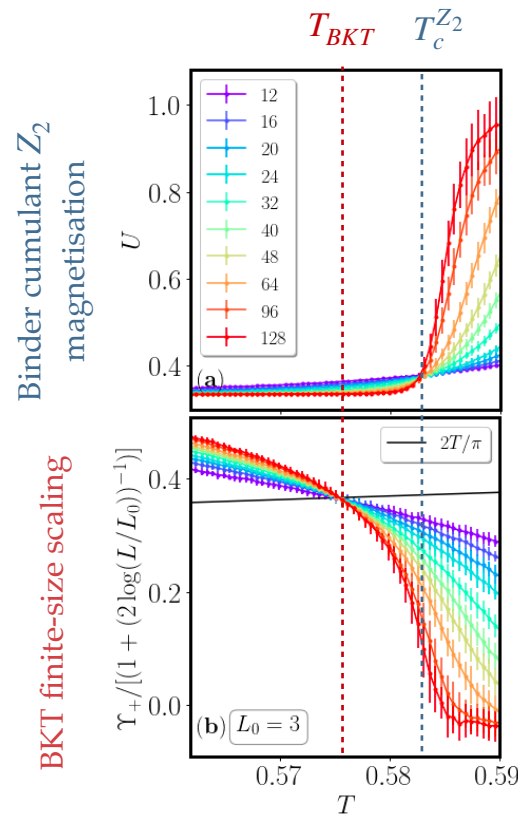
I. Maccari, J. Carlström and E. Babaev, *Phys. Rev. B* 107 (6), 064501 (2023)

Large-scale Monte Carlo simulations $K > 0$ $e = 0$

$$f = \frac{1}{2\rho^2} \left[|\Delta_1|^2 |\vec{\nabla} \phi_1 + |\Delta_2|^2 |\vec{\nabla} \phi_2 \right]^2 + \frac{|\Delta_1|^2 |\Delta_2|^2}{2\rho^2} \left[|\vec{\nabla}(\phi_1 - \phi_2)|^2 + \frac{1}{2} \left[(\vec{\nabla} |\Delta_1|)^2 + (\vec{\nabla} |\Delta_2|)^2 \right] + 2K |\Delta_1|^2 |\Delta_2|^2 [\cos(2(\phi_1 - \phi_2)) - 1] \right]$$

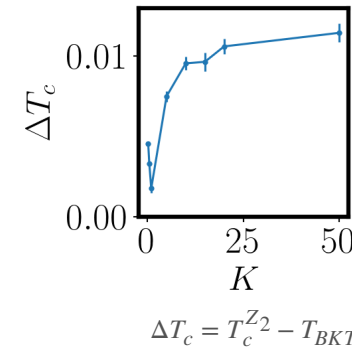
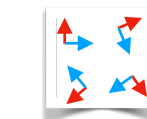
$$\rho^2 = |\Delta_1|^2 + |\Delta_2|^2 = 1$$

*We looked at the limit of very large penetration length, so we neglect the coupling to the vector potential



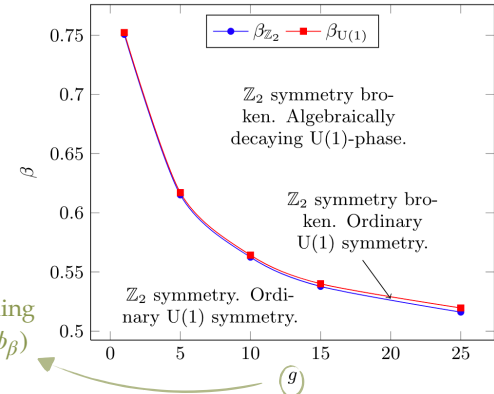
$$\langle \psi_{1,2} \rangle = 0$$

$$\langle \psi_1 \psi_2^* + \psi_1^* \psi_2 \rangle \neq 0$$

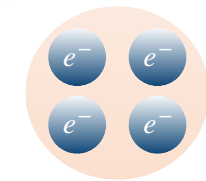


In other models this phase was found only with very strong Josephson coupling

- 2D three-band model with phase frustration



T. A. Bojesen, E. Babaev, and A. Sudbø, *Phys. Rev. B* 88, 220511 (R) (2013).



Fermionic quadrupling condensate for all finite values of K considered!!

Promising candidate to observe fermion quadruplets above T_c !

Hunting fermion quadruplets: the case of twisted bilayer graphene

I. Maccari, J. Carlström and E. Babaev, *Phys. Rev. B* 107 (6), 064501 (2023)

Large-scale Monte Carlo simulations $K > 0$

$$f = \frac{1}{2\rho^2} \left[|\Delta_1|^2 \vec{\nabla} \phi_1 + |\Delta_2|^2 \vec{\nabla} \phi_2 \right]^2 + \frac{|\Delta_1|^2 |\Delta_2|^2}{2\rho^2} \left[\vec{\nabla}(\phi_1 - \phi_2) \right]^2 + \frac{1}{2} \left[(\vec{\nabla} |\Delta_1|)^2 + (\vec{\nabla} |\Delta_2|)^2 \right] + 2K |\Delta_1|^2 |\Delta_2|^2 [\cos(2(\phi_1 - \phi_2)) - 1]$$

$$\rho^2 = |\Delta_1|^2 + |\Delta_2|^2 = 1$$

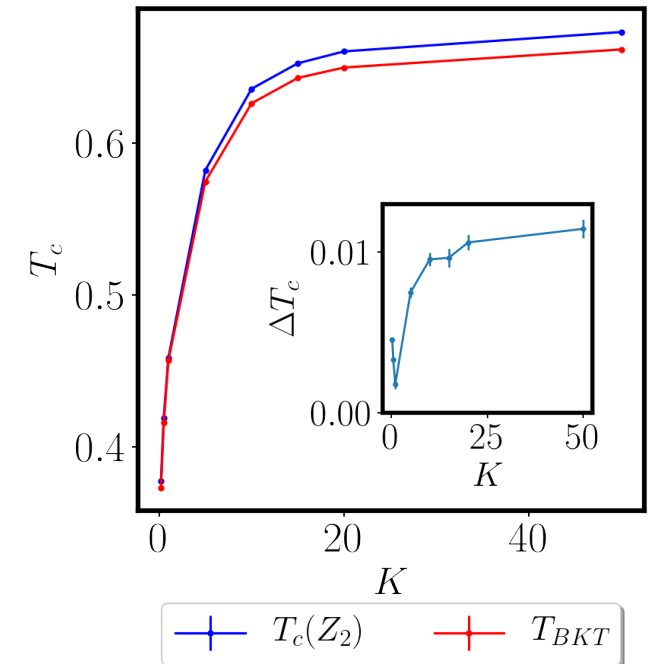
*We looked at the limit of very large penetration length, so we neglect the coupling to the vector potential

Great, but...

- Why do vortices proliferate always at lower temperatures than domain walls?
- At very small values of K , finite-size effects become prominent, are the two transition still split apart in the thermodynamic limit?
- What is the role of relative density fluctuations?

RG approaches could be crucial to shed light on these main questions

- Related, but different model previously studied via RG:
G. Bighin et al, *PRL* 123, 100601 (2019)
Coupled XY models studied with an interlayer coupling $\propto \cos(\phi_1 - \phi_2)$



Working in progress with
Benjamin Liégeois,
Chitra Ramasubramanian,
and Nicolò Defenu

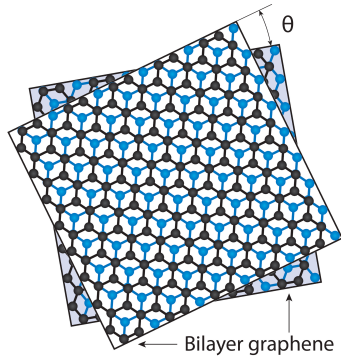
Hunting fermion quadruplets: the case of twisted bilayer graphene

I. Maccari, J. Carlström and E. Babaev, *Phys. Rev. B* 107 (6), 064501 (2023)

I. Maccari, J. Carlström and E. Babaev, *In preparation* (2024)

MATBG promising candidate to stabilize fermion quadrupling condensates

1. 2D \rightarrow more phase fluctuations
2. Single-band SC unconventional ground state (multicomponent)



Picture from:
X. Liu et al., *Nature* 583, 221 (2020)

Proposed pairing ad $n = -2$:
 $d \pm id \rightarrow (\Delta_{E_1}, \Delta_{E_2})$

