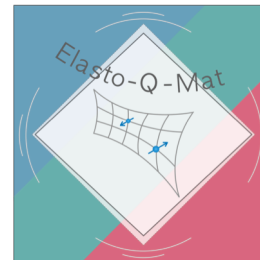


# Strain manipulation of multi-component order parameters

Jörg Schmalian  
Karlsruhe Institute of Technology

**DFG** Deutsche  
Forschungsgemeinschaft  
German Research Foundation

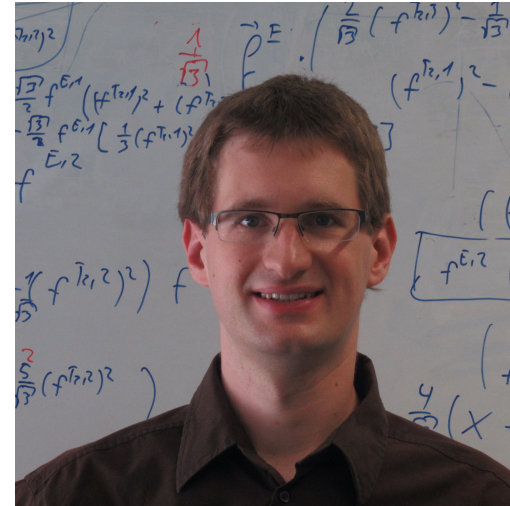
CRC– TRR 288



# collaborators



**Matthias Hecker (KIT)**



**Roland Willa (KIT)**



**Rafael M. Fernandes (Minnesota)**



**Rolf Lortz (Hong-Kong UST)**

- brief recap: vestigial order in iron-based systems
- vestigial order in doped  $\text{Bi}_2\text{Se}_3$   
with Matthias Hecker (theory) + Rolf Lortz (experiment)

Matthias Hecker and J.S., npj Quantum Mater. **3**, 26 (2018)

C.-w. Cho, et al., Nature Comm. **11**, 3056 (2020)

- proposal for (non) TRS-breaking in  $\text{Sr}_2\text{RuO}_4$   
with Roland Willa and Rafael Fernandes

unpublished

# Landau theory of phase transitions: 1937



Lev D. Landau  
(1908-1968)

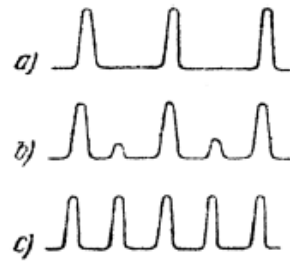


Fig. 1

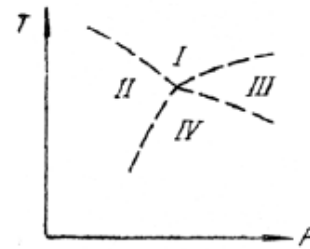


Fig. 3

$$\delta\rho(\mathbf{r}) = \sum_n \sum_{\mu} \eta_{\mu}^{(n)} \varphi_{\mu}^{(n)}(\mathbf{r})$$

- expand w.r.t. the irreducible representations of the symmetry group
- Schur's orthogonality relations  $\rightarrow$

$$f = \sum_n r_n \sum_{\mu} \eta_{\mu}^{(n)} \eta_{\mu}^{(n)} + \dots$$

# symmetry breaking fluctuations:

fluctuating field  $\eta_\mu$  with multiple components  $\mu = 1, \dots, d_{\text{irr}}$

$$\varphi = \sum_{\mu, \nu} r_{\mu\nu} \eta_\mu^* \eta_\nu$$

$$\varphi = \sum_{\mu, \nu, \lambda} r_{\mu\nu\lambda} \eta_\mu \eta_\nu \eta_\lambda$$

composite order of a fluctuating variable can break a symmetry

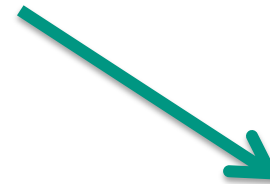
$$\langle \varphi \rangle \neq 0$$

# fluctuations or frustration



long range order is suppressed

$$\langle \eta_\mu \rangle = 0$$



a liquid of spins,  
of Cooper pairs etc.  
forms

composite order  
(condensation of fluctuations)

$$\langle \eta_\mu \eta_\nu \rangle \neq 0 \quad \langle \eta_\mu \eta_\nu \eta_\lambda \rangle \neq 0$$

symmetry: must be a nontrivial combination  
energetics: less fluctuations or frustration

**vestigial order**

## Ising Transition in Frustrated Heisenberg Models

P. Chandra

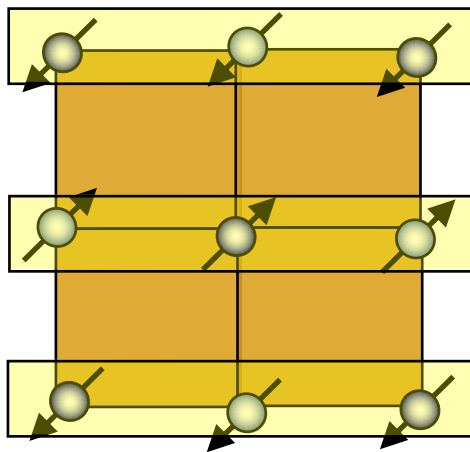
*Corporate Research Science Laboratories, Exxon Research and Engineering Company, Annandale, New Jersey 08801*

P. Coleman and A. I. Larkin<sup>(a)</sup>

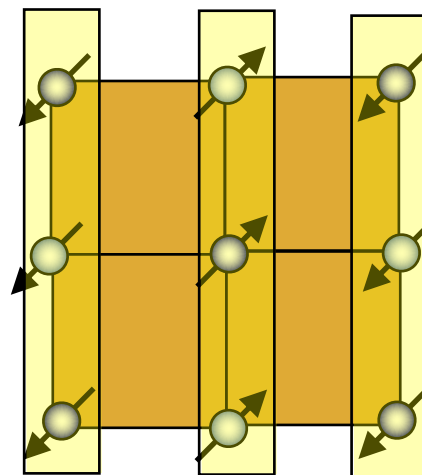
*Serin Physics Laboratory, Rutgers University, P.O. Box 849, Piscataway, New Jersey 08854*

(Received 5 June 1989)

### $J_1$ - $J_2$ Heisenberg model



$$\varphi < 0$$



$$\varphi > 0$$

additional  $Z_2$  degeneracy

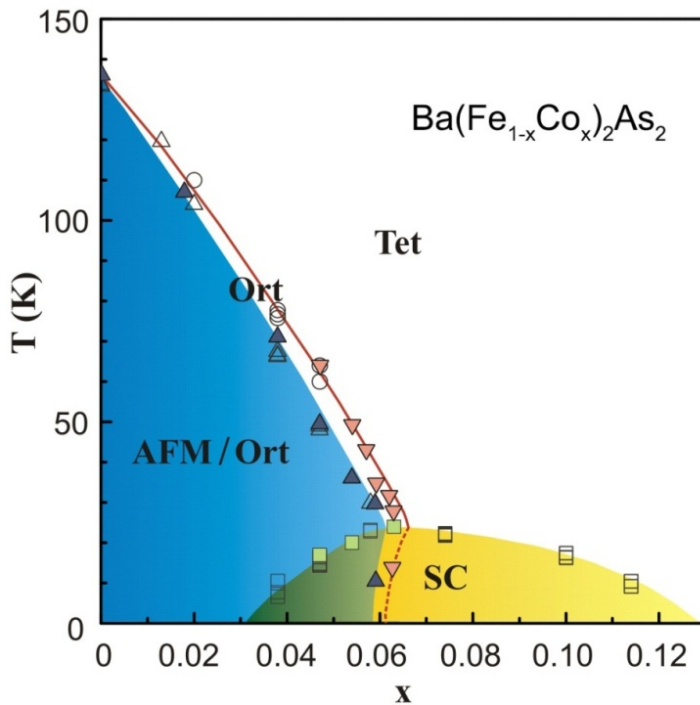
$$\mathbf{S}(\mathbf{r}) = \mathbf{S}_x(\mathbf{r}) e^{i\mathbf{Q}_x \cdot \mathbf{r}} + \mathbf{S}_y(\mathbf{r}) e^{i\mathbf{Q}_y \cdot \mathbf{r}}$$

emergent Ising order parameter

$$\varphi = \langle \mathbf{S}_x^2 - \mathbf{S}_y^2 \rangle$$

# vestigial nematicity in iron-based systems

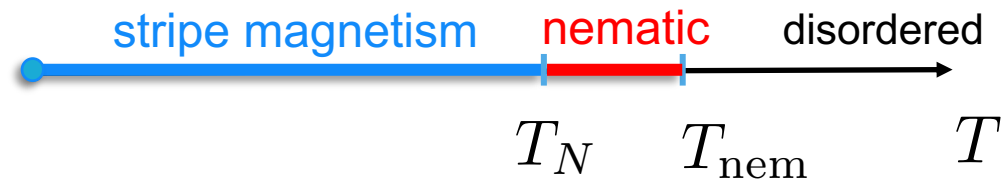
time-reversal symmetry and rotational symmetry are separately broken



C. Xu, M. Müller, and S. Sachdev, PRB **78**, 020501(R) (2008)

C. Fang, H. Yao, W.-F. Tsai, J. P. Hu, and S. A. Kivelson, PRB **77**, 224509 (2008)

$$\varphi = \langle \mathbf{S}_x^2 - \mathbf{S}_y^2 \rangle$$



Ni et al, PRB (2008), Fernandes et al, PRB (2010)



