

2024 Theory Winter School

New Frontiers in Superconductivity

URANIUM DITELLURIDE

A VARIETY OF SPIN-TRIPLET, HIGH MAGNETIC FIELD, AND/OR
PRESSURE-INDUCED SUPERCONDUCTING PHASES

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ACKNOWLEDGMENTS



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Research supported by



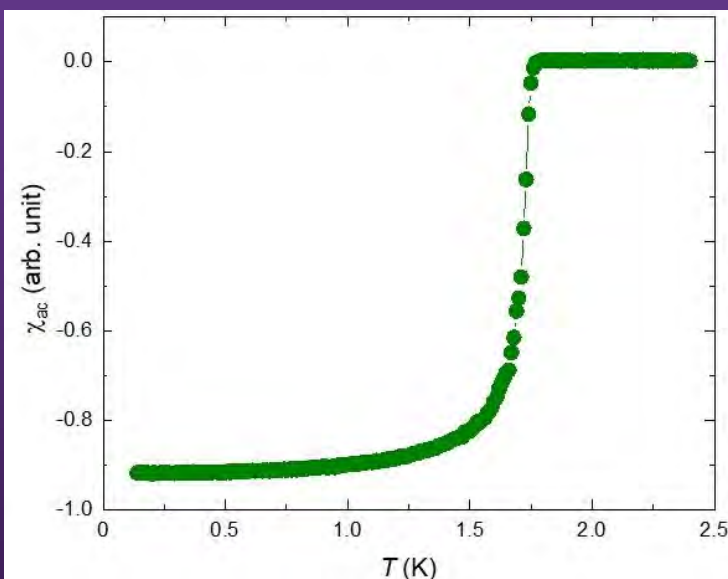
NIST NRC Postdoctoral Fellowship (US citizens only)
nicholas.butch@nist.gov

OUTLINE

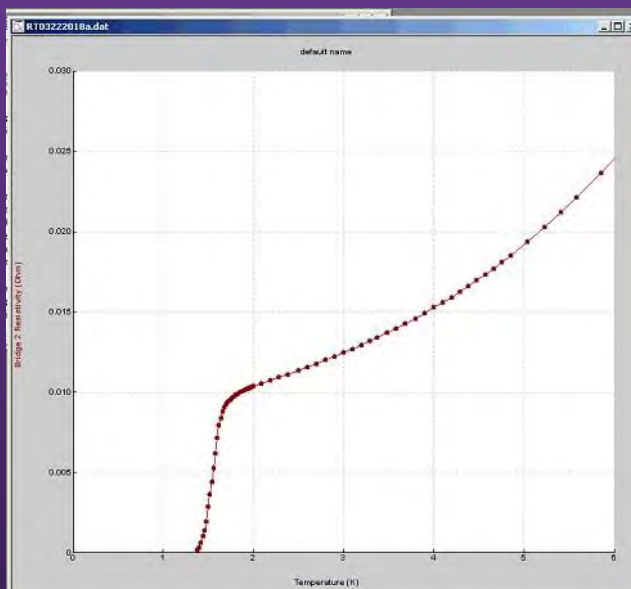
- BASIC PROPERTIES UTe_2
- SPIN TRIPLET PAIRING – SC ORDER PARAMETER?
- MULTIPLE SC PHASES – “LAZARUS” AND PRESSURE TUNING

SUPERCONDUCTIVITY IN UTe_2

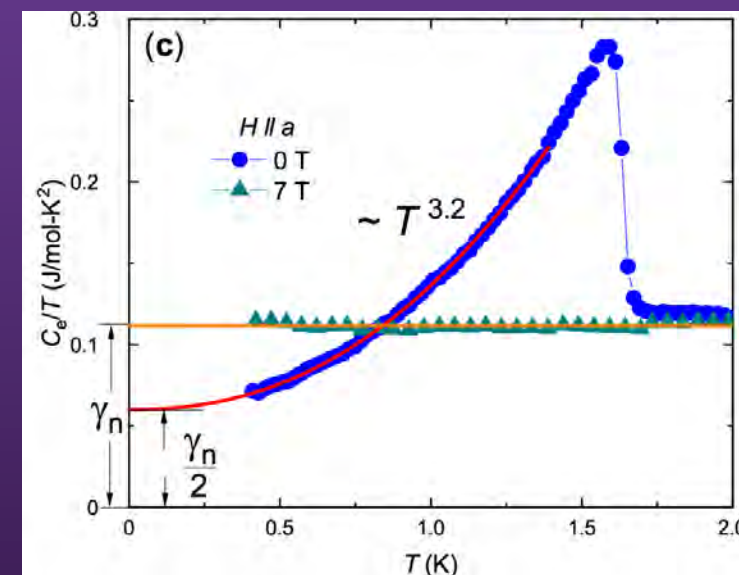
AC magnetic susceptibility



Electrical resistivity



Heat capacity



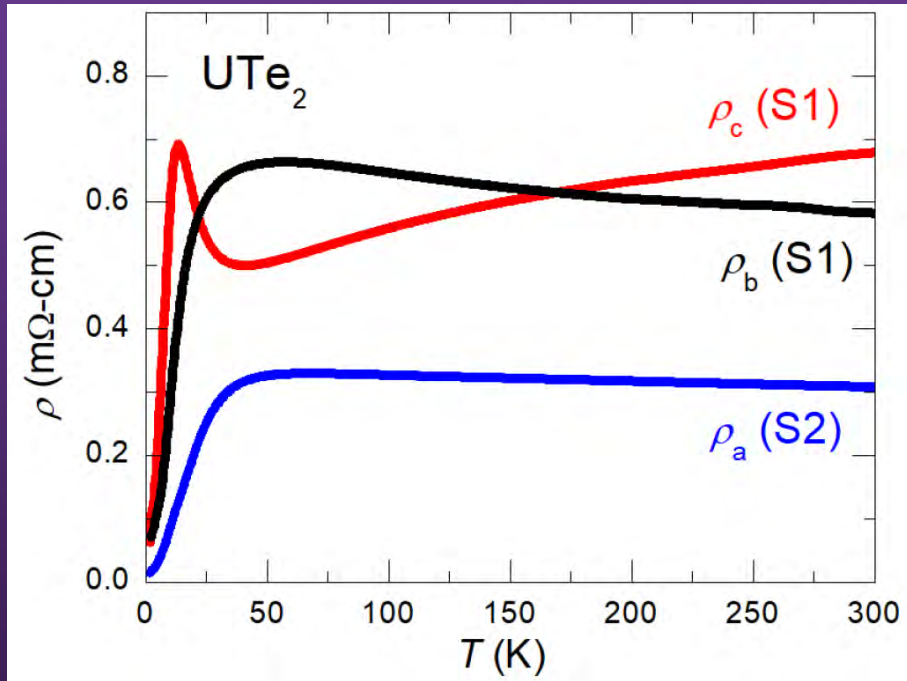
First results from Butch group, measured at UMD (March 2018)