Key inter-component potential term

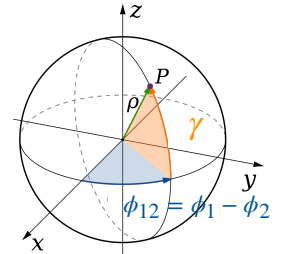
$$V(\Delta_{E_1}, \Delta_{E_2}) = 2K |\Delta_1|^2 |\Delta_2|^2 [\cos(2(\phi_1 - \phi_2)) - 1]$$

[D. V. Chichinadze, L. Classen, A. Chubokov, *PRB* 101, 224513 (2020)]

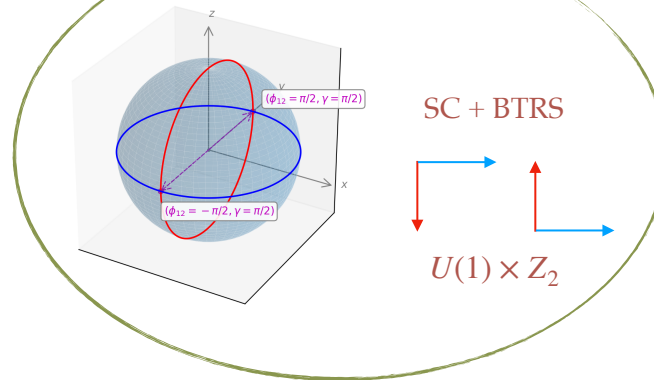
$$f = \frac{1}{2}(\vec{\nabla} \times \vec{A})^2 + \sum_{i=1,2} \left[\frac{1}{2} |(\vec{\nabla} - ie\vec{A})\Delta_i|^2 \right] + V(\Delta_1, \Delta_2); \quad V(\Delta_1, \Delta_2) = 2K |\Delta_1|^2 |\Delta_2|^2 [\cos(2(\phi_1 - \phi_2)) - 1]$$

$$\begin{cases} \Delta_1 = |\Delta_1| e^{i\phi_1}; \\ \Delta_2 = |\Delta_2| e^{i\phi_2}; \\ |\Delta_1|^2 + |\Delta_2|^2 = \rho^2. \end{cases} \rightarrow \begin{cases} \Delta_1 = |\cos(\frac{\gamma}{2})| e^{i\phi_1}; \\ \Delta_2 = |\sin(\frac{\gamma}{2})| e^{i\phi_2}; \end{cases}$$

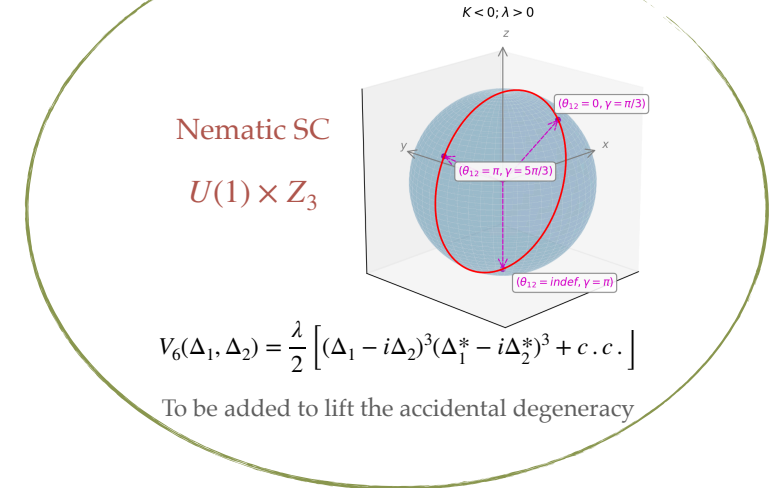
$$V(\Delta_1, \Delta_2) = \frac{K}{2} \sin^2(\gamma) [\cos(2(\phi_1 - \phi_2)) - 1]$$



$K > 0$



$K < 0$



Two complementary regions of parameter space describing a chiral ($K > 0$) and a nematic ($K < 0$) SC

Hunting fermion quadruplets: the case of twisted bilayer graphene

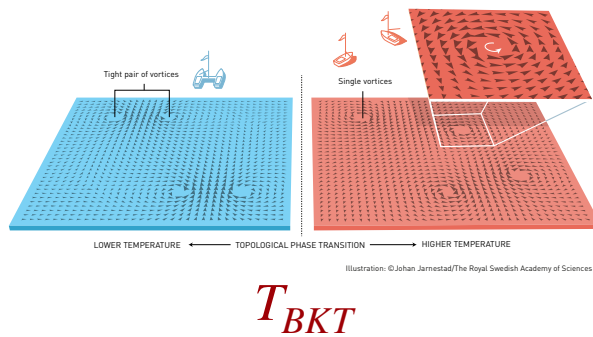
I. Maccari, J. Carlström and E. Babaev, *In preparation* (2024)

Large-scale Monte Carlo simulations $K < 0$

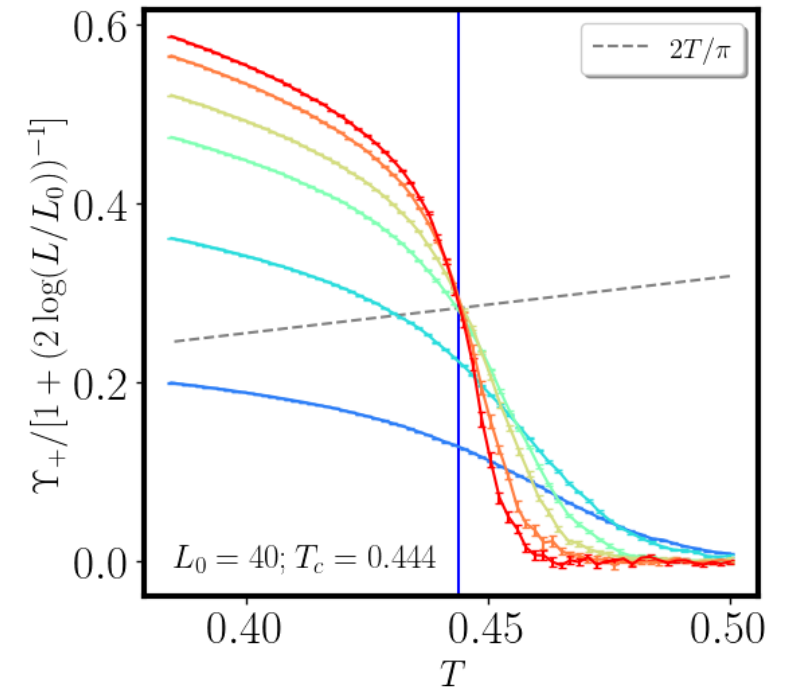
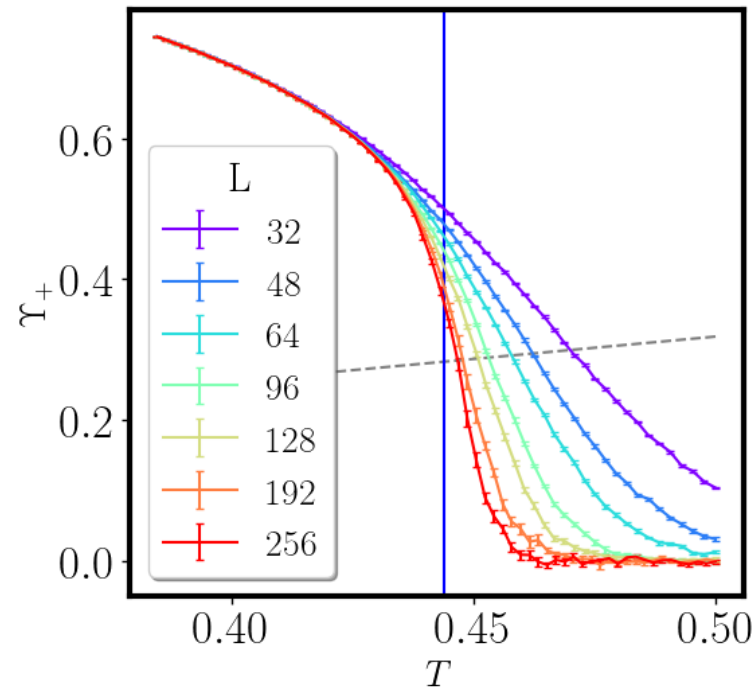
$$f = \frac{1}{2\rho^2} \left[|\Delta_1|^2 \vec{\nabla} \phi_1 + |\Delta_2|^2 \vec{\nabla} \phi_2 \right]^2 + \frac{|\Delta_1|^2 |\Delta_2|^2}{2\rho^2} \left[\vec{\nabla}(\phi_1 - \phi_2) \right]^2 + \frac{1}{2} \left[(\vec{\nabla} |\Delta_1|)^2 + (\vec{\nabla} |\Delta_2|)^2 \right] + \gamma = 2 \arctan(|\Delta_2|/|\Delta_1|)$$

$$- 2|K| |\Delta_1|^2 |\Delta_2|^2 [\cos(2(\phi_1 - \phi_2)) - 1] + \lambda [\cos(3\gamma) + 3 \cos(\gamma) \sin^2(\gamma) \sin^2((\phi_1 - \phi_2))]$$