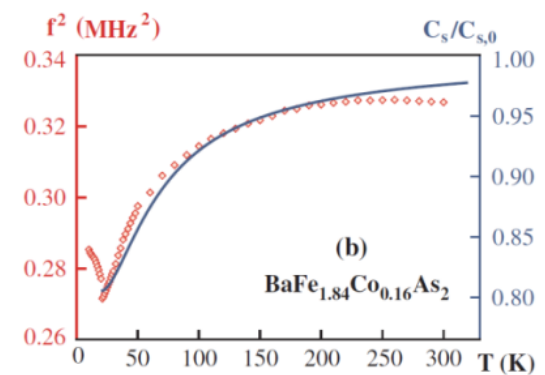
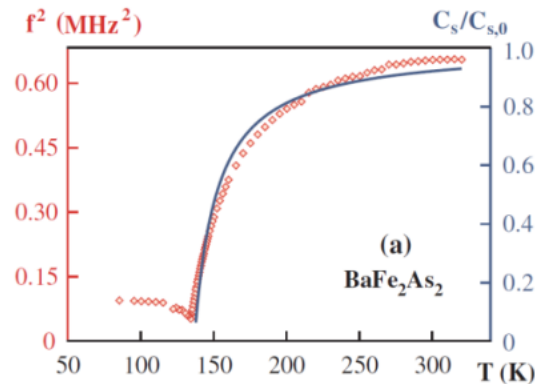
# nematic susceptibility: elastic constants

coupling to the lattice  $H_{mag.-elast.} = -\lambda \int_x \varepsilon_{66}(x) \varphi(x)$

renormalization of the elastic constants

$$C_{66}^{-1} = C_{66,0}^{-1} + \lambda^2 C_{66,0}^{-2} \chi_{\text{nem}}$$

elastic constants  
measure  
the static nematic  
susceptibility

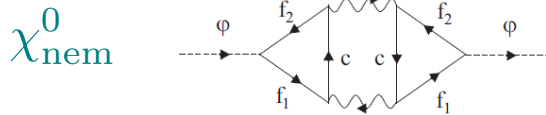


# nematic dynamics: Raman scattering

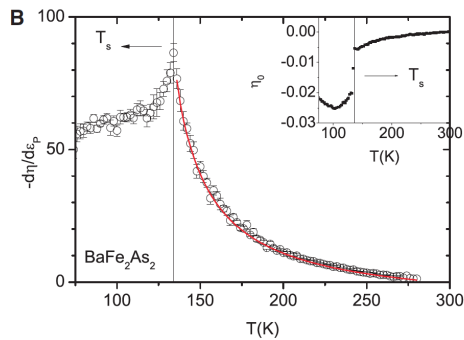
$$\chi_{\text{nem}}(q, \omega) \propto \left\langle \rho_{q, \omega}^{B_{1g}} \rho_{-q, -\omega}^{B_{1g}} \right\rangle$$

Y. Gallais et al. Phys. Rev. Lett. **111**, 267001 (2013)

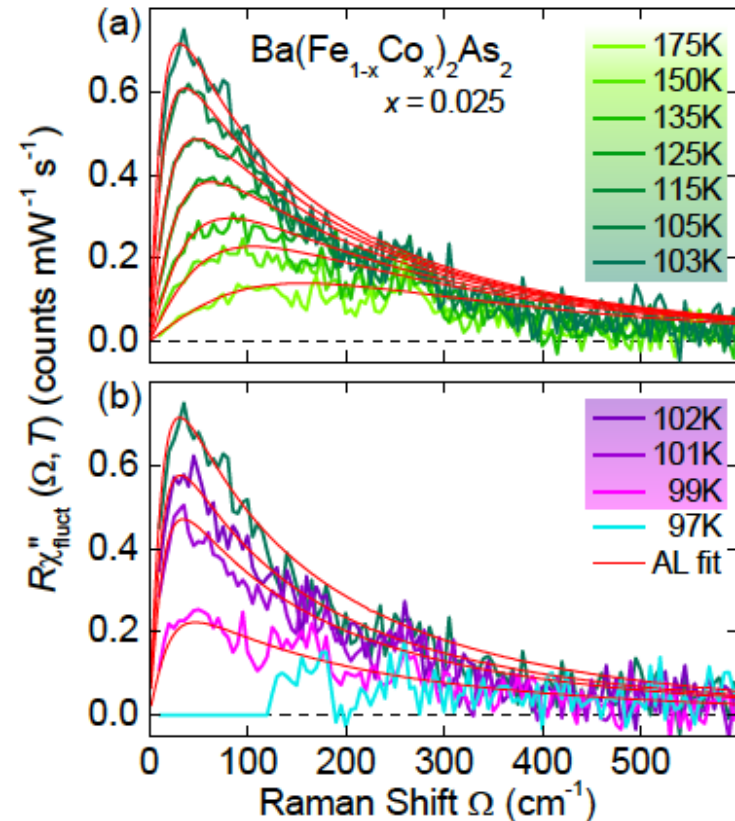
$$\chi_{\text{nem}} = \frac{\chi_{\text{nem}}^0}{1 - g\chi_{\text{nem}}^0}$$



low-frequency slope = elasto-resistivity



J.H. Chu, H.-H. Kuo, J. G. Analytis, I. R. Fisher  
Science **337**, 710 (2012)



F. Kretzschmar et al.  
Nature Physics **12**, 560 (2016)

# spin-driven composite order?

using NMR to probe the origin of lattice softening

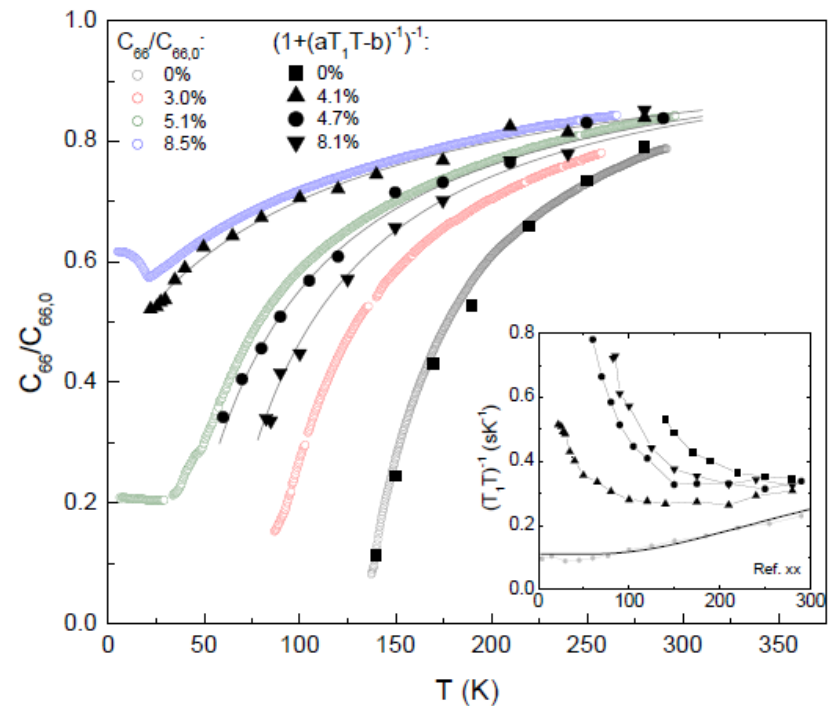
$$\varphi = \langle \mathbf{S}_x^2 - \mathbf{S}_y^2 \rangle$$

$$\frac{1}{T_1 T} \iff \chi_{\text{nem}} \iff C_{66}$$

assume overdamped spin response

$$\chi_{\text{spin}}(q, \omega) \propto \frac{1}{\epsilon(q) - i\omega/\Gamma}$$

D. S. Inosov, et al. Nature Phys. **6**, 178 (2010)



# primary and vestigial order

order parameter expansion  $\delta f = a(T) \sum_{\mu=1}^{n_{\text{irr}}} \eta_{\mu}^* \eta_{\mu} + \dots$

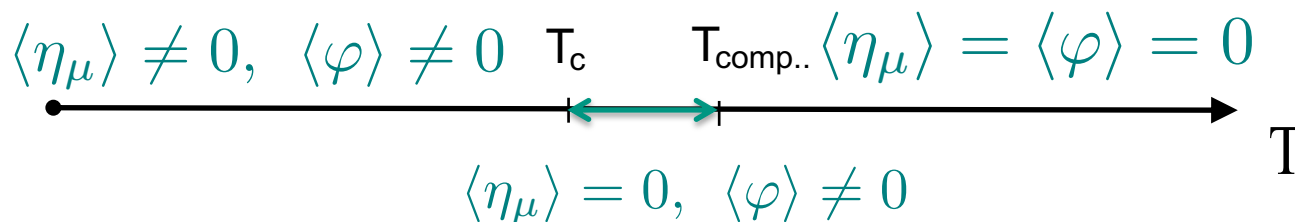
form bilinear combinations  
that transform non-trivially

$$\varphi = \sum_{\mu\nu} r_{\mu\nu} \eta_{\mu}^* \eta_{\nu}$$

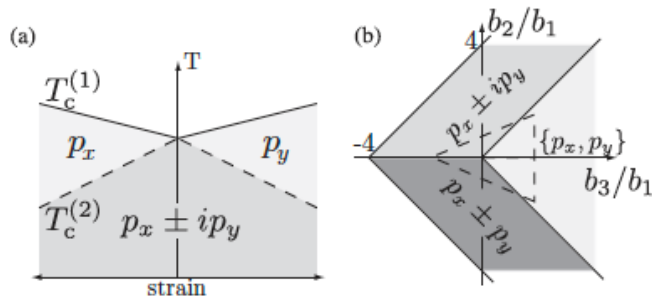
$$\Gamma^* \otimes \Gamma = E \oplus \Gamma_1 \oplus \dots \oplus \Gamma_n$$