Ran Science 365, 684 (2019)

Highest transition temperature 2.1 K

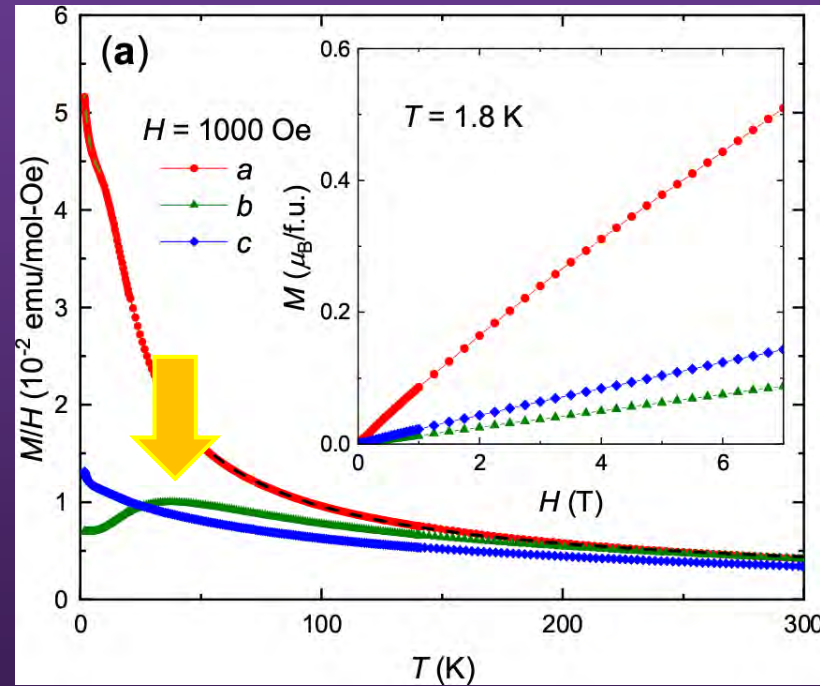
UTe₂ – 3D HEAVY FERMION (FERMI LIQUID)

a- and b-axes: local maximum resistivity
c-axis: more complicated

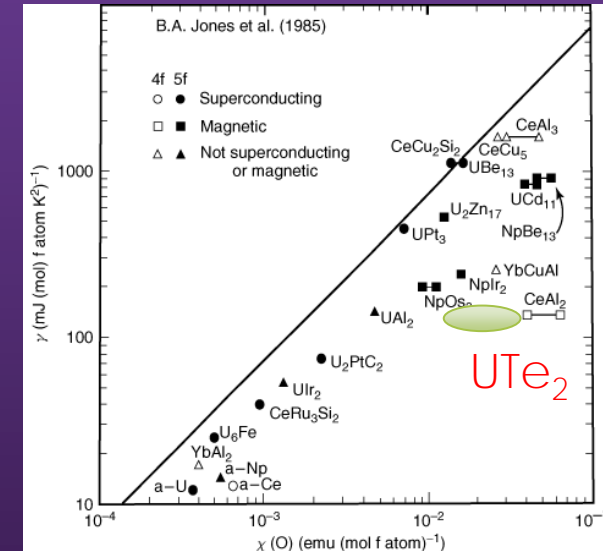
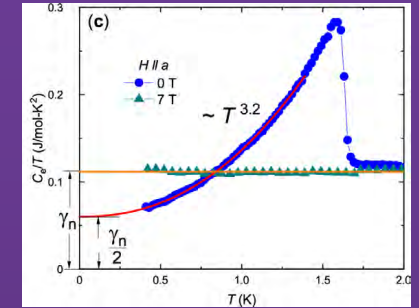


Eo Phys. Rev. B 106, L060505 (2022)

b-axis: local maximum susceptibility

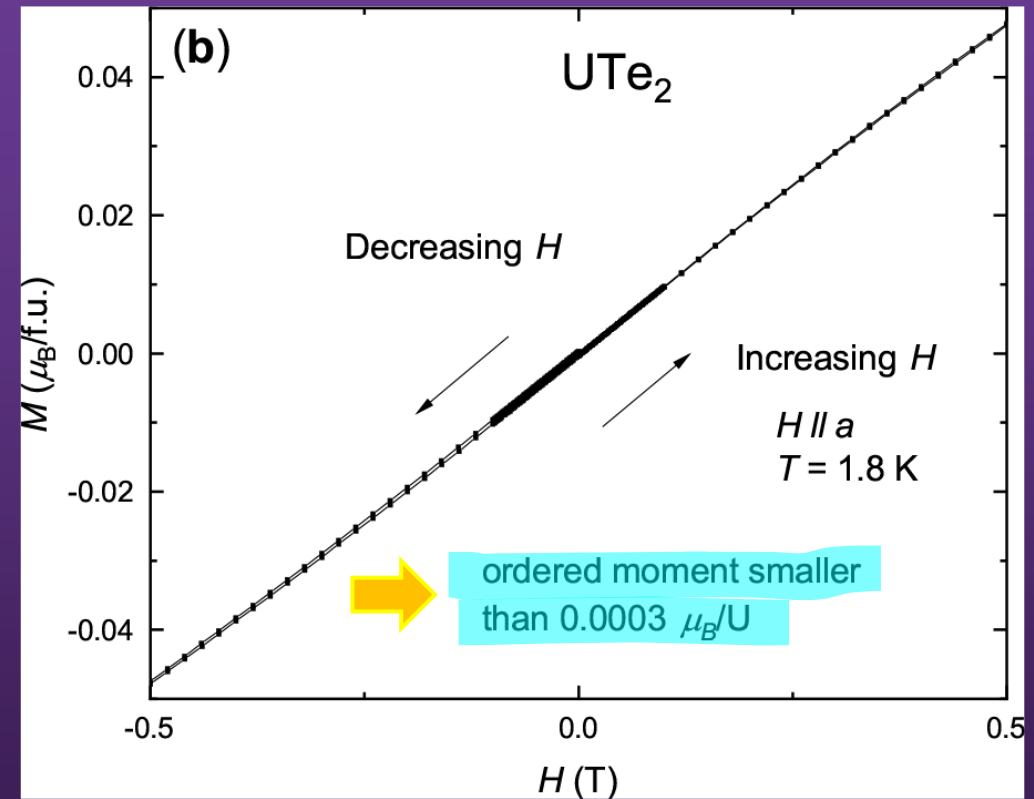
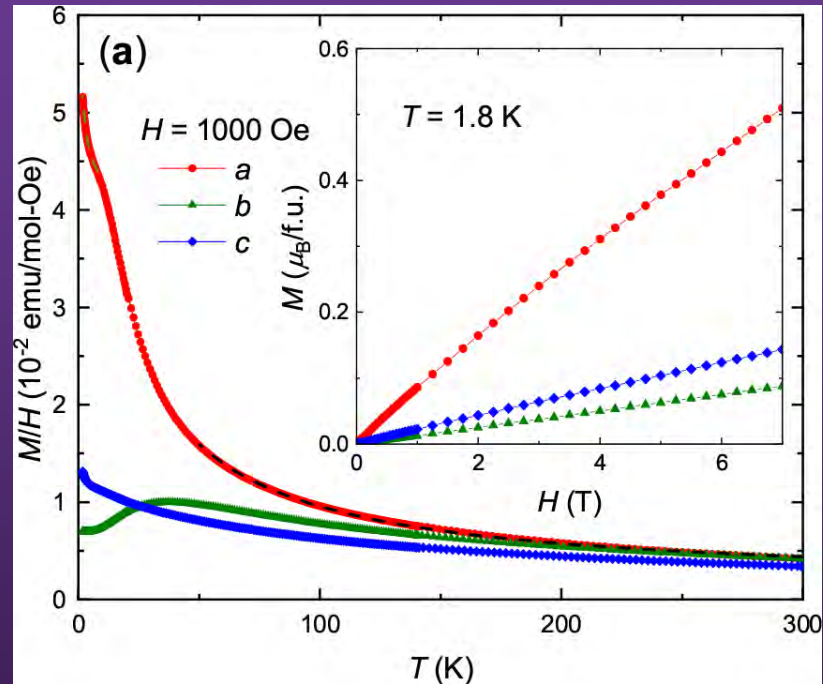