BKT transition



$K = -1; \lambda = 1$



Hunting fermion quadruplets: the case of twisted bilayer graphene

I. Maccari, J. Carlström and E. Babaev, *In preparation* (2024)

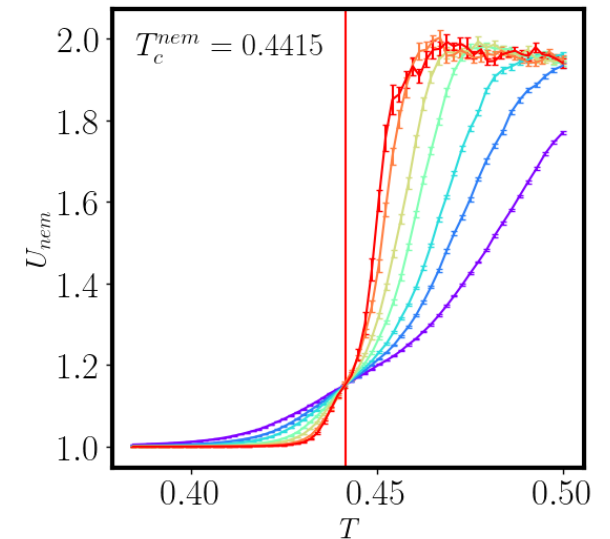
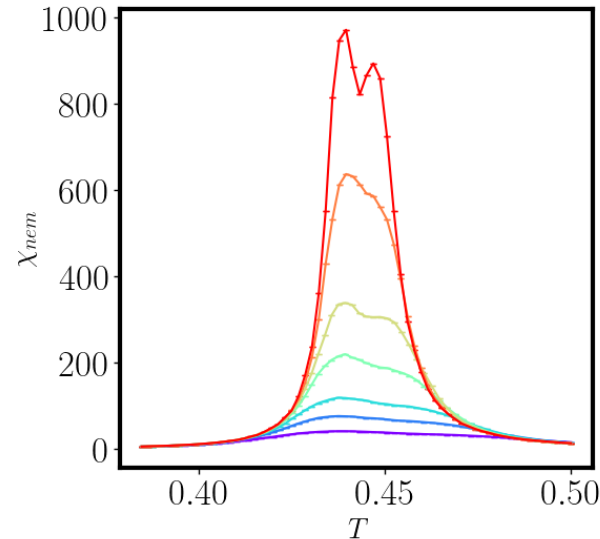
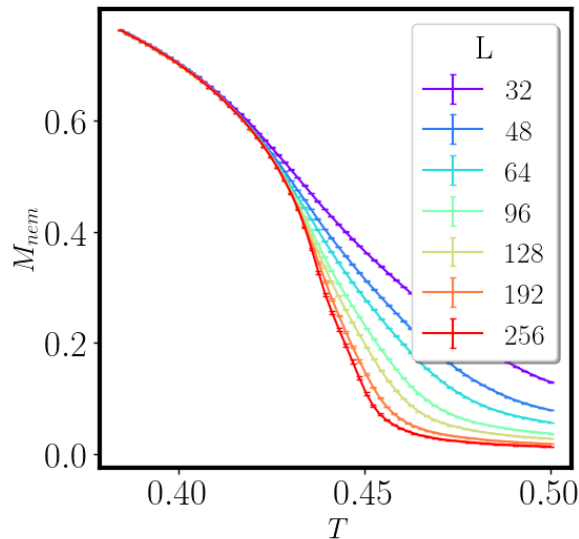
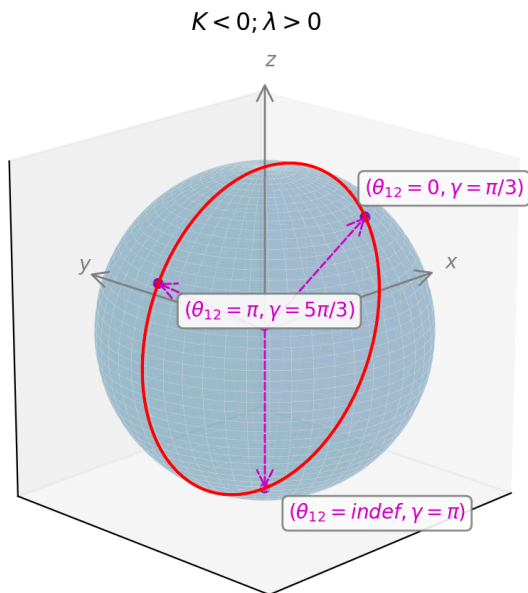
Large-scale Monte Carlo simulations $K < 0$

$$f = \frac{1}{2\rho^2} \left[|\Delta_1|^2 \vec{\nabla} \phi_1 + |\Delta_2|^2 \vec{\nabla} \phi_2 \right]^2 + \frac{|\Delta_1|^2 |\Delta_2|^2}{2\rho^2} \left[\vec{\nabla}(\phi_1 - \phi_2) \right]^2 + \frac{1}{2} \left[(\vec{\nabla} |\Delta_1|)^2 + (\vec{\nabla} |\Delta_2|)^2 \right] + \gamma = 2 \arctan(|\Delta_2|/|\Delta_1|)$$

$$- 2|K| |\Delta_1|^2 |\Delta_2|^2 [\cos(2(\phi_1 - \phi_2)) - 1] + \lambda [\cos(3\gamma) + 3 \cos(\gamma) \sin^2(\gamma) \sin^2((\phi_1 - \phi_2))]$$

Nematic Z_3 phase transition

$$M_{nematic}^2 = \frac{1}{N^2} \left[\left(\sum_i \sin(\gamma_i) \cos(\theta_{12,i}) \right)^2 + \left(\sum_i \sin(\gamma_i) \sin(\theta_{12,i}) \right)^2 + \left(\sum_i \cos(\gamma_i) \right)^2 \right]$$



Hunting fermion quadruplets: the case of twisted bilayer graphene

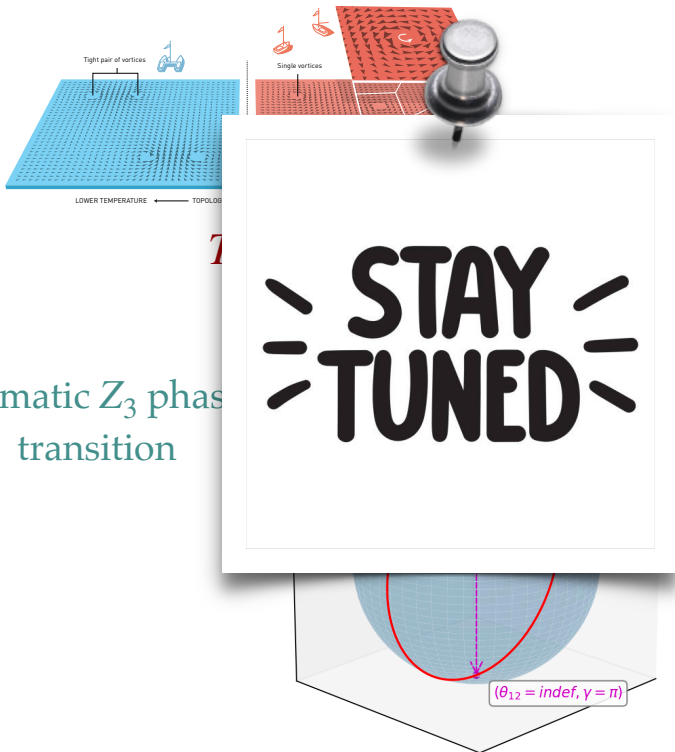
I. Maccari, J. Carlström and E. Babaev, *In preparation* (2024)