nontrivial bilinear  
forms  
=  
intertwined  
symmetry breakings

## order made from fluctuations



# Vestigial phases in multi-component superconductors



M. H. Fischer and Erez Berg PRB **93**, 054501 (2016)

**charge-4e  
superconductivity**

E. Berg, E. Fradkin, and S.A. Kivelson, Nat. Phys. **5**, 830 (2009)

D. F. Agterberg and H. Tsunetsugu, Nat. Phys. **4**, 639 (2008)

L. Radzihovsky and A. Vishwanath, PRL **103**, 010404 (2009)

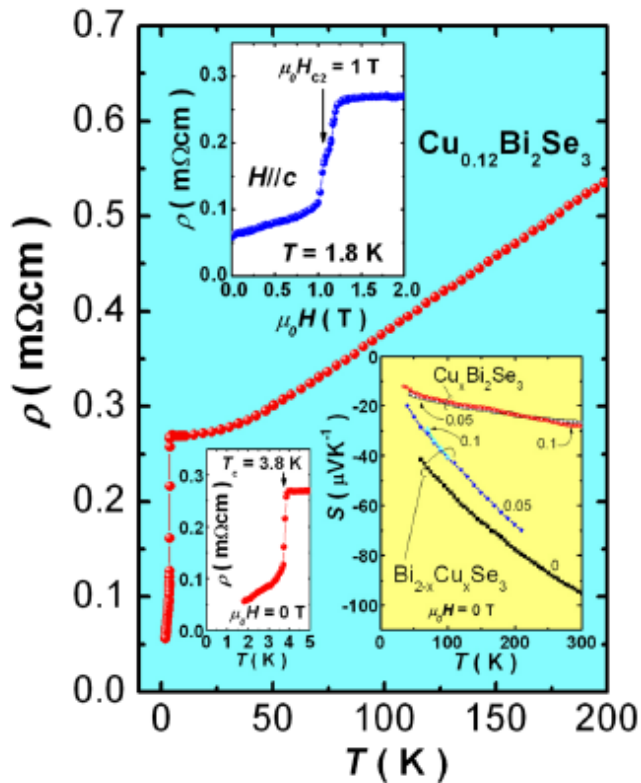
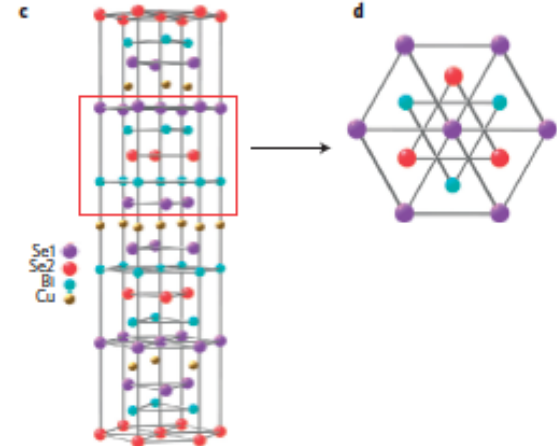
**Notice: superconductors don't fluctuate very much!**

$$\Delta \ll E_F \implies \xi_0 \approx v_F / \Delta \gg \lambda_F \approx v_F / E_F$$

# doping the 3d topological insulator $\text{Bi}_2\text{Se}_3$

## superconductivity in $\text{Cu}_x\text{Bi}_2\text{Se}_3$

Y. S. Hor, et al. Phys. Rev. Lett. **104**, 057001 (2010)  
 M. Kriener, et al., Phys. Rev. Lett. **106**, 127004 (2011)



$$T_c \sim 3\text{K}$$

$$n \sim 10^{20} \text{cm}^{-3}$$

low carrier concentration

$$\xi_{ab} / \lambda_F \approx 2 - 4$$

short coherence length

S.C. fluctuations

# Two-component superconducting states

$$\text{singlet: } E_g \quad \Delta_{\mathbf{p}} = \Delta_x (p_x p_y + \eta p_y p_z) + \Delta_y (p_x^2 - p_y^2 + \eta p_x p_z)$$

$$\text{triplet: } E_u \quad \mathbf{d}_{\mathbf{p}} = \Delta_x (\hat{\mathbf{x}} p_z - \eta \hat{\mathbf{z}} p_x) + \Delta_y (\hat{\mathbf{y}} p_z - \eta \hat{\mathbf{z}} p_y)$$

two options: **nematic** or **chiral** (TRS-breaking)

candidate for topological superconductivity

L. Fu and E. Berg, Phys. Rev. Lett. **105**, 097001 (2010)

J. W. F. Venderbos, V. Kozii, and L. Fu, B **94**, 180504(R) (2016)

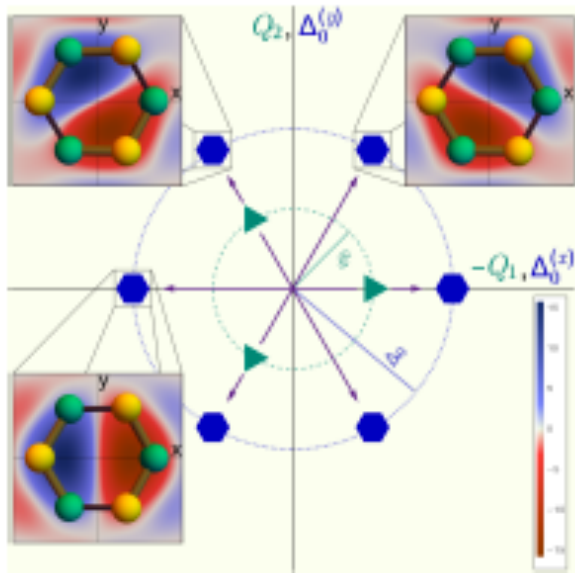
# Two-component superconducting states

singlet:  $E_g$   $\Delta_{\mathbf{p}} = \Delta_x (p_x p_y + \eta p_y p_z) + \Delta_y (p_x^2 - p_y^2 + \eta p_x p_z)$

triplet:  $E_u$   $\mathbf{d}_{\mathbf{p}} = \Delta_x (\hat{\mathbf{x}} p_z - \eta \hat{\mathbf{z}} p_x) + \Delta_y (\hat{\mathbf{y}} p_z - \eta \hat{\mathbf{z}} p_y)$

two options: **nematic** or chiral (TRS-breaking)

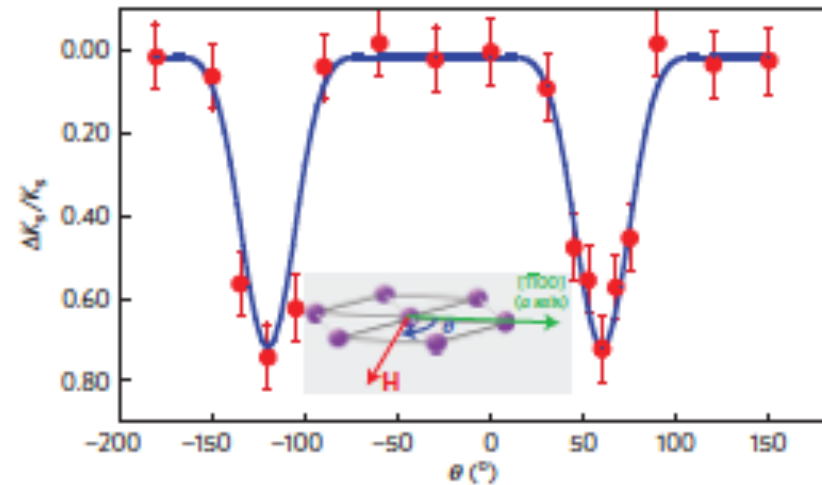
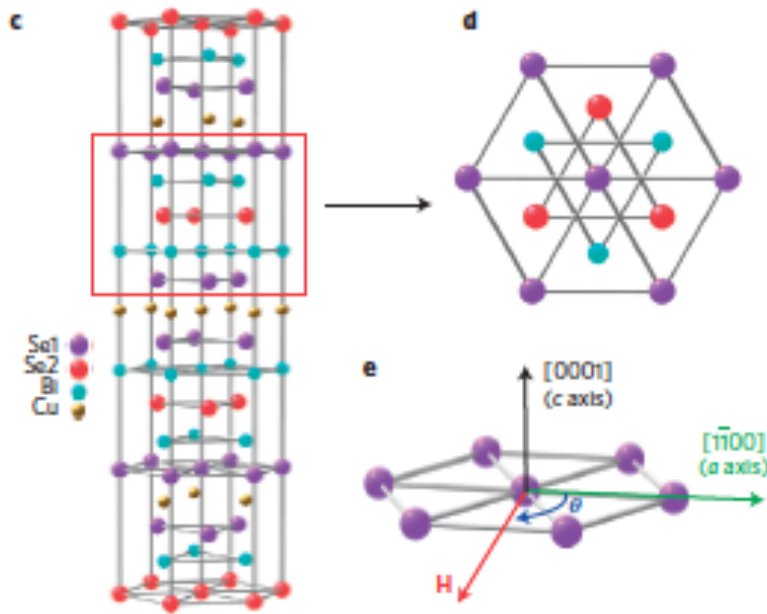
$d_{\mathbf{p},z}$



nematic state:  
superconductor breaks rotation symmetry

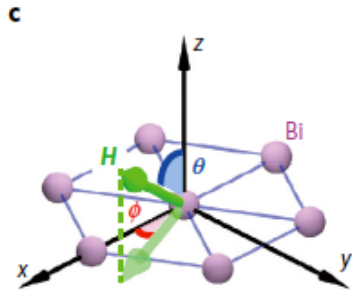


# rotational symmetry breaking

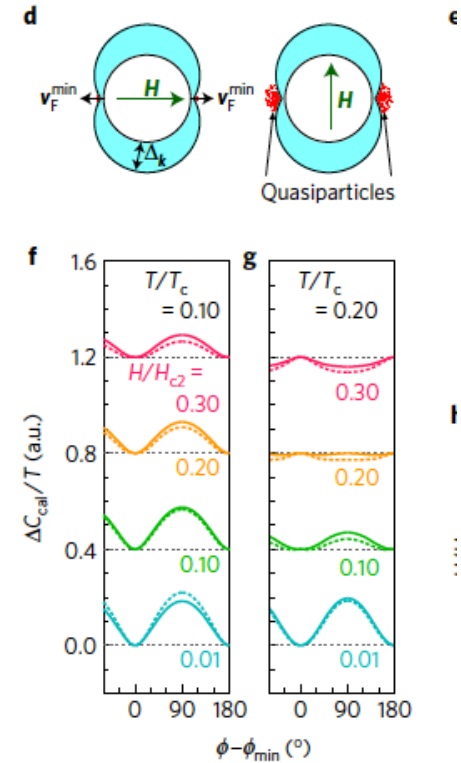
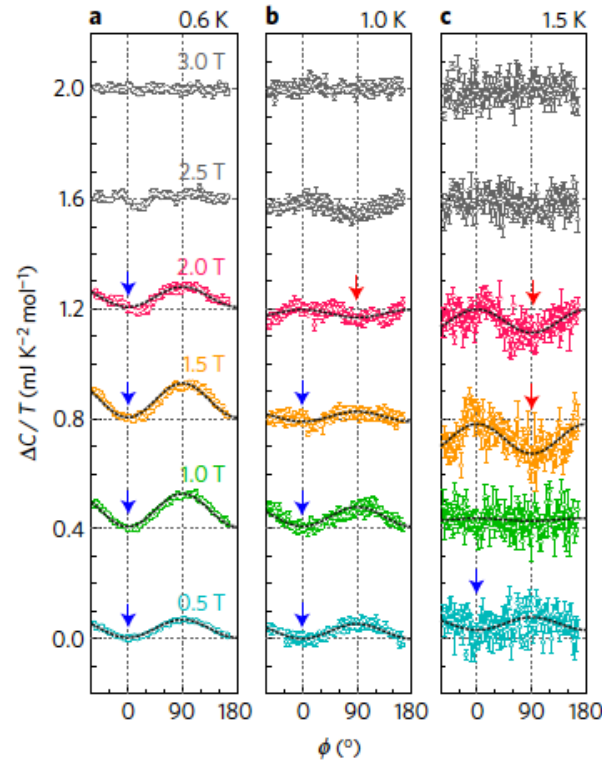