Ran Science 365, 684 (2019)



Wilson ratio χ/γ typical

UTe₂ REMAINS PARAMAGNETIC TO LOWEST TEMPERATURES



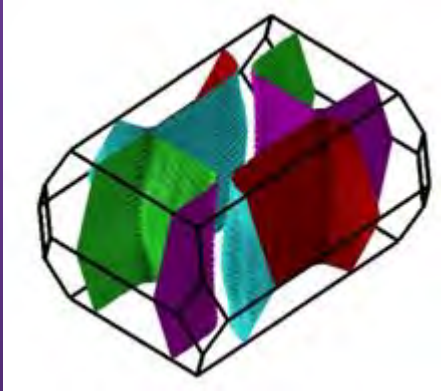
Ran Science 365, 684 (2019)

Neutron diffraction: No magnetic transitions down to 2.7 K (or reported since)

Hutanu et al, Acta Cryst. B 76, 137 (2020)

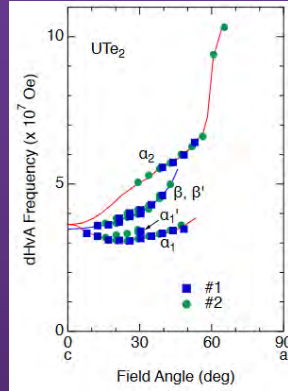
UTe₂ ELECTRONIC STRUCTURE – QUASI 2-DIMENSIONAL

LDA – 2D FS “ThTe2”

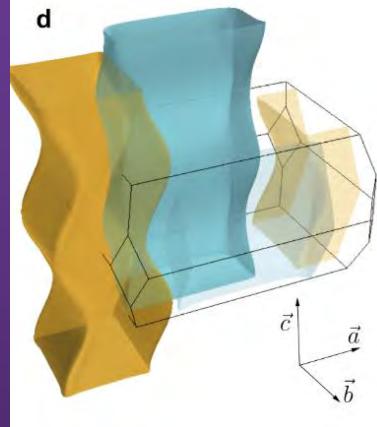


f-less Fermi Surface

dHvA oscillations – heavy

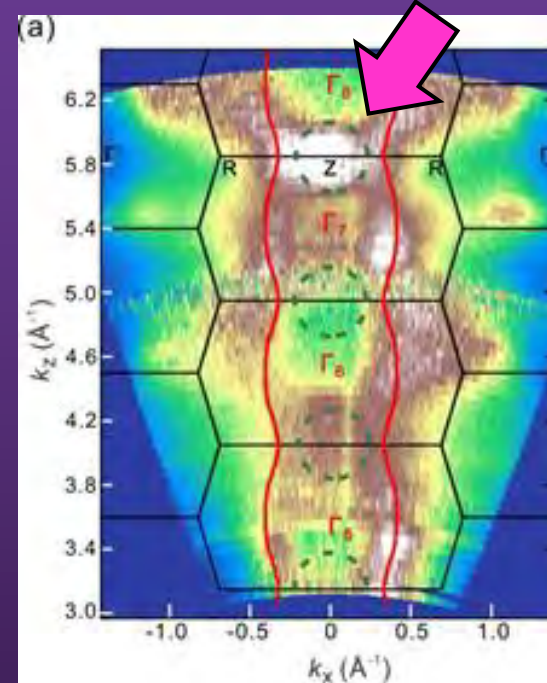


Aoki JPSJ 91, 083704 (2022)



Eaton arxiv:2302.04758 (2023)

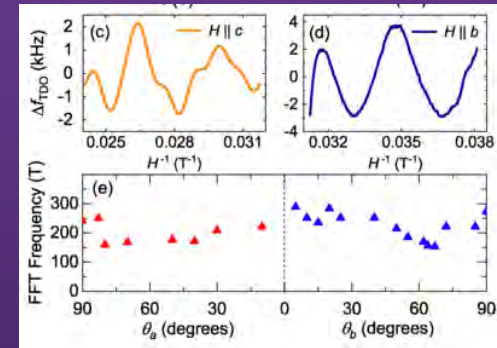
ARPES: additional 3D pocket



Miao et al, PRL 124, 076401 (2020)

Also Fujimori JPSJ 88, 103701 (2019)

dHvA - pocket



Broyles PRL 131, 036501 (2023)

ARPES did not detect the effects of f-electron hybridization near chemical potential
 Quantum oscillations suggest heavy mass, but
 FS cross section similar to “light” FS
 More 3D pocket at Z unexpected in calculations