Large-scale Monte Carlo simulations $K < 0$

$$f = \frac{1}{2\rho^2} \left[|\Delta_1|^2 \vec{\nabla} \phi_1 + |\Delta_2|^2 \vec{\nabla} \phi_2 \right]^2 + \frac{|\Delta_1|^2 |\Delta_2|^2}{2\rho^2} \left[\vec{\nabla}(\phi_1 - \phi_2) \right]^2 + \frac{1}{2} \left[(\vec{\nabla} |\Delta_1|)^2 + (\vec{\nabla} |\Delta_2|)^2 \right] +$$

$$-2|K| |\Delta_1|^2 |\Delta_2|^2 \left[\cos(2(\phi_1 - \phi_2)) - 1 \right] + \lambda \left[\cos(3\gamma) + 3 \cos(\gamma) \sin^2(\gamma) \sin^2((\phi_1 - \phi_2)) \right]$$

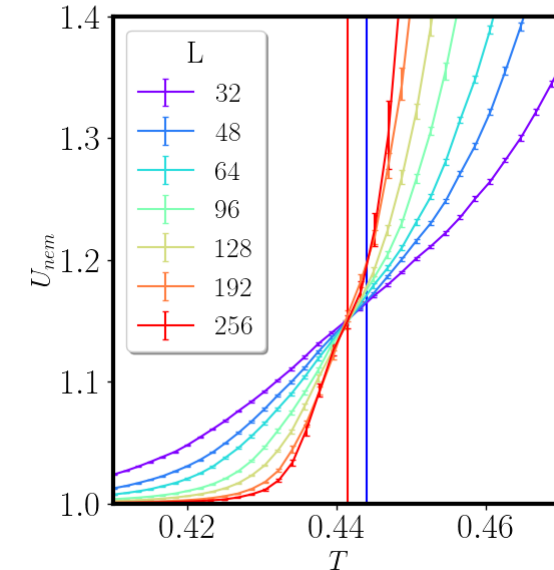
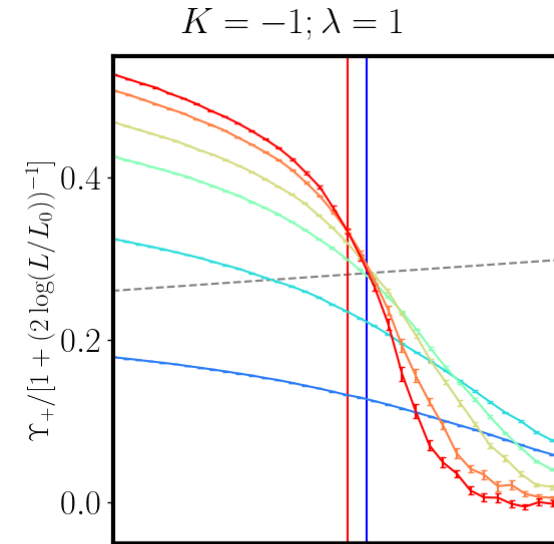
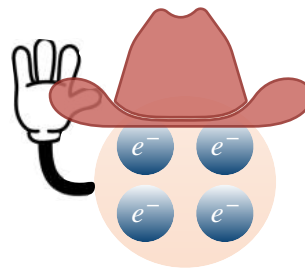
BKT transition



These preliminary results seem to suggest:

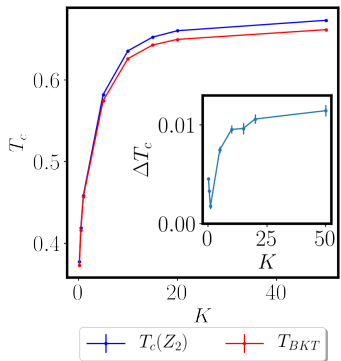
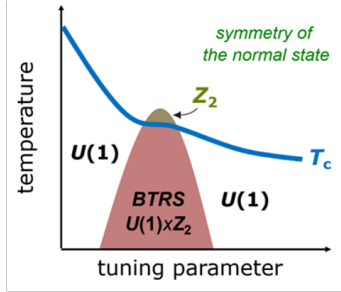
$$T_{BKT} > T_c^{Z_3}$$

Charge-4e SC phase?



Conclusions and future directions

non-SC fermion quadruplets



- Fermion quadrupling condensates are novel fascinating states of matter so far observed only in two real materials.
 - ★ V. Grinenko, D. Weston, F. Caglieris, C. Wuttke, C. Hess, T. Gottschall, **I. Maccari**, et al., *Nature Physics* **17**, 1254–1259 (2021)
 - ★ I. Maccari and E. Babaev, *Phys. Rev. B* **105**, 214520 (2022)
 - ★ I. Shipulin, N. Stegani, **I. Maccari**, et al., *Nature Communications* **14**, 6734 (2023)
- Twisted-bilayer graphene may be a promising candidate for the observation of fermion quadruplets above T_c
 - ★ **I. Maccari**, J. Carlström and E. Babaev, *Phys. Rev. B* **107** (6), 064501 (2023)
 - ★ **I. Maccari**, J. Carlström and E. Babaev, *In preparation* (2024)

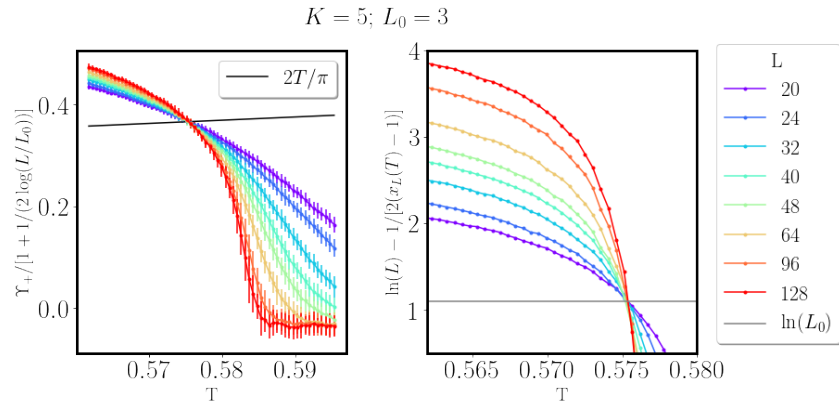
- ➔ How to engineer materials in such a way as to observe higher order fermionic condensates: what symmetries are important? what interactions?
- ➔ How to manipulate the nucleation of specific topological excitations?
- ➔ What are the new emergent properties of these states?

Thank you for your attention!