**nematic superconductor: either  $E_g$  or  $E_u$**

# anisotropic thermodynamic response



heat capacity as function of in-plane magnetic field



# vestigial order in $\text{Cu}_x\text{Bi}_2\text{Se}_3$

the symmetry group (trigonal) =  $D_{3d} \otimes U(1)$

$$E_u^* \otimes E_u = A_{1g} \oplus A_{2g} \oplus E_g$$

nematic superconductivity

time reversal symmetry

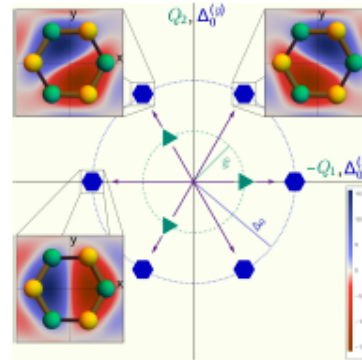
possibly relevant for thin films  
 L. Chirulli, PRB **98**, 014505 (2018)

$$q_1 = \Delta_x^* \Delta_x - \Delta_y^* \Delta_y \quad \hat{Q} = \begin{pmatrix} q_1 & q_2 \\ q_2 & -q_1 \end{pmatrix}$$

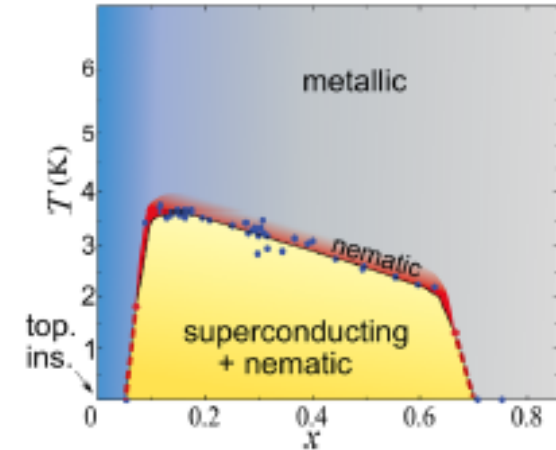
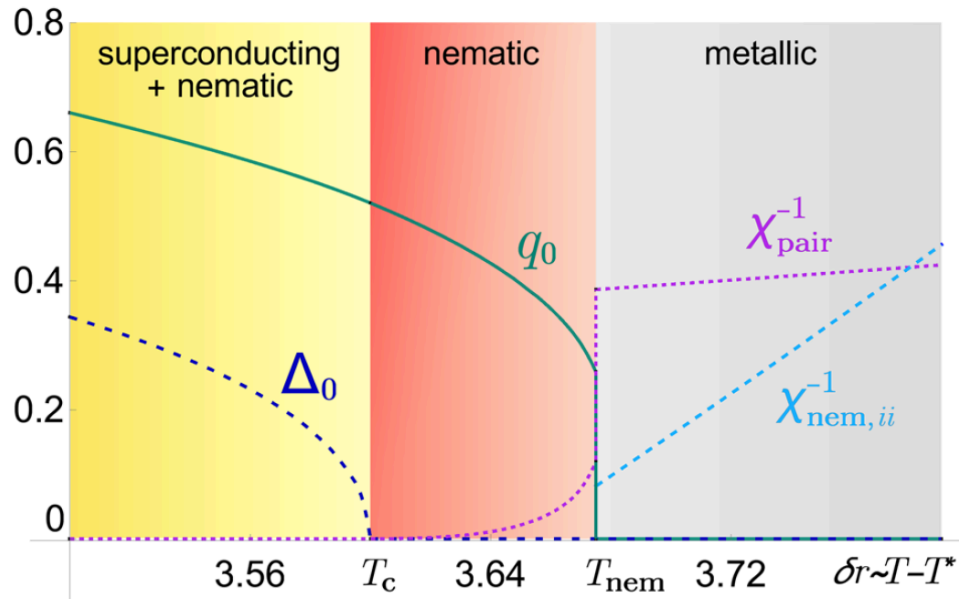
$$q_2 = \Delta_x^* \Delta_y + \Delta_y^* \Delta_x$$

$$f = \frac{r}{2} (q_1^2 + q_2^2) - \frac{g}{3} q_1 (q_1^2 - 3q_2^2) + \frac{u}{4} (q_1^2 + q_2^2)^2$$

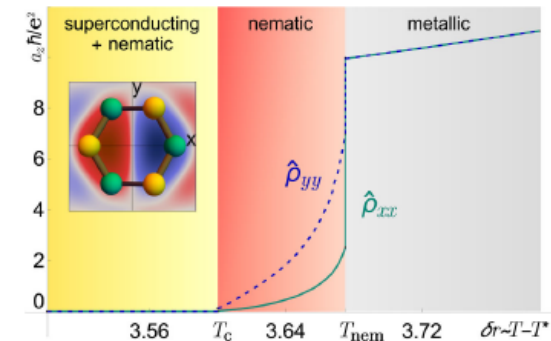
**3-state Potts model**  
 three degenerate  
 solutions that break  
 rotation invariance



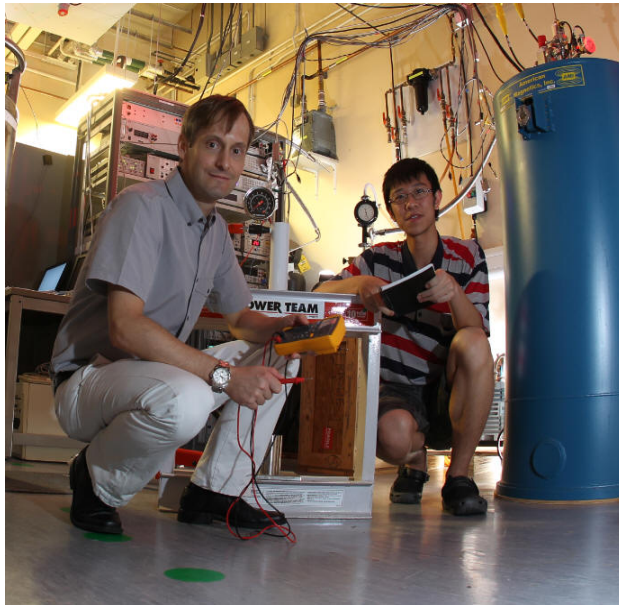
# vestigial order in $\text{Cu}_x\text{Bi}_2\text{Se}_3$



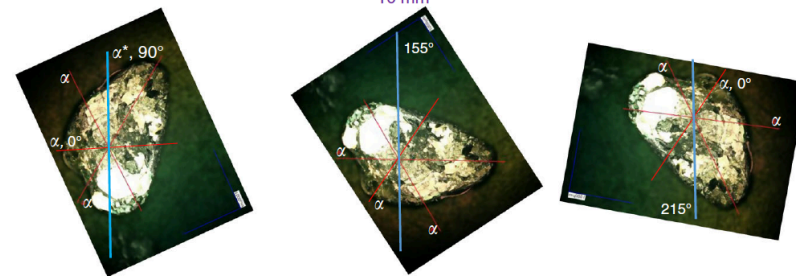
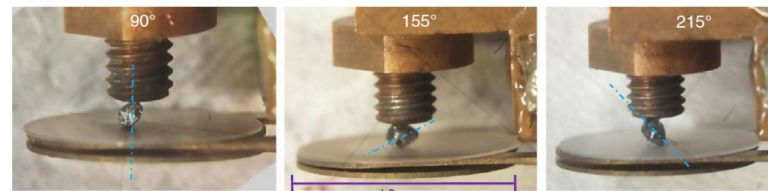
- large anisotropic para-conductivity
- coupling to the lattice
- Raman anomaly



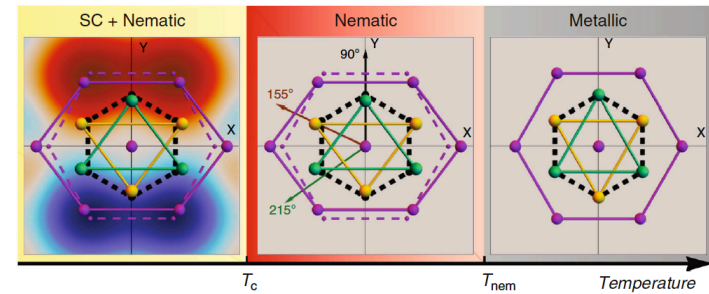
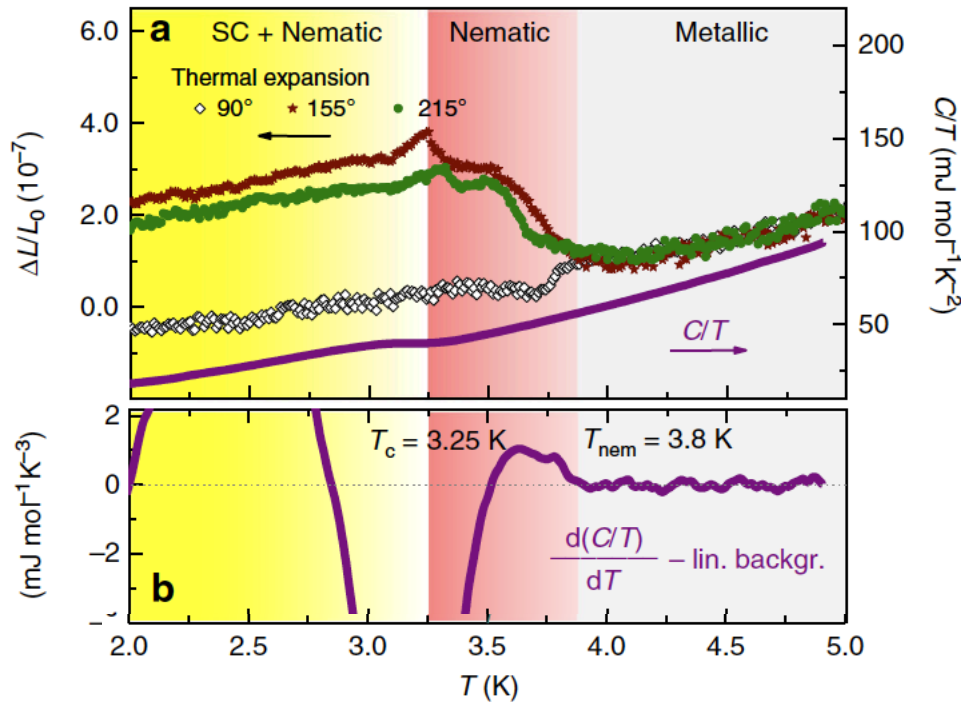
**Rolf Lortz**  
The Hong Kong University  
of Science & Technology