REMINDER: MAGNETIC FIELD DESTROYS SUPERCONDUCTIVITY

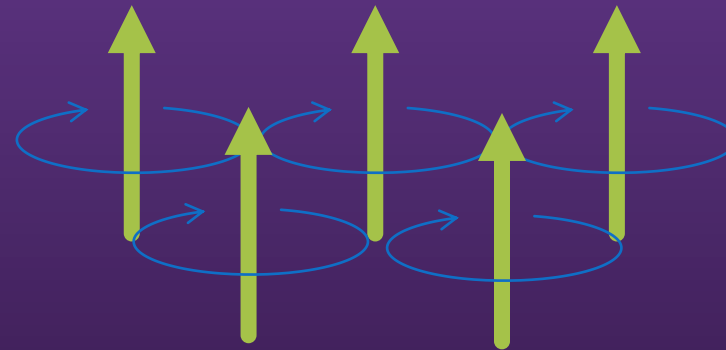
PARAMAGNETIC (SPIN) LIMIT



Zeeman effect breaks *singlets*

ORBITAL LIMIT

Mag. field penetrates in quantized vortices with circulating supercurrent

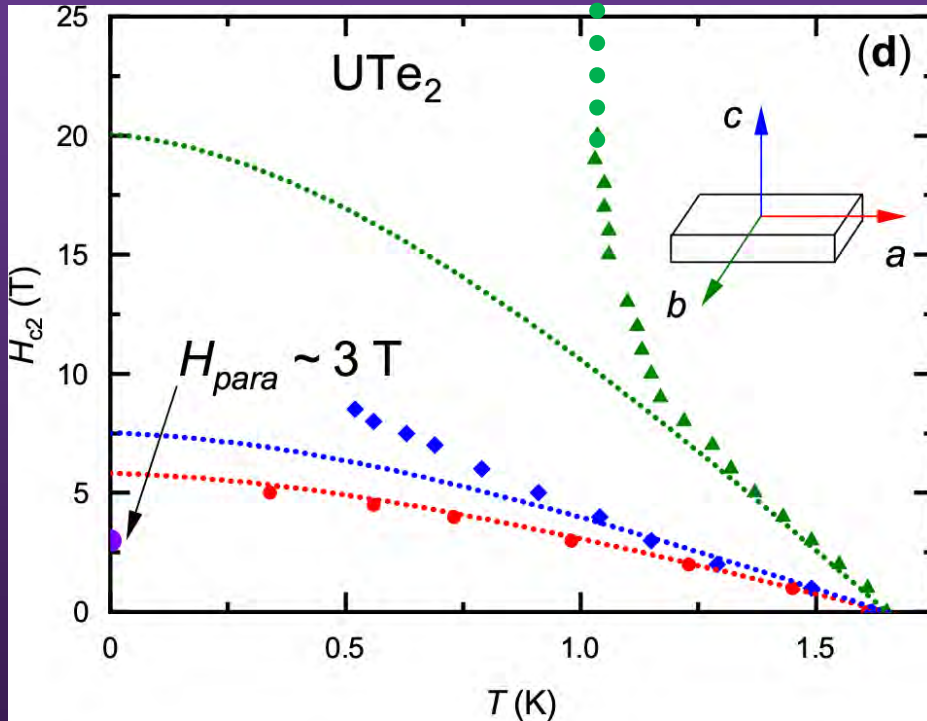


Overlap of vortex cores, *general*

UTe₂: ANISOTROPIC, LARGE UPPER CRITICAL FIELD

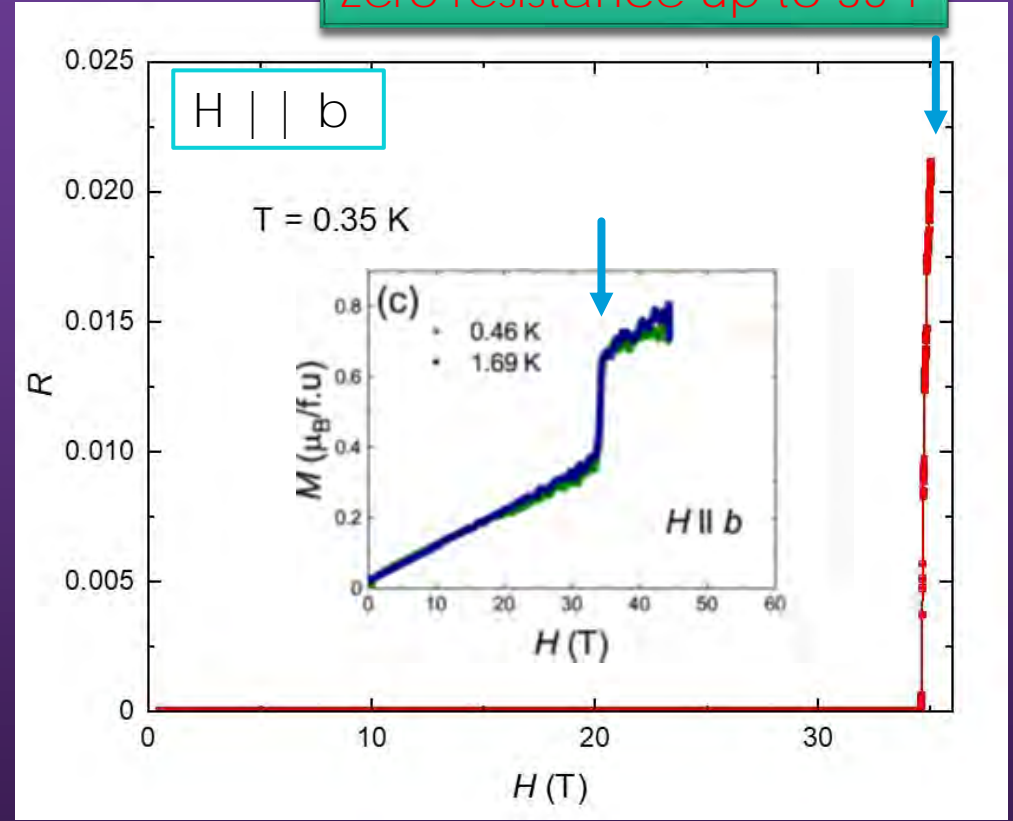
35 T

Ran Science 365, 684 (2019)