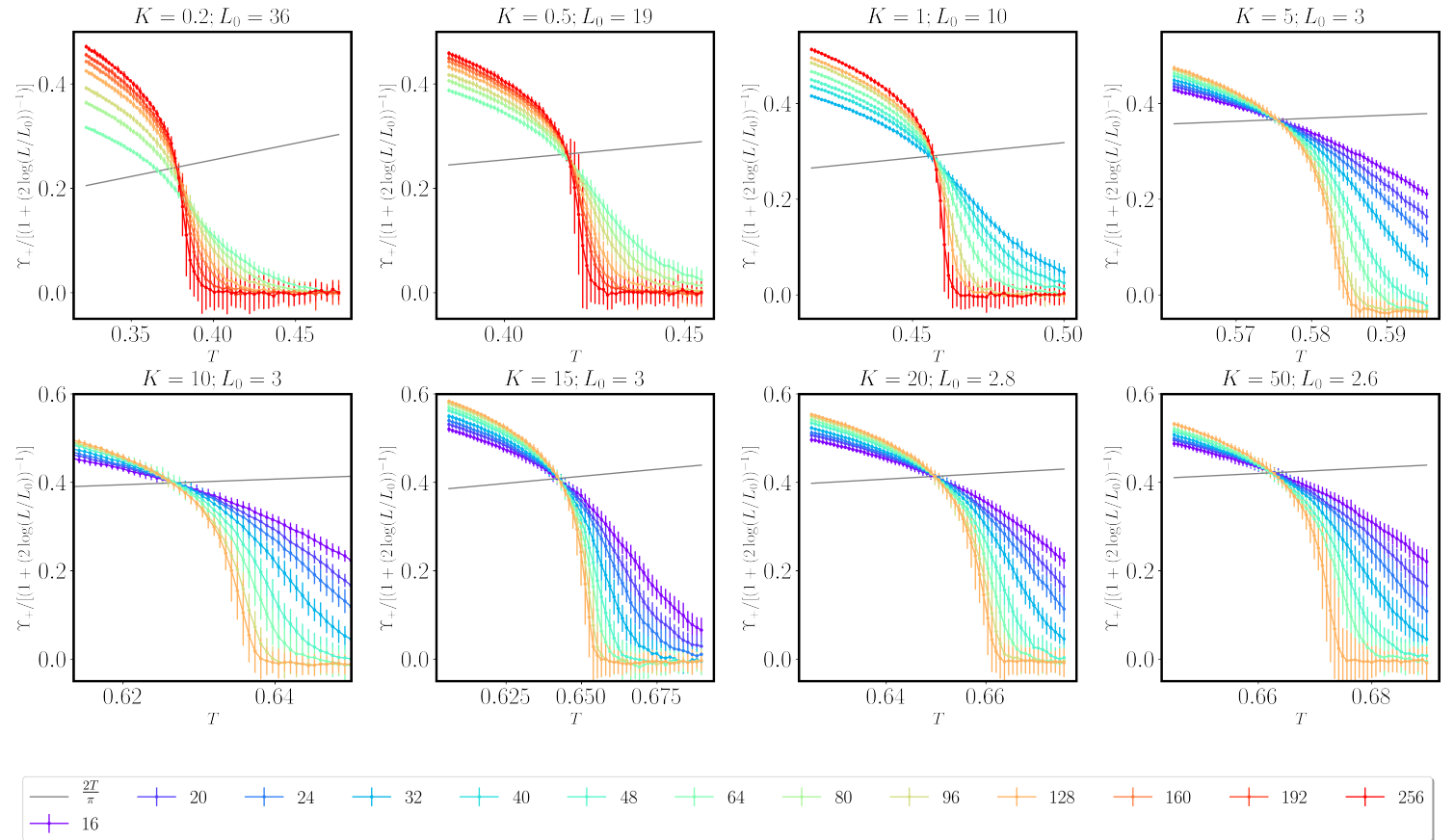
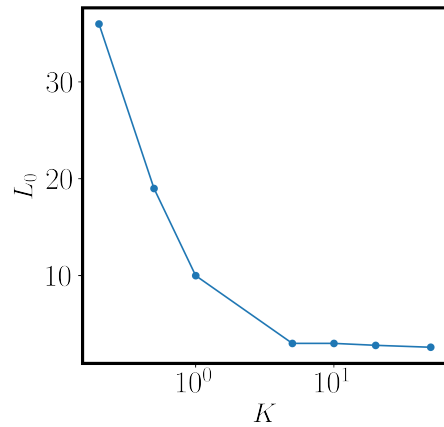


Backup slides

I. Maccari, J. Carlström and E. Babaev, *Phys. Rev. B* 107 (6), 064501 (2023)



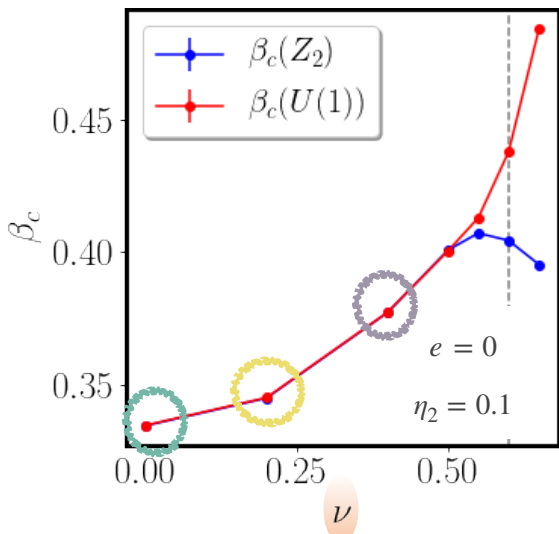
$$\gamma_+(\infty, T_{BKT}) = \frac{\gamma_+(L, T_{BKT})}{1 + (2 \log(L/L_0))^{-1}}$$



Backup slides

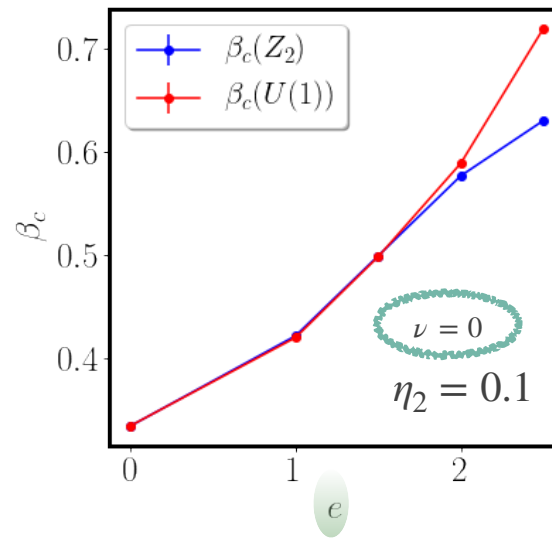
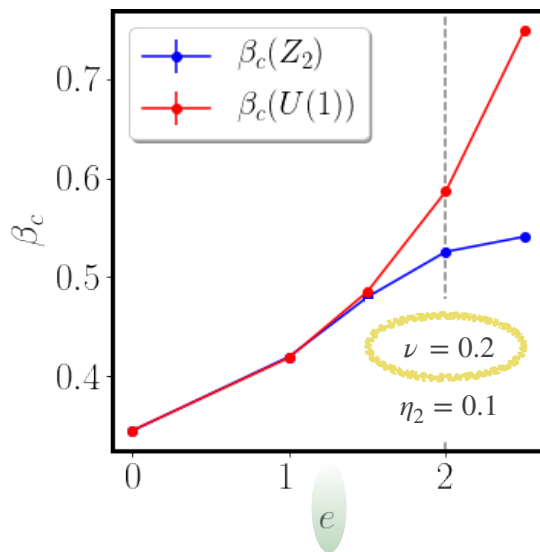
I. Maccari and E. Babaev *Phys. Rev. B* 105, 214520 (2022)

$$\rho_1 = \rho_2 = 1 \quad f = \frac{1-\nu}{4} (\nabla\phi_1 + \nabla\phi_2 - 2e\mathbf{A})^2 + \frac{1+\nu}{4} (\nabla\phi_1 - \nabla\phi_2)^2 + \eta_2 \cos[2(\phi_1 - \phi_2)] + \frac{1}{2}(\nabla \times \mathbf{A})^2$$