$\text{Nb}_{0.25}\text{Bi}_2\text{Se}_3$  single crystal mounted in the capacitive dilatometer along the three orientations  
(similar results for  $\text{Cu}_x\text{Bi}_2\text{Se}_3$ )

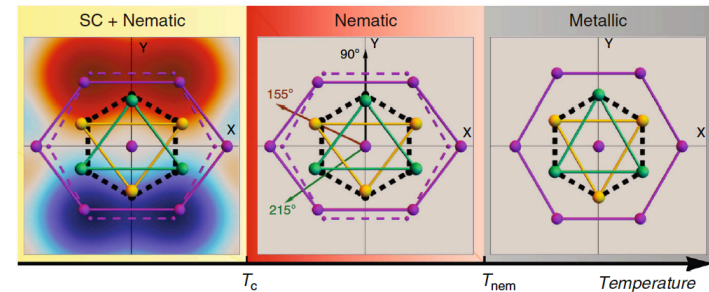
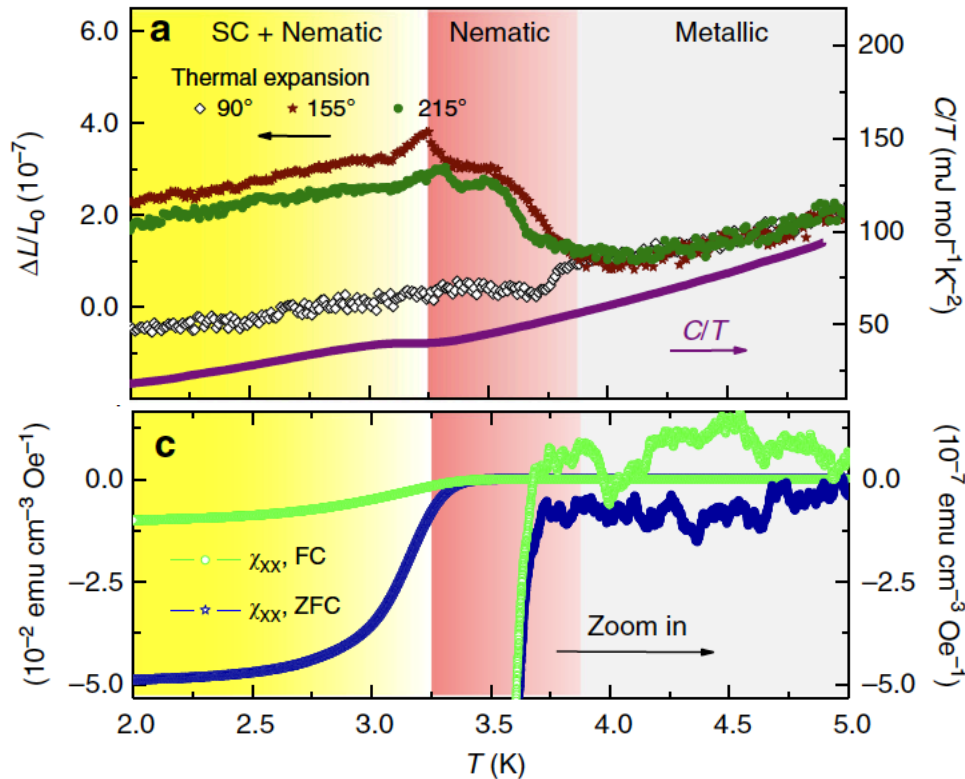


# High-precision thermal expansion measurements



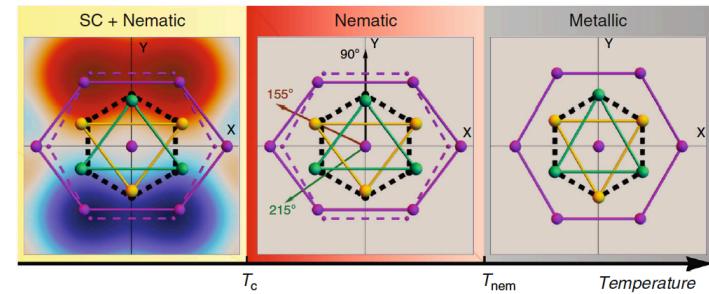
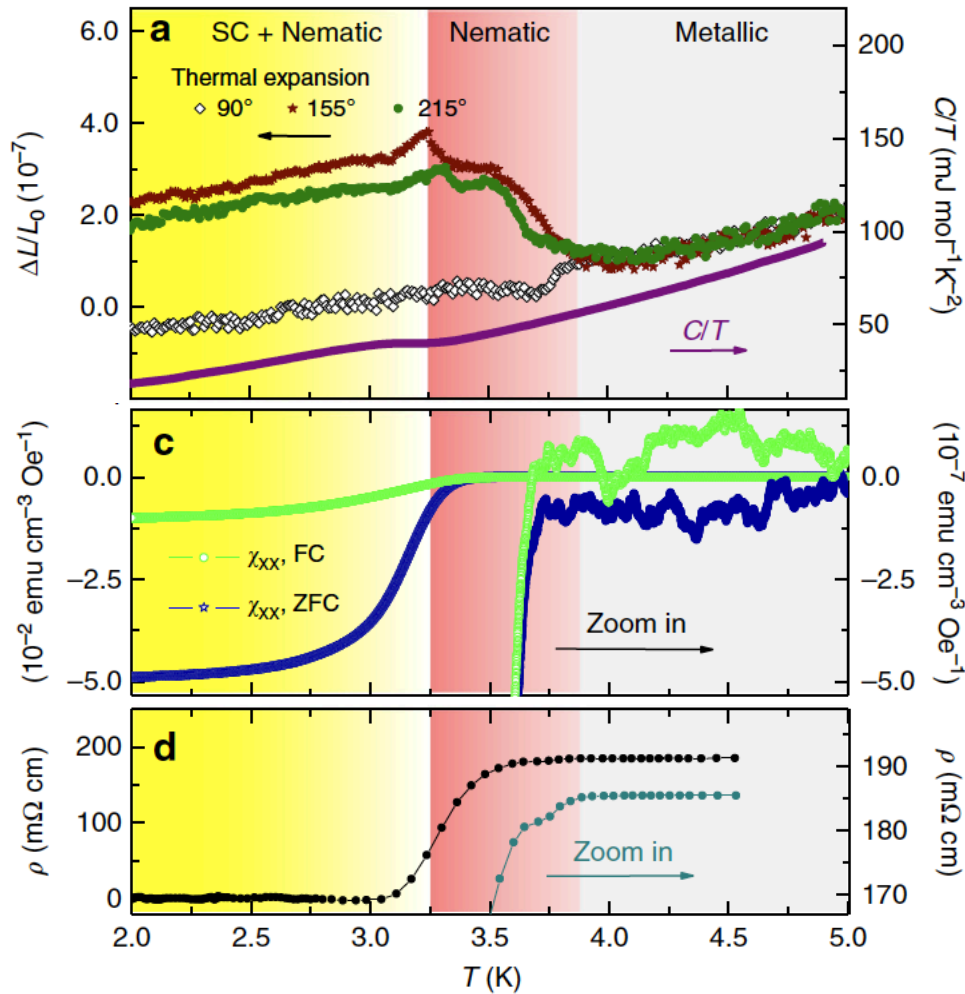
$Z_3$ -Potts symmetry breaking slightly above  $T_c$

# High-precision thermal expansion measurements



**susceptibility:** onset of diamagnetic fluctuations at  $T_{\text{nem}}$

# High-precision thermal expansion measurements

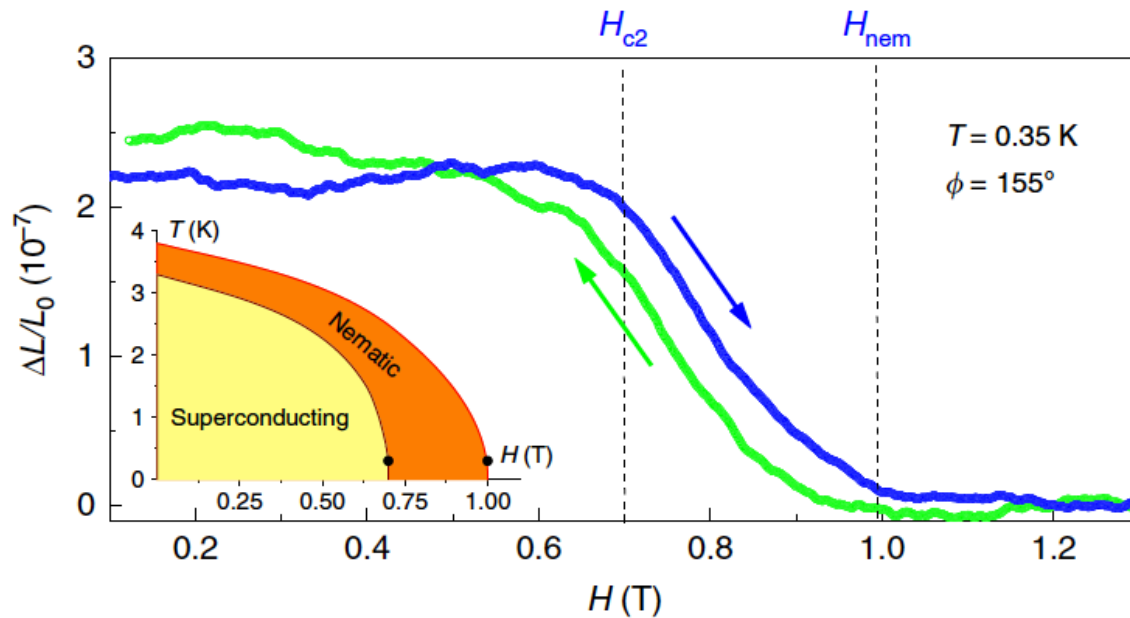


**susceptibility:** onset of diamagnetic fluctuations at  $T_{nem}$

**transport:** onset of paraconductivity at  $T_{nem}$



# magnetostriction measurements



- nematic order is tied to superconductivity
- possibility of an isolated  $Z_3$  quantum Potts transition