Paramagnetic limit exceeded along every direction

Zero resistance up to 35 T

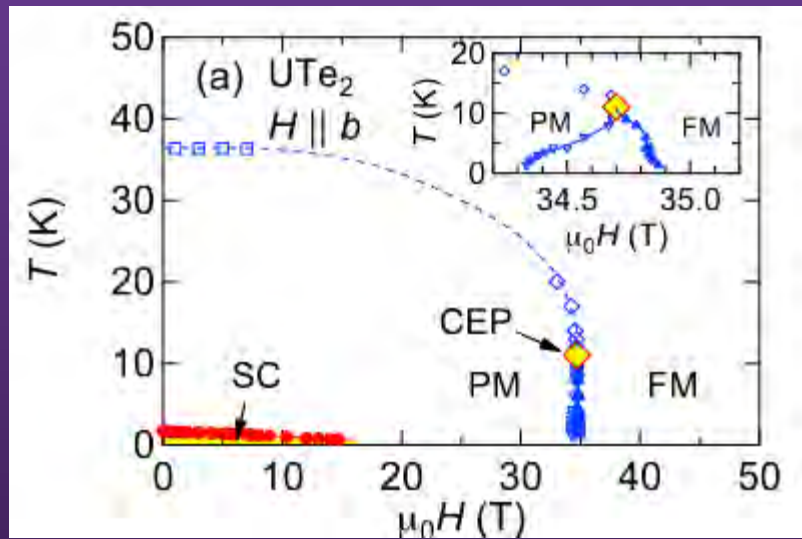


Ran Nature Physics 15, 1250 (2019)

B-axis: step in $M(H)$ at 35 T – not orbital limit

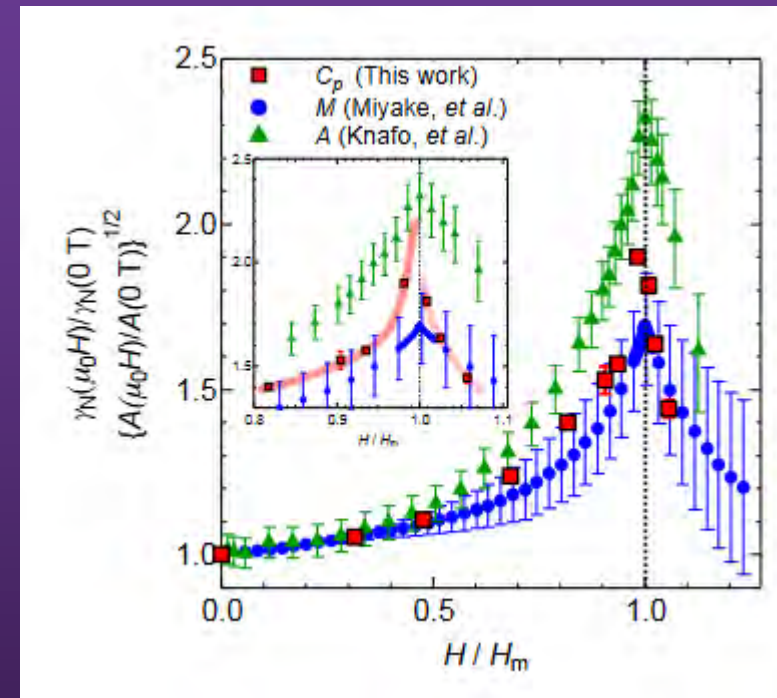
Also, Tohoku/Grenoble collaboration - JPSJ 88, 063706 (2019); JPSJ 88, 063707 (2019)

METAMAGNETIC TRANSITION – NOT ORBITAL LIMIT



Miyake JPSJ 88, 063706 (2019)

Extends to higher temperatures



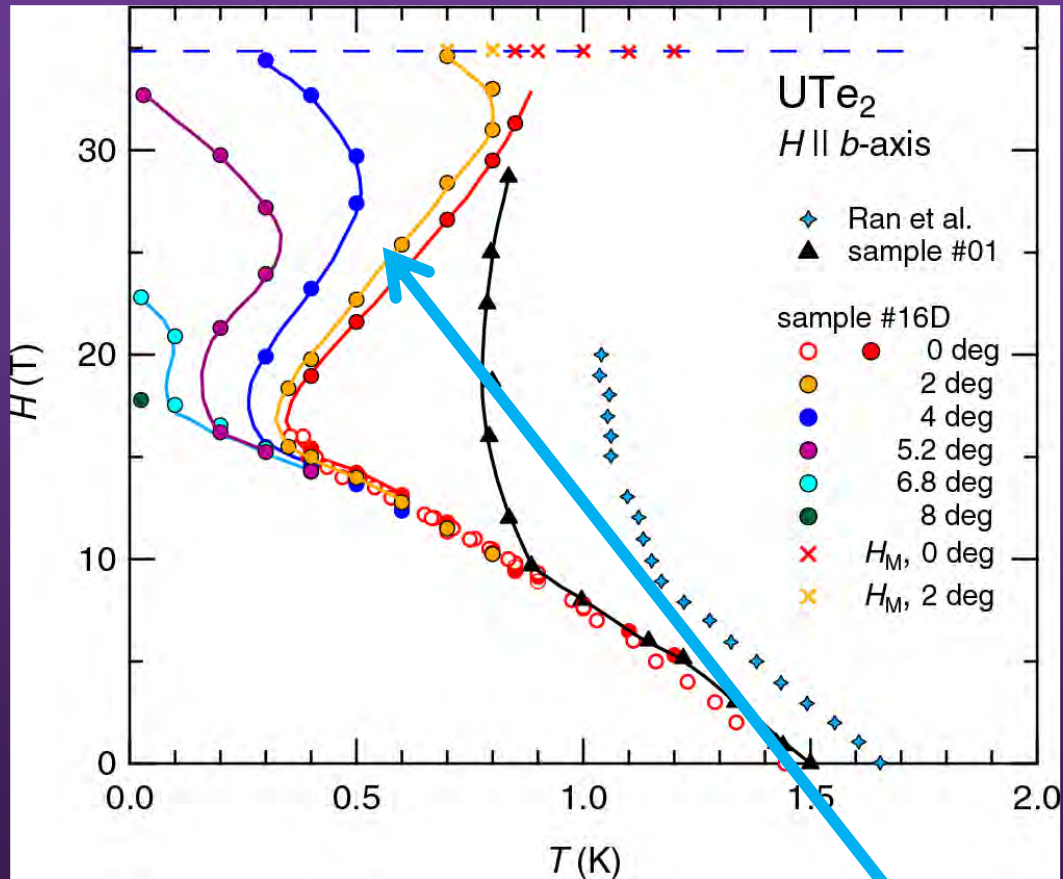
Imajo JPSJ 88, 083705 (2019)

Low-T effective mass enhancement

MAGNETISM?