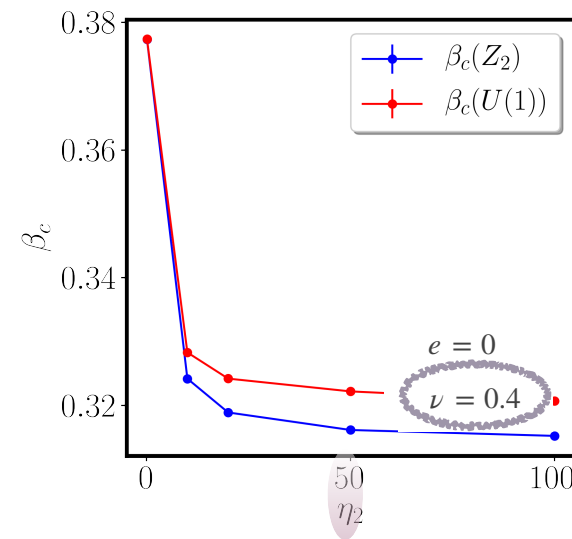


It decreases the relative cost of phase sum fluctuations

It decreases the cost of (1,1) vortices

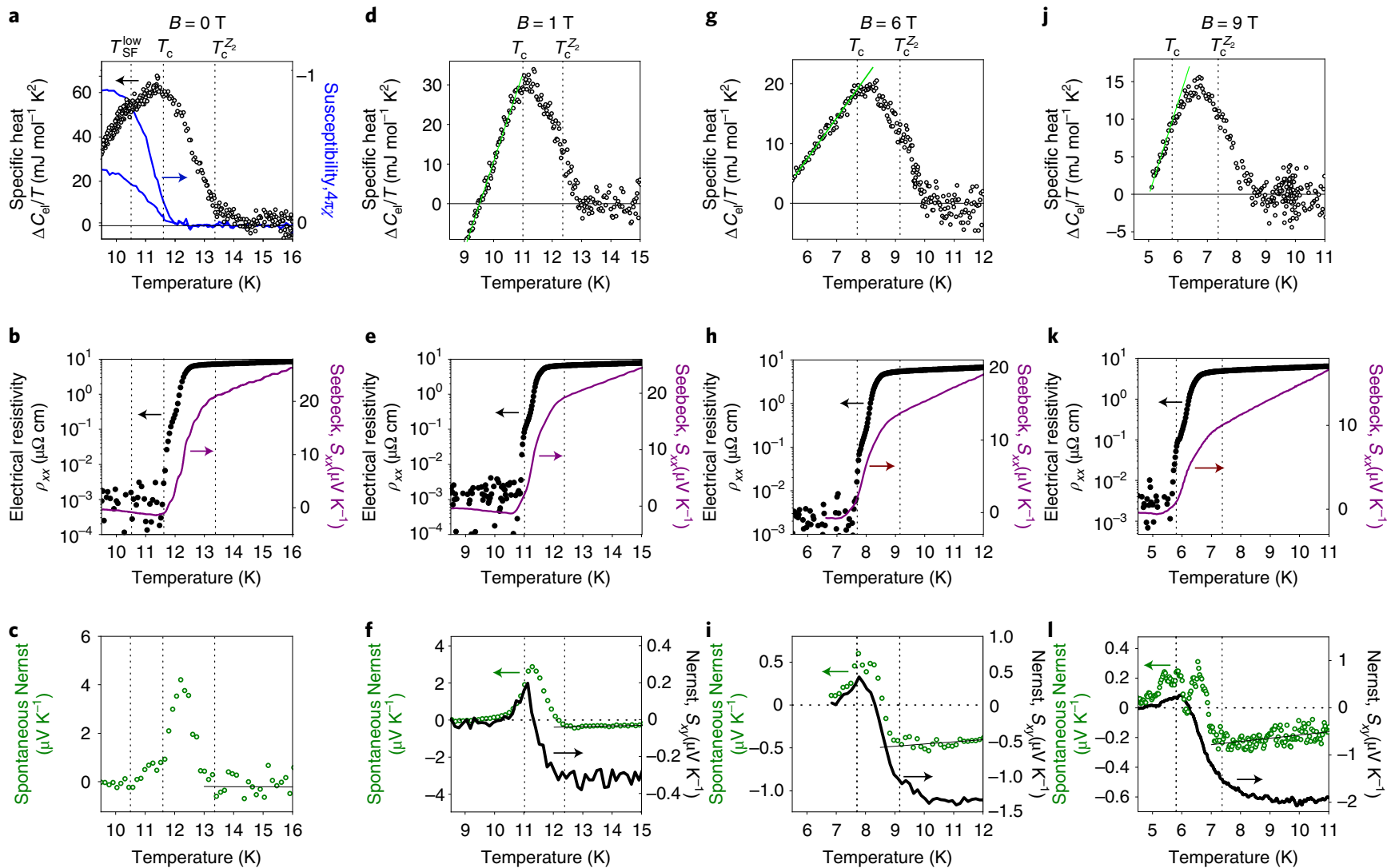


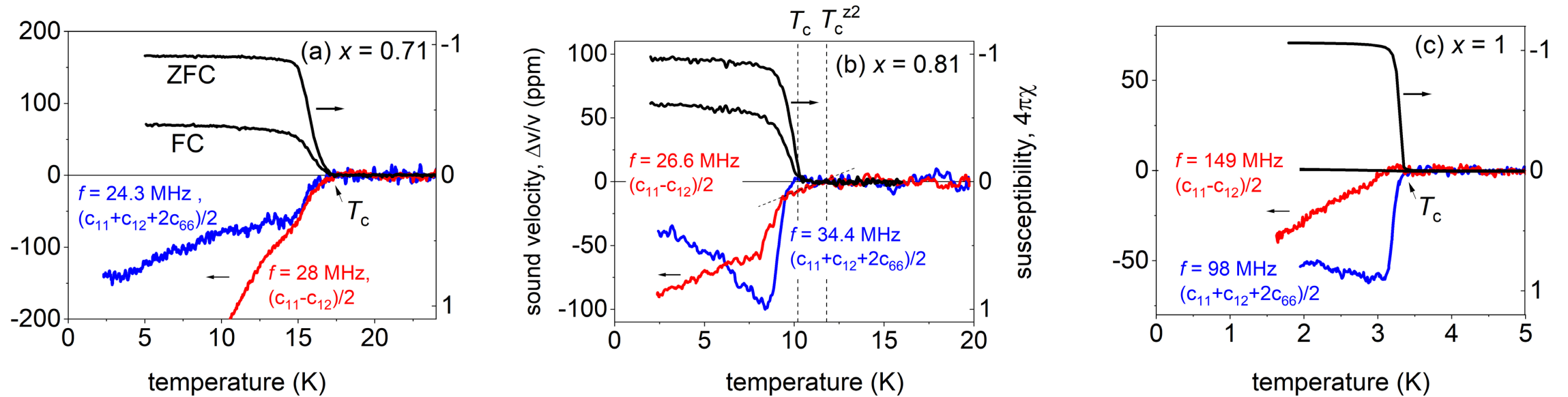
It increases the cost of domain walls



Backup slides

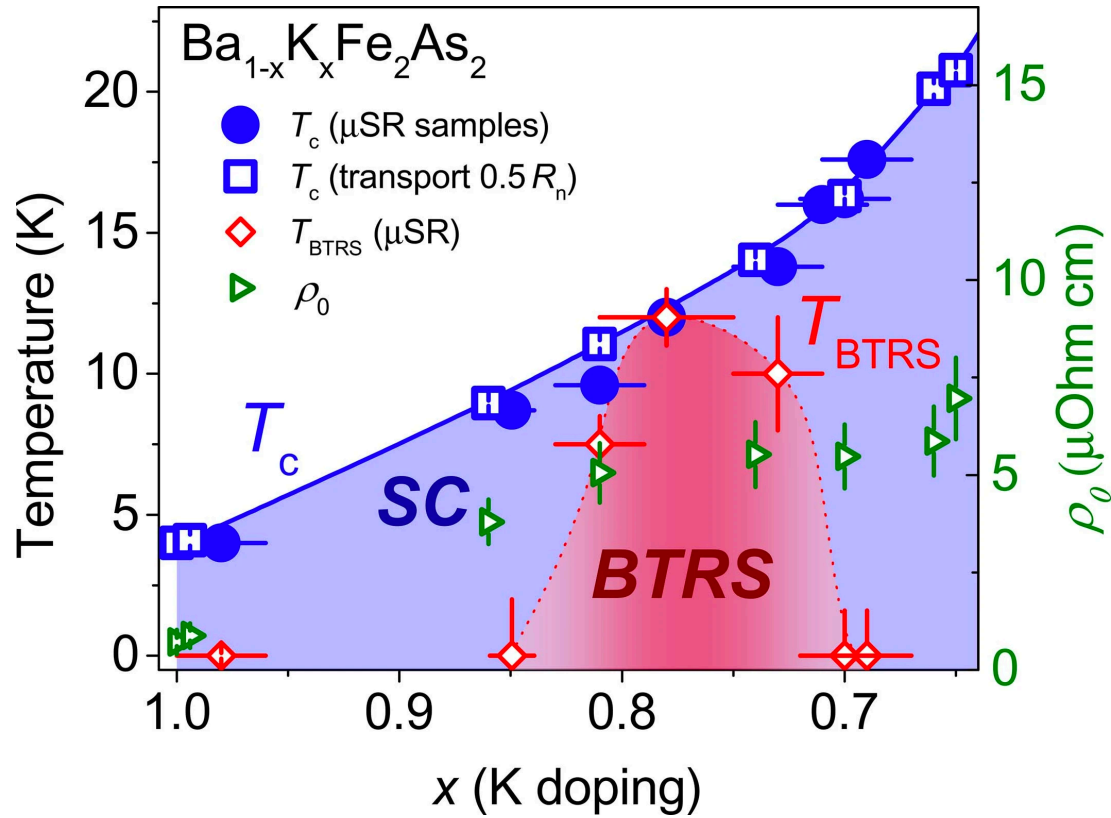
V. Grinenko, et al., *Nat. Phys.* 17, 1254 - 1259 (2021).