**strong evidence for vestigial superconducting phase !**

$$\langle \Delta_{x,y} \rangle = 0$$

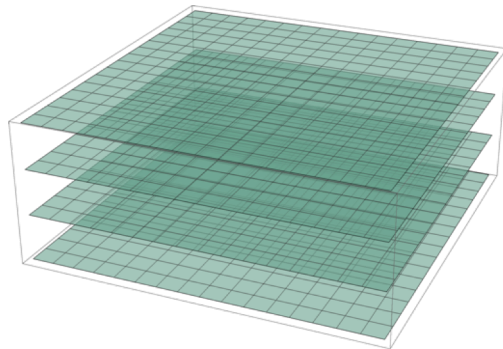
$$\langle \Delta_x^* \Delta_x - \Delta_y^* \Delta_y \rangle \neq 0$$

**condensation of anisotropic pairing fluctuations**

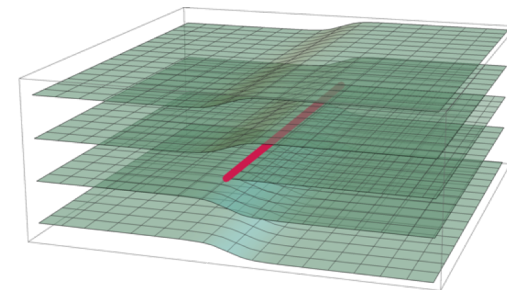
# the pairing state of $\text{Sr}_2\text{RuO}_4$

(in collaboration with Roland Willa and Rafael Fernandes)

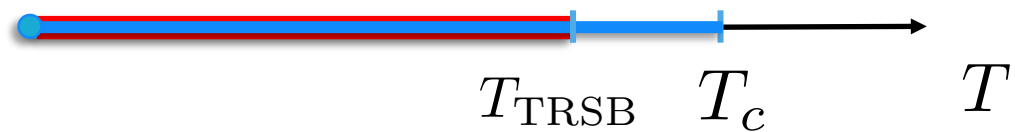
perfect  $\text{Sr}_2\text{RuO}_4$   
 time-reversal symmetric, single-component  
 (d-wave) superconductor



TRS-breaking is no bulk effect but  
 occurs near edge dislocations



TRSB near the dislocation  $d + e^{i\phi(\mathbf{r})}g$



consistent with quasiparticle  
 interference data

R. Sharma et al., PNAS **117**, 5222 (2020)

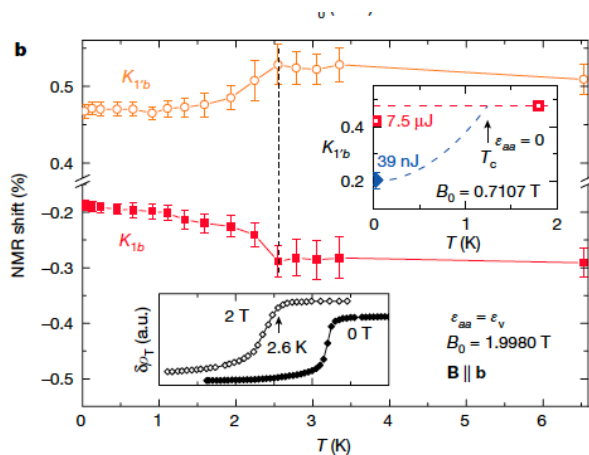
d-wave bulk superconductivity

# the pairing state of $\text{Sr}_2\text{RuO}_4$

A. P. Mackenzie, T. Scaffidi, C. W. Hicks, and Y. Maeno, npj Quantum Mat. **2**, 40 (2017)

## singlet pairing

A. Pustogow et al., Nature **574**, 72 (2019)

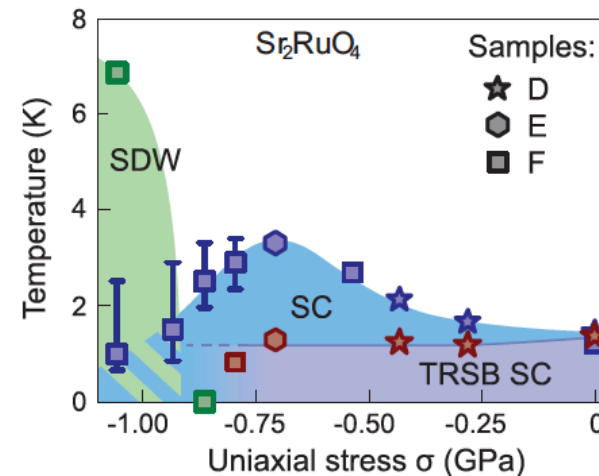


$$d_{xz} \pm id_{yz} ?$$

nontrivial orbital pairing ?

## split transitions under strain

V. Grinenko et al., arXiv:2001.08152



O. Gingras et al. PRL **123**, 217005 (2019)

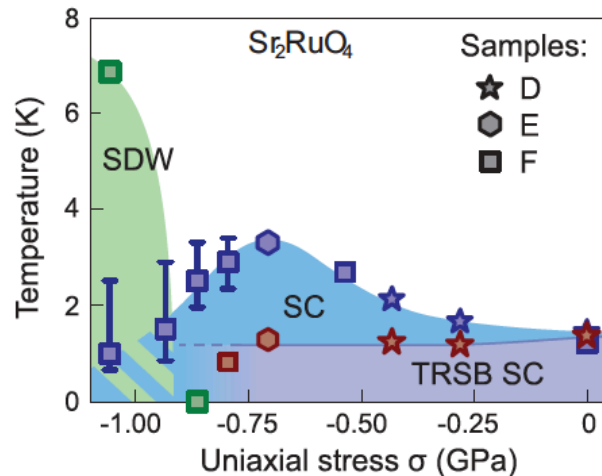
A. Ramires and M. Sigrist PRB **100**, 104501 (2019)

W. Huang et al., PRB **100** 134506 (2019)

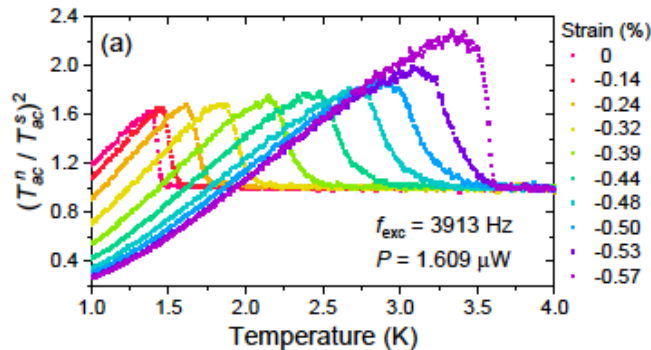
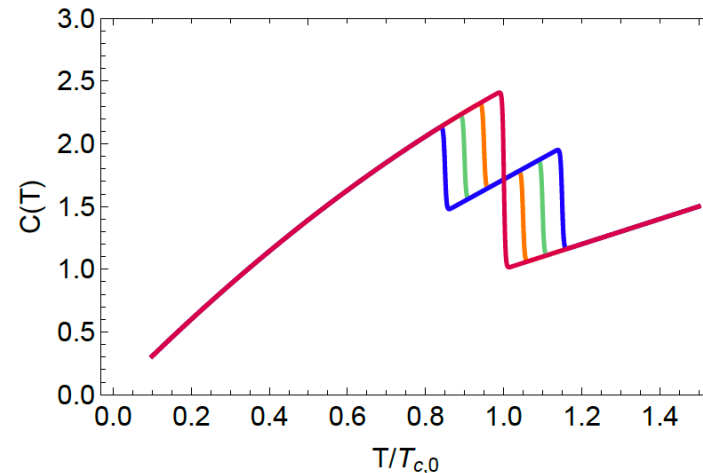
H.-G. Suh et al., PRR **2**, 032023(R) (2020)

# the pairing state of $\text{Sr}_2\text{RuO}_4$

split transitions under strain  
 V. Grinenko et al. arXiv:2001.08152



$E_{g,u}$ : expect at least comparable heat capacity jumps at the two transitions