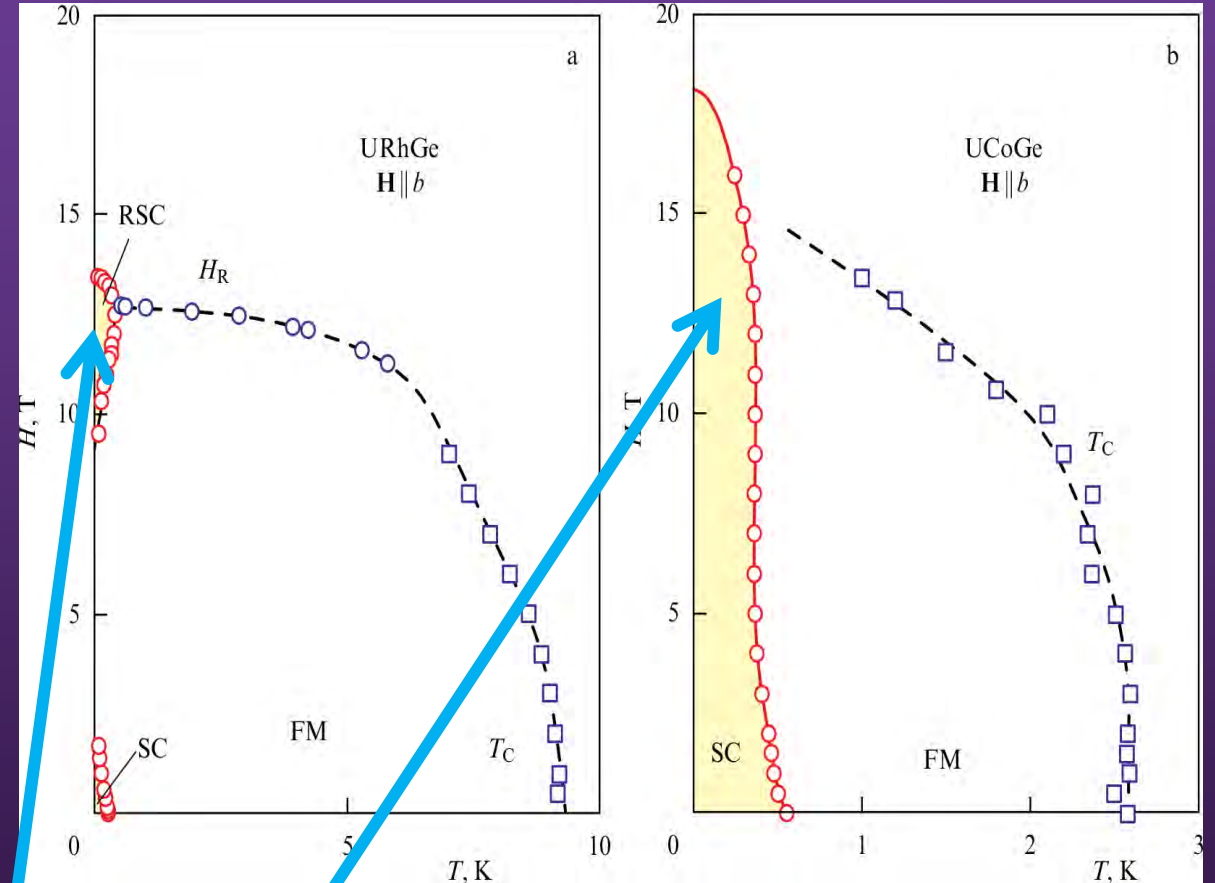
UTe₂ FIELD DEPENDENCE RESEMBLES FM SUPERCONDUCTOR

UTe₂ (paramagnet)



Knebel JPSJ 88, 063707 (2019)

URhGe and UCoGe (ferromagnets)

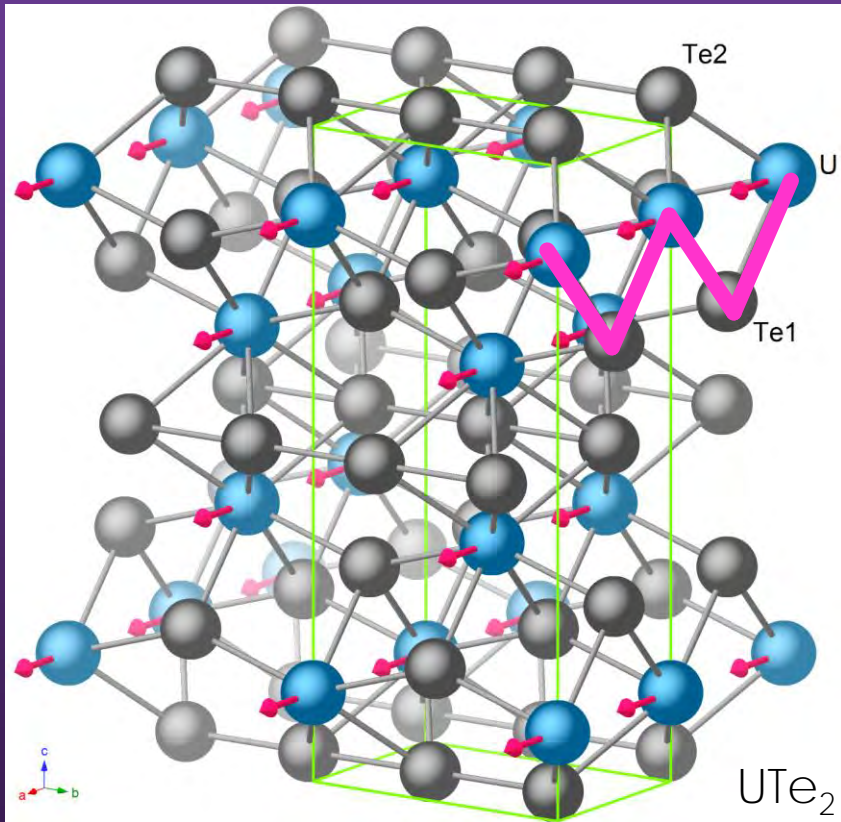


Mineev Phys.-Usp. 60, 121 (2017)