Extended Data Fig. 6 | Ultrasound data. Temperature dependence of the relative change of the sound velocity (left) for the longitudinal $(c_{11} + c_{12} + 2c_{66})/2$ and transverse $(c_{11} - c_{12})/2$ acoustic modes with subtracted background (see Fig. S3 in the supplementary information) compared with temperature dependence of the magnetic susceptibility in $B||ab = 0.5$ mT (right) measured after cooling in zero magnetic field (ZFC) and cooling in the applied field (FC). The data for the samples with $x = 0.71$, 0.81 , and 1 are shown in panels (a), (b), and (c), respectively. The sample with $x = 0.81$ exhibits a kink in the velocity of the transverse acoustic mode at a position which agrees with the onset of the broad feature in the specific heat shown in Extended Data Fig. 8 for the same sample. The position of the kink agrees with the position of the Z_2 transition indicated by the thermal transport experiments for the sample with $x \approx 0.8$ (Fig. 2 in the main text).

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



The residual resistivity ρ_0 very weakly dependent on doping

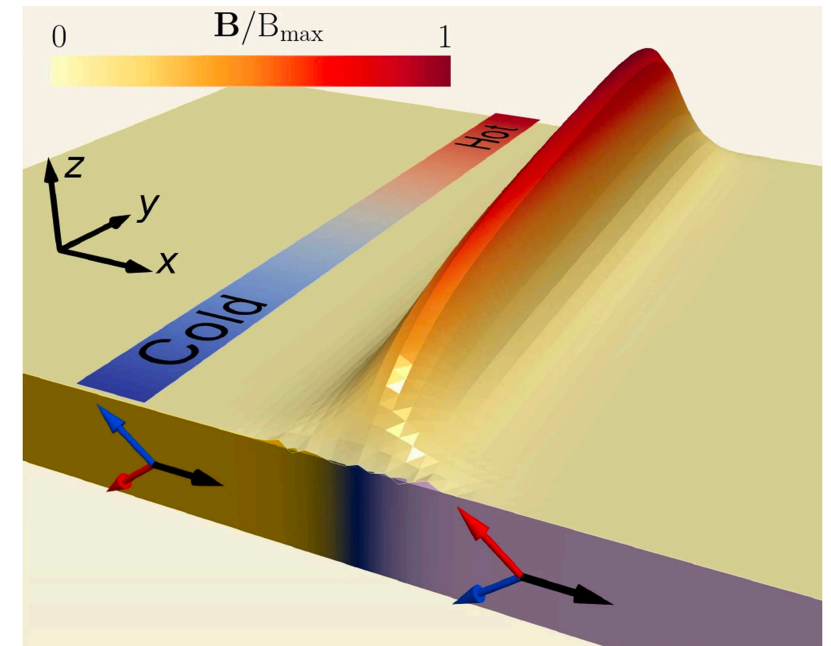
$$\rho(T) = \rho_0 + A_{FL}T^2$$

$$F = \frac{\mathbf{J}^2}{2e^2\varrho^2} + \frac{1}{2}\mathbf{B}^2 + \sum_i \frac{1}{2}(\nabla|\psi_i|)^2 + a_i|\psi_i|^2 + \frac{b_i}{2}|\psi_i|^4 + \sum_{i<j} \frac{|\psi_i|^2|\psi_j|^2}{\rho^2} \left(\frac{[\nabla(\phi_i - \phi_j)]^2}{2} + \frac{\eta_{ij}\rho^2 \cos(\phi_i - \phi_j)}{|\psi_i||\psi_j|} \right).$$

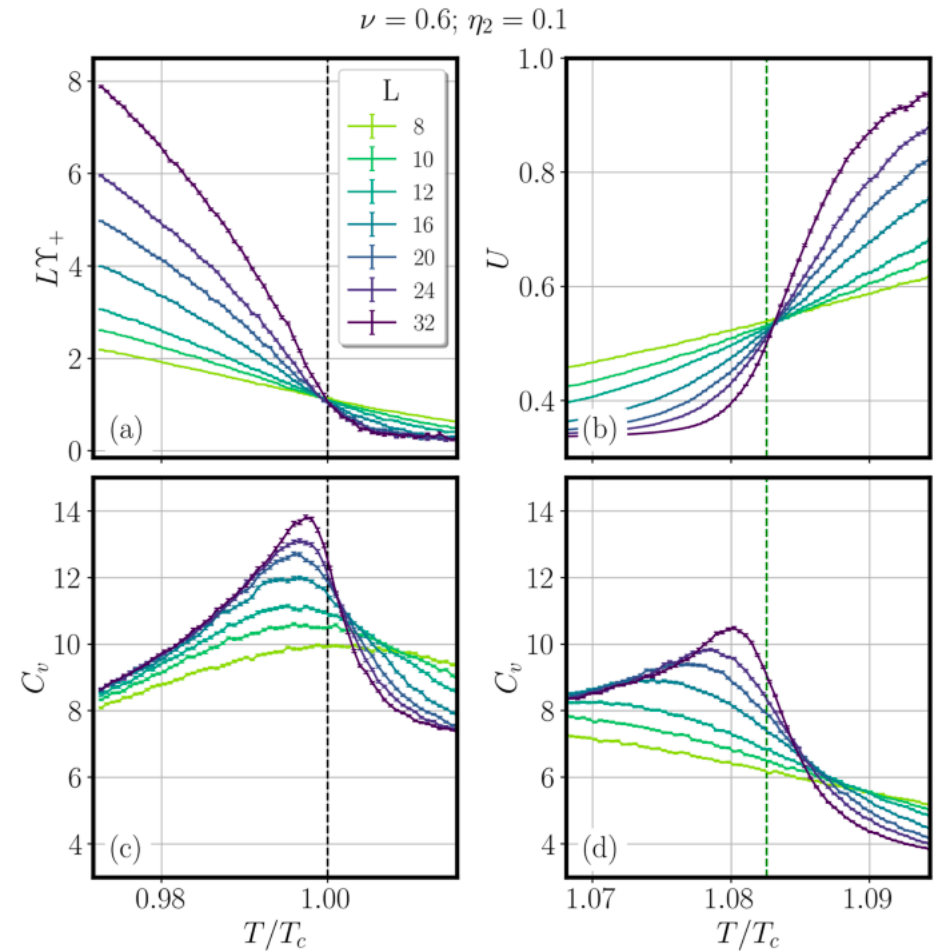
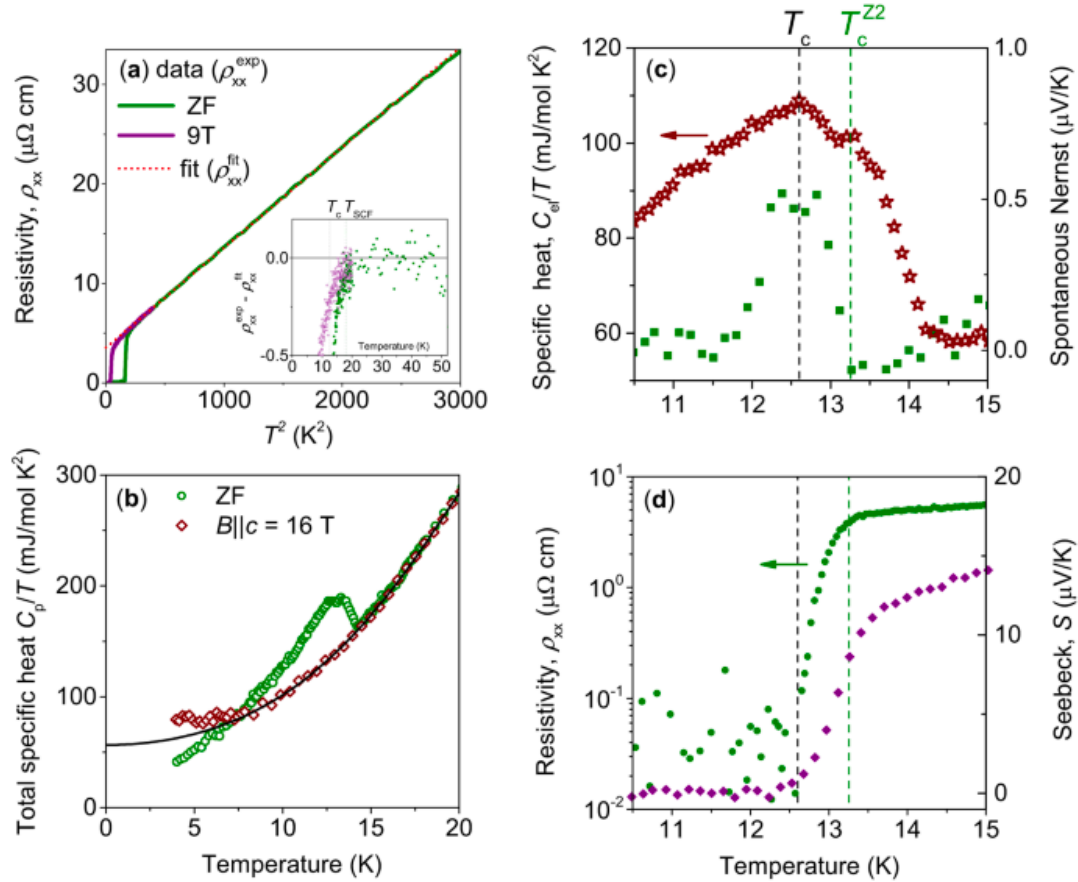
$$\mathbf{J} \equiv \sum_i e \operatorname{Im}(\psi_i^* D\psi_i)$$