no second heat capacity jump  
 $\rightarrow$  inconsistent with  $E_{g,u}$

$\rightarrow$  accidental degeneracy

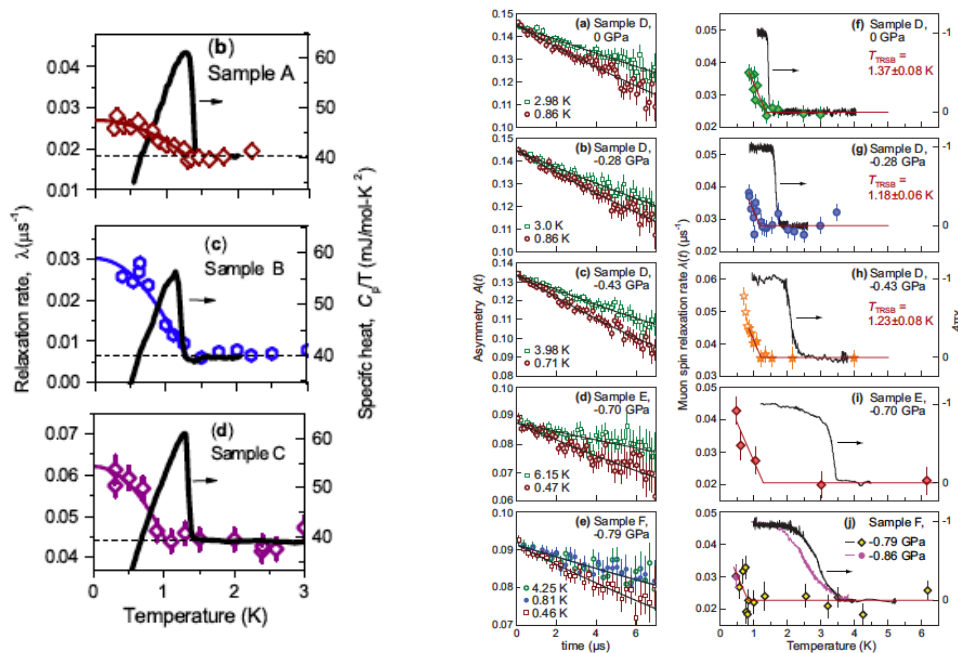
$$d_{x^2-y^2} \pm ig_{xy}(x^2-y^2)$$

Y.-S. Li et al. arXiv:1906.07597

S. A. Kivelson et al. arXiv:2002.00016

# $\mu$ -SR data under strain

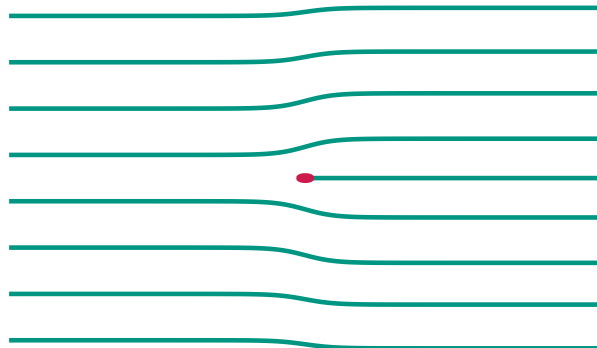
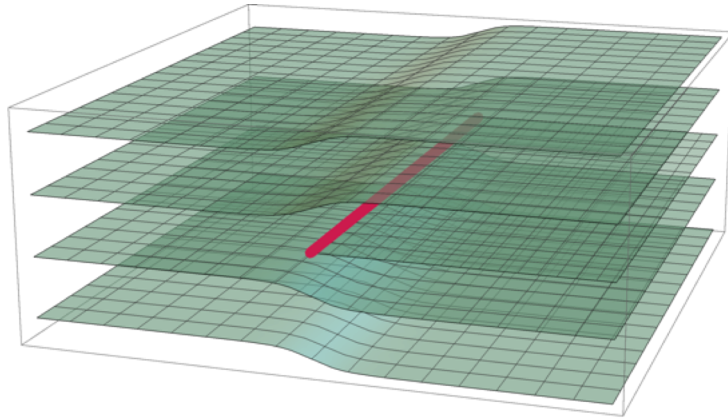
V. Grinenko et al. arXiv:2001.08152



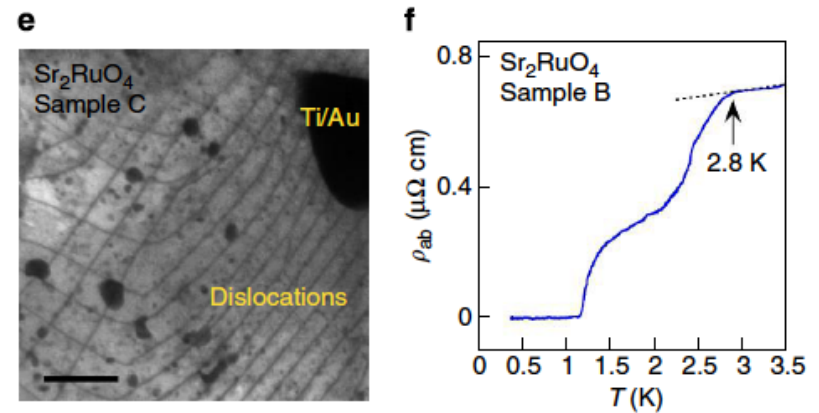
- sample dependence of  $T_C - T_{\text{TRSB}}$  even at zero strain
- broad distribution of local fields (like spin glasses, only smaller in magnitude)

*“The internal field is thought to arise at edges, defects, and domain walls ...”*

# edge dislocations in $\text{Sr}_2\text{RuO}_4$

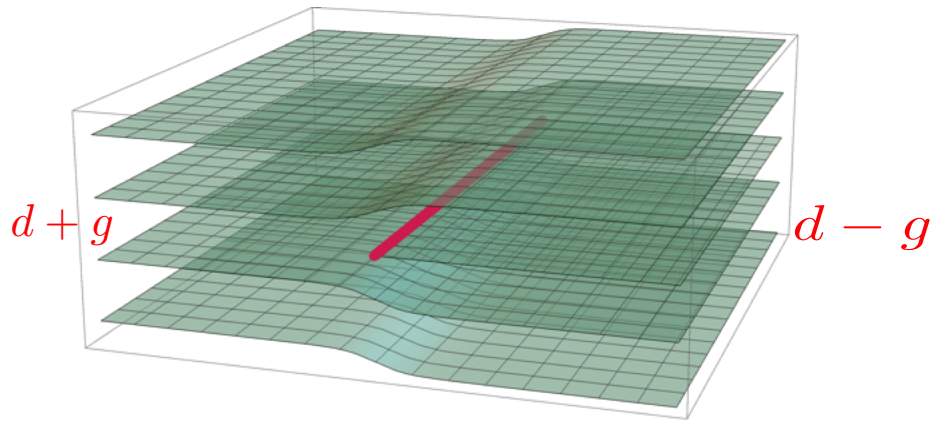


impact of edge dislocations on  
superconductivity



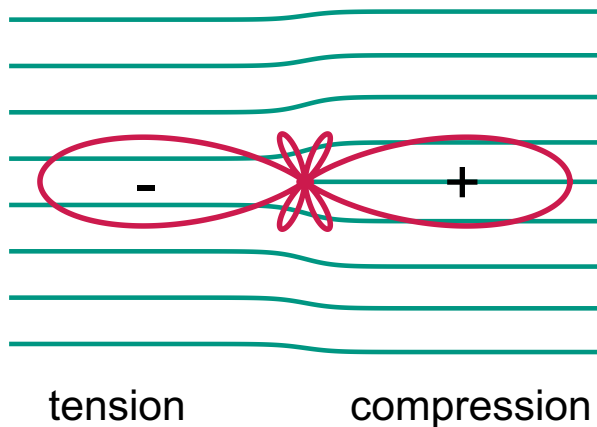
Y.A. Ying, N.E. Staley, Y. Xin, K. Sun, X. Cai, D. Fobes,  
T.J. Liu, Z.Q. Mao & Y. Liu, Nature Comm. **4**, 2596 (2013)

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slowly-decaying  
secondary component

strain fields near the dislocation



$$f_c \sim \varepsilon_{xy} (\psi_d^* \psi_g + h.c.)$$

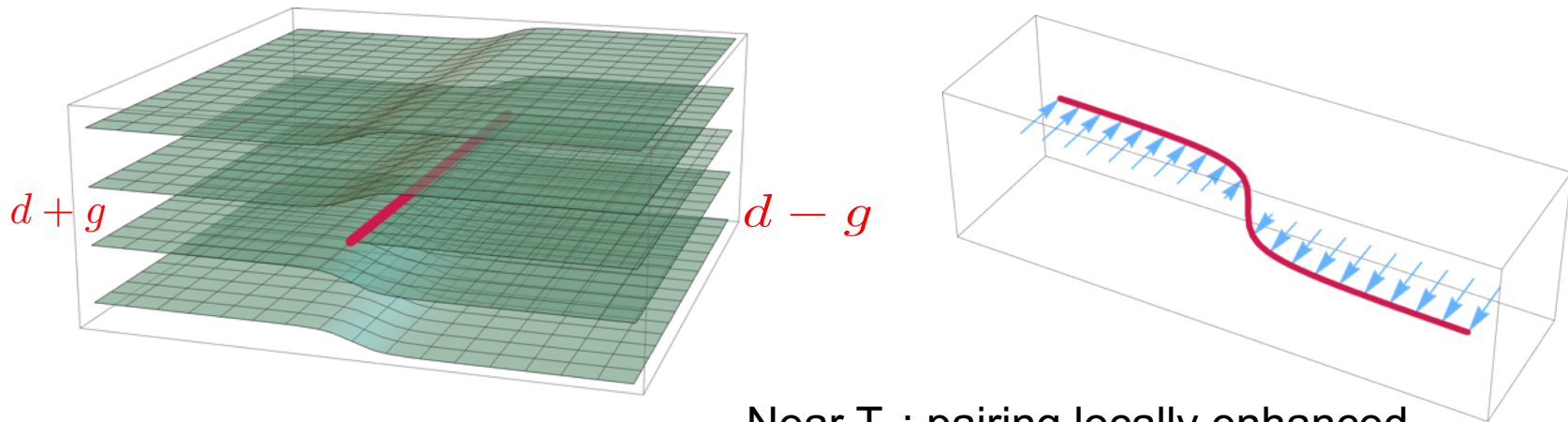
(strong local mixing of order parameters)

$$+iH_z \varepsilon_{x^2-y^2} (\psi_d^* \psi_g - h.c.)$$

(TRS-breaking: trainable by a magnetic field)

# edge dislocations in $\text{Sr}_2\text{RuO}_4$

variational ansatz: sign-change through the complex plane  
 similar to twin boundaries in FeSe: T. Watashige et al. PRX 5, 031022 (2015)