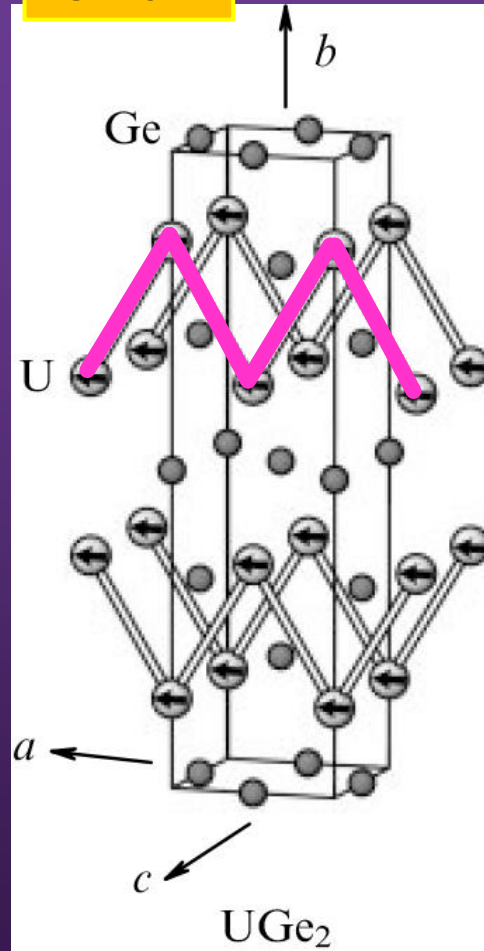
Reentrant Superconductivity

URANIUM CHAINS ALSO IN FM SUPERCONDUCTORS

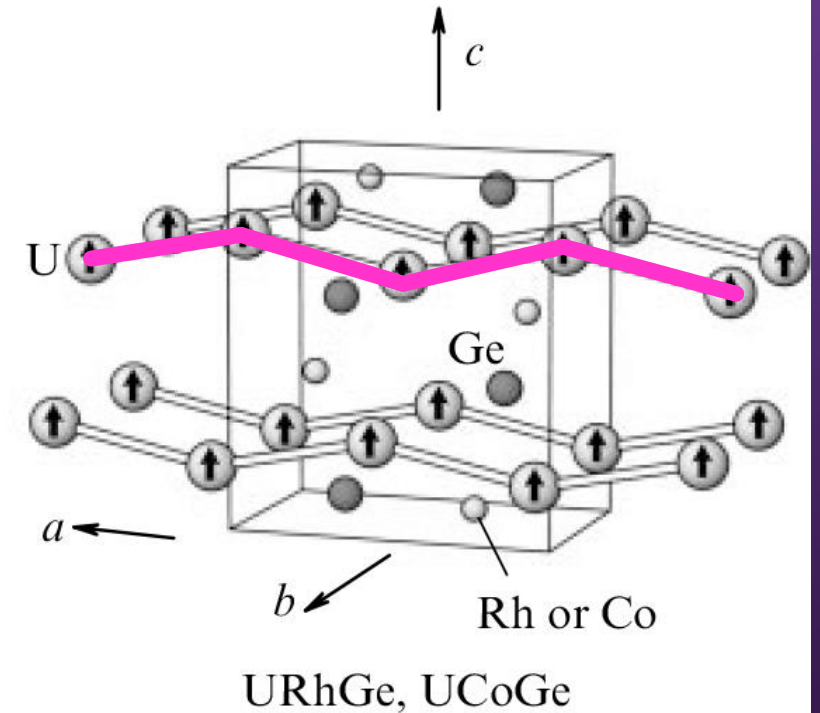
No magnetic order



$T_c = 52K$

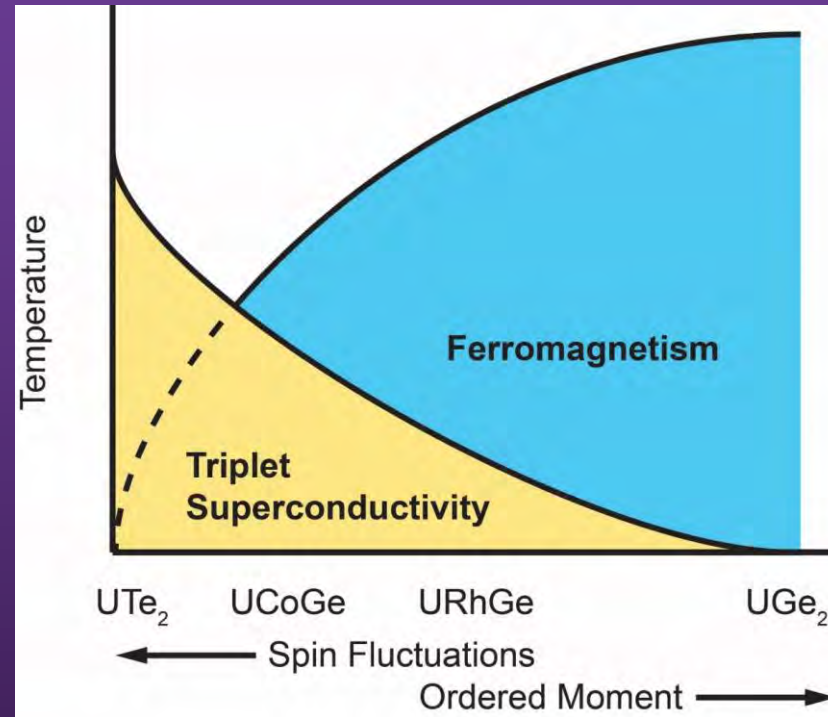


$T_c < 10K$



Mineev Phys.-Usp. 60 121 (2017)

IS UTe_2 THE END MEMBER OF THE FERROMAGNETIC SC'S?



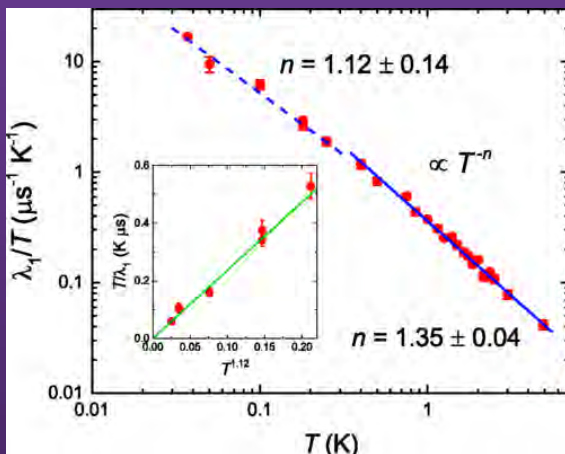
Ran Science 365, 684 (2019)

The equal spin pairing state... generically support[s] topologically protected surface Majorana arcs and bulk Weyl fermions as gapless excitations.