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

Spontaneous Magnetic Fields

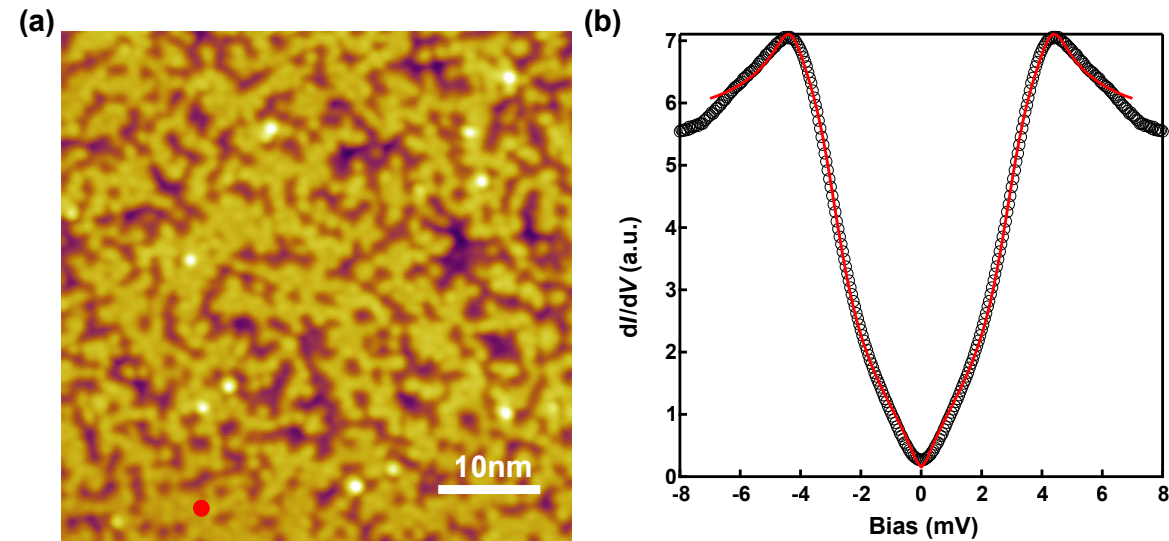


I. Shipulin, N. Stegani, IM et al. *Nat. Comm.* 14, 6734 (2023)



No CDW evidences from STM data

$T = 300 \text{ mK}$

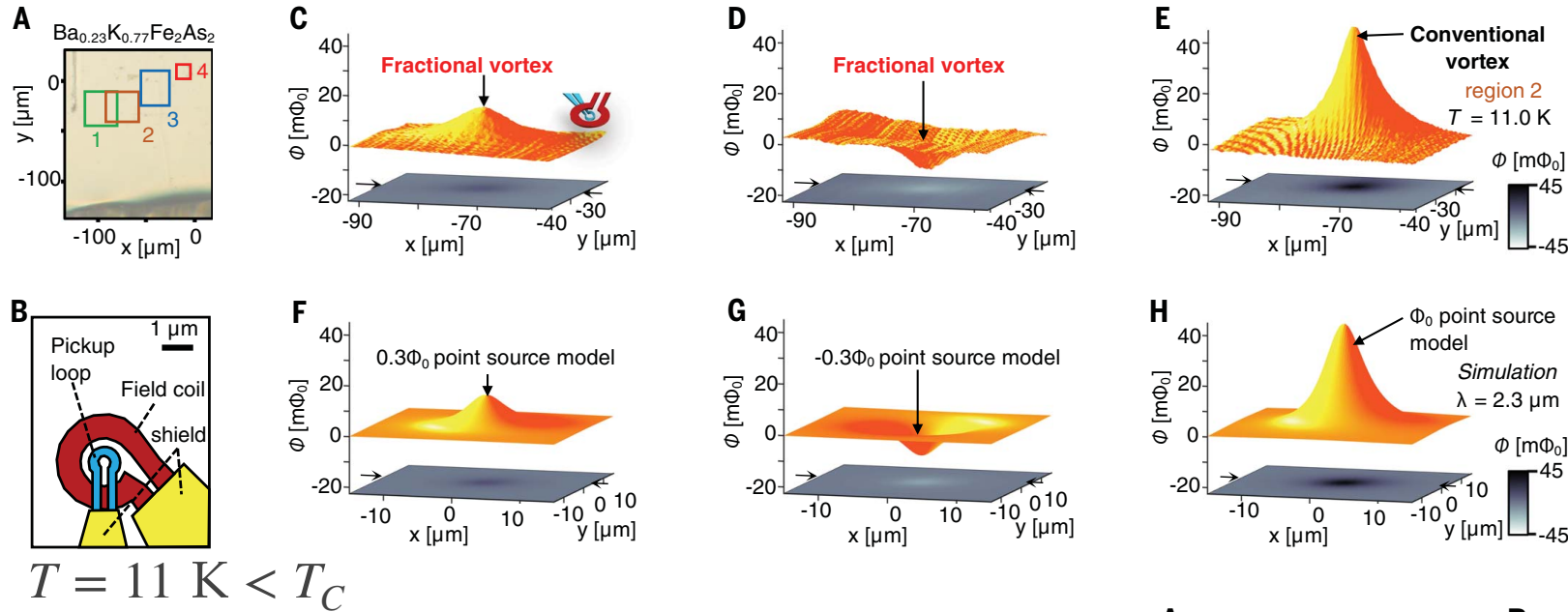


I. Shipulin, N. Stegani, IM et al.
Nat. Comm. 14, 6734 (2023)

Figure S3. STM data for the sample S_{NP} (a) STM Topograph recorded from the disordered surface of a $Ba_{1-x}K_xFe_2As_2$ sample with $x = 0.77$ (image size: $(50 \times 50)nm^2$, $V_b = 80mV$, $I_t = 100pA$). The disordered surface was likely caused by the mixture of Ba atoms and K atoms.(b) $dI/dV - V$ spectrum taken from a defect-free position (the red point) on the surface shown in (a) (Spectroscopic set-point: $V_s = 10mV$, $I_s = 200pA$, amplitude of bias modulation used $V_{mod} = 0.25mV$), showing a "V"-shaped superconducting gap. The black circle was experiment result. Red solid curve was fitting result using double-gap Dynes equation with a larger gap ($\Delta_2 = 3.1meV$) and a smaller gap with node($\Delta_1 = 2.1meV$). There is no "CDW" gap feature in the spectrum.

SC vortices carrying a temperature-dependent fractional quantum flux

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



Observation of vortices carrying fractional quantum fluxes

$$\oint \vec{J}_s \vec{dl} = -e\rho^2 \oint \left(\hbar \frac{|\psi_1|^2}{\rho^2} \vec{\nabla} \phi_1 - \frac{2e}{c} \vec{A} \right) \vec{dl} = 0$$

$$\hbar \frac{|\psi_1|^2}{\rho^2} 2\pi N = \frac{2e}{c} \Phi \implies \Phi = \Phi_0 \frac{|\psi_1|^2}{\rho^2} N$$

The fractional quantum flux shows a temperature dependence, that cannot be explained just in terms of $\lambda_L(T)$. Decreasing the temperature also the fractional quantum flux decreases.