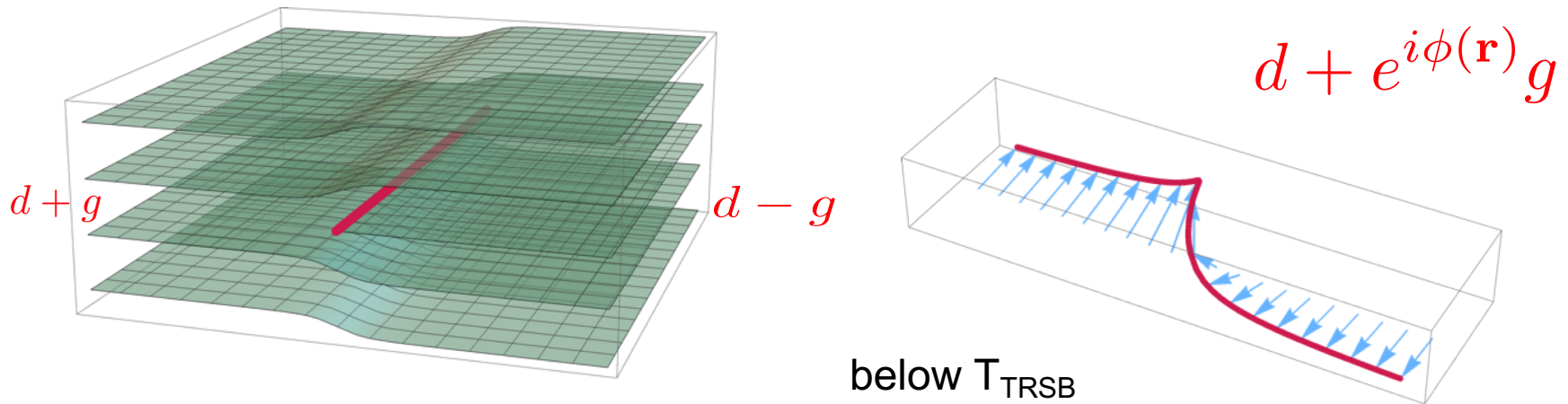
Near  $T_c$ : pairing locally enhanced



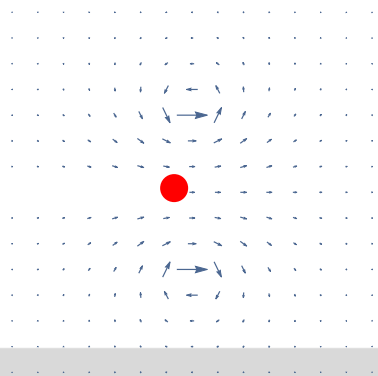


# edge dislocations in $\text{Sr}_2\text{RuO}_4$

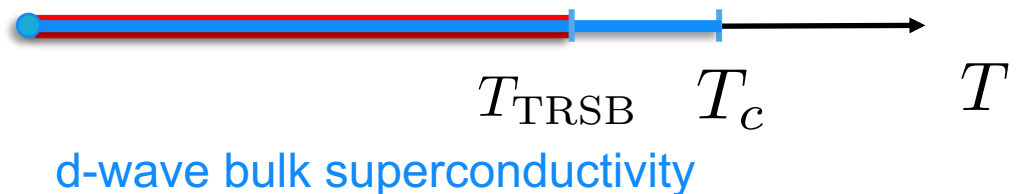
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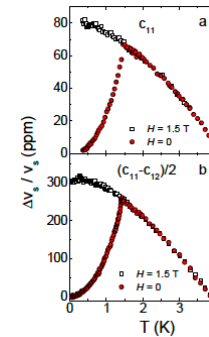
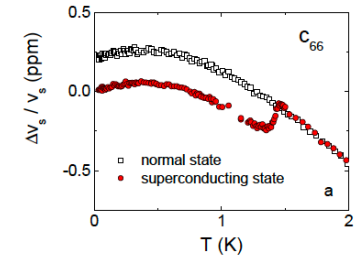
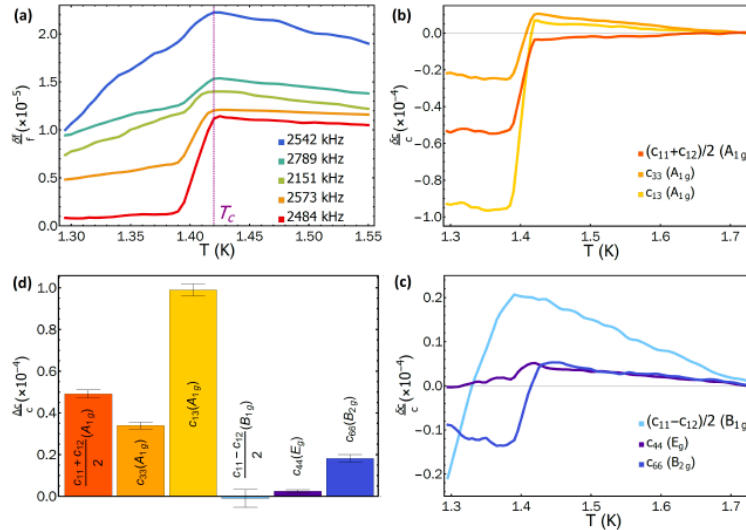
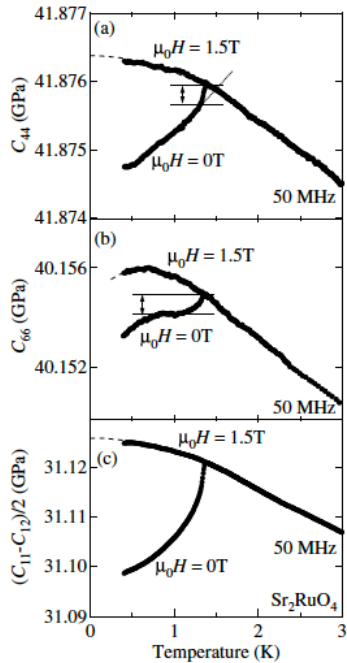
one-dim. structures with complex current pattern



TRSB near the dislocation



# problem: elastic constant discontinuity in $C_{66}$



N. Okuda et al. JPSJ 71, 1134 (2002)

S. Ghosh et al. arXiv:2002.06130

S. Benhabib et al. arXiv:2002.05916

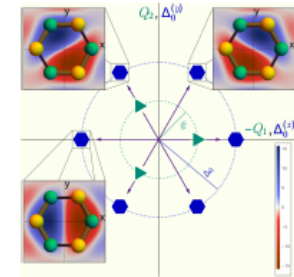
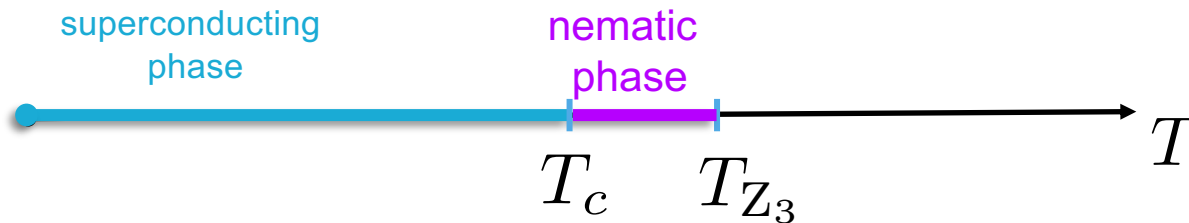
dislocation-induced  
change

$$\left. \frac{\delta C}{C} \right|_{\text{disl.}} \sim 0.1 \rho_{\text{disl}} l^2 \sim 2 - 10\% \gg \left. \frac{\Delta C}{C} \right|_{\text{sc.}}$$

G. Grimwall, *Thermophysical properties of materials*, North-Holland (1999)

# conclusions

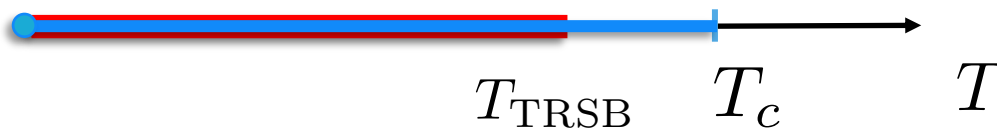
## 1. doped $\text{Bi}_2\text{Se}_3$ Superconducting fluctuations split the superconducting and nematic phase transitions



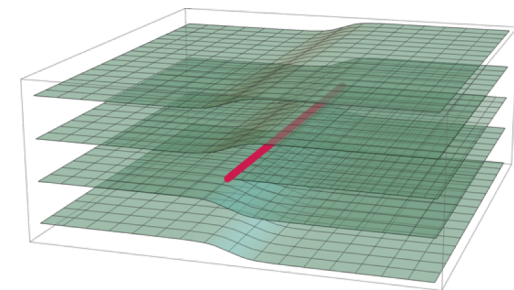
U(1) symmetry and  $Z_3$  rotational symmetry separately broken

## 2. absence of heat capacity anomaly at TRSB transition + broad distribution of field seen in $\mu$ -SR

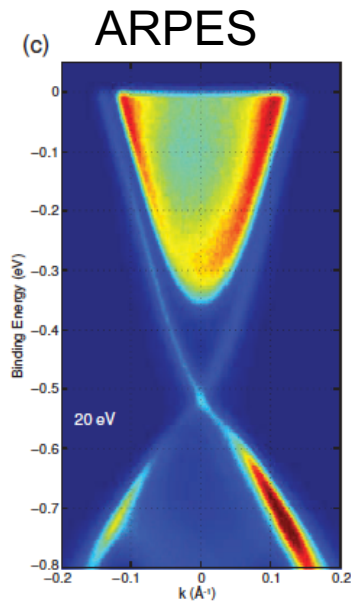
TRS-breaking only near dislocations  
 $\rightarrow$  one-dimensional structures with complex order parameter and current pattern



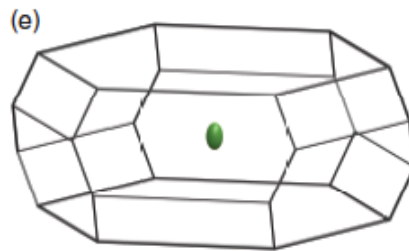
d-wave bulk superconductivity



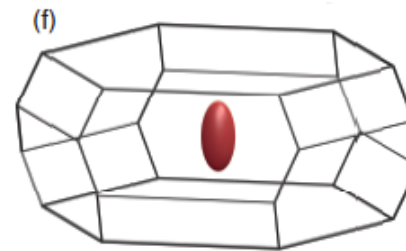
# evolution of the Fermi surface with carrier concentration



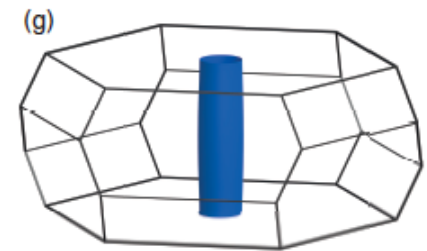
SdH



$$n \sim 10^{17} \text{ cm}^{-3}$$



$$n \sim 10^{19} \text{ cm}^{-3}$$



$$n \sim 10^{20} \text{ cm}^{-3}$$

electronic structure becomes increasingly anisotropic