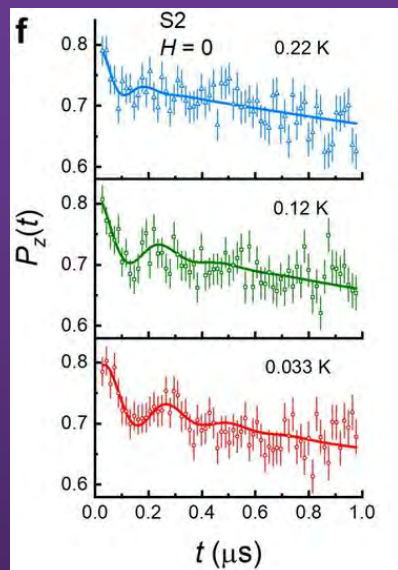
Sau & Tewari PRB 86, 104509 (2012)

UTe₂ FERROMAGNETIC CORRELATIONS

Muon spin relaxation

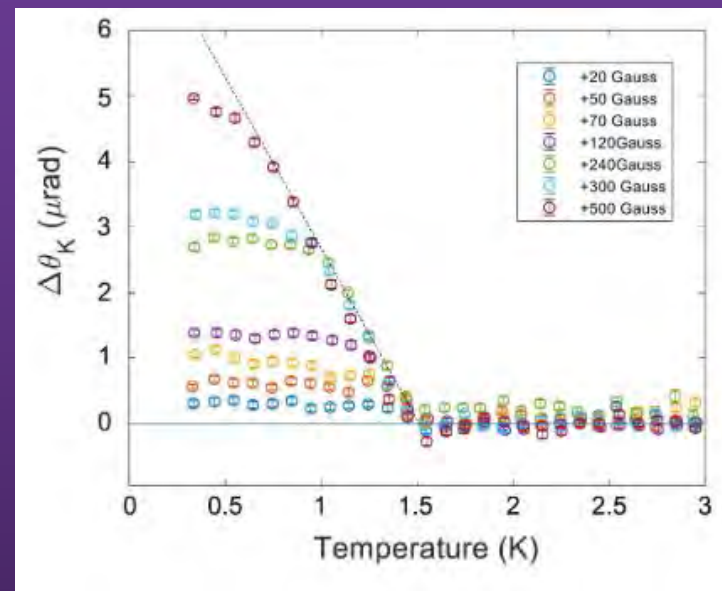


Sundar PRB 100, 140502(R) (2019)



Sundar Commun Phys 6, 24 (2023)

Optical Kerr rotation



Wei, PRB 105, 024521 (2022)

Relaxation time consistent with 3D FM quantum criticality



Oscillations from magnetic short-range order at low T (Not seen in neutron scattering)

Strongly polarized normal state → near FM instability

However, recent muSR → no magnetic clusters

Azari PRL 131, 226504 (2023)

However, recent Kerr → inhomogeneous magnetic regions

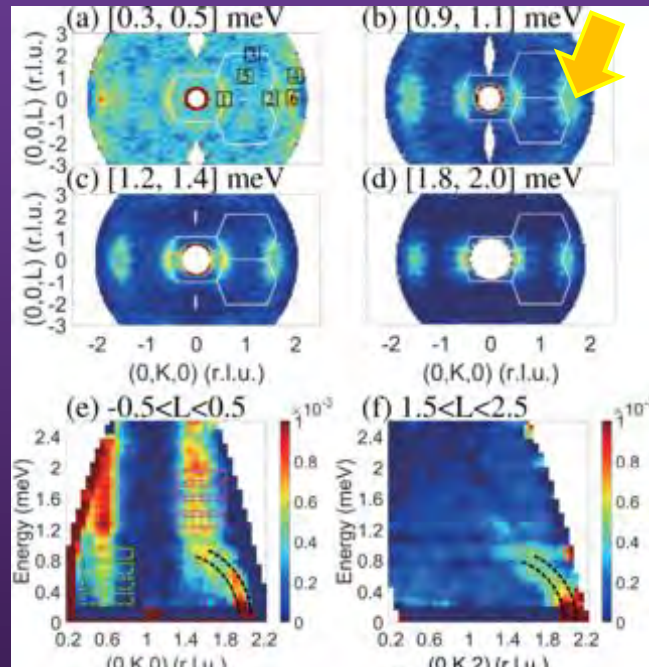
Ajeesh PRX 13, 041019 (2023)



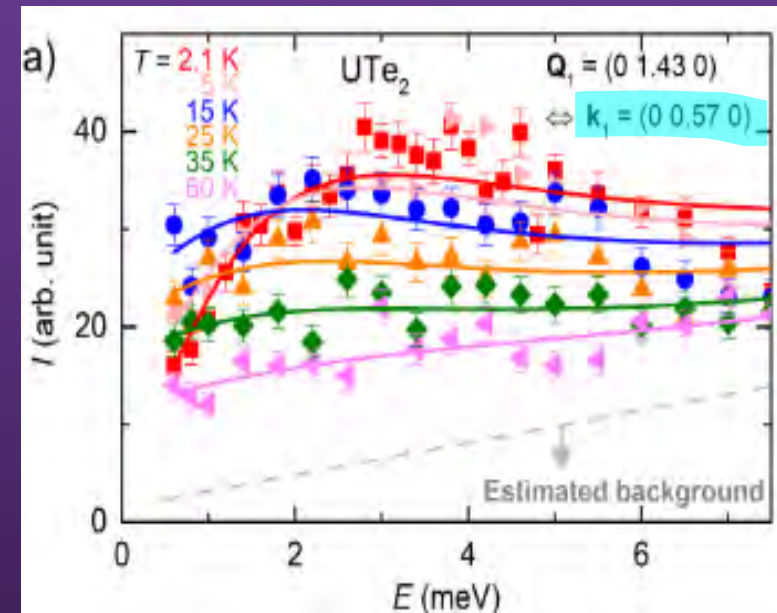
UTe₂ INELASTIC NEUTRON SCATTERING – AFM CORRELATIONS?

No FM-like inelastic signal near Bragg peaks

Excitations at incommensurate momentum transfer
Therefore (?) antiferromagnetic spin fluctuations



Duan et al, PRL 125, 237003 (2020)

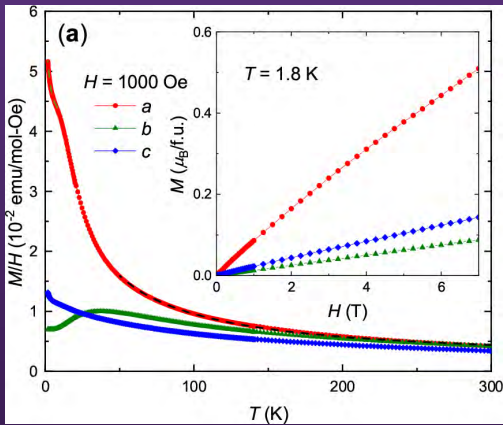


Knafo et al, PRB 104, L100409 (2021)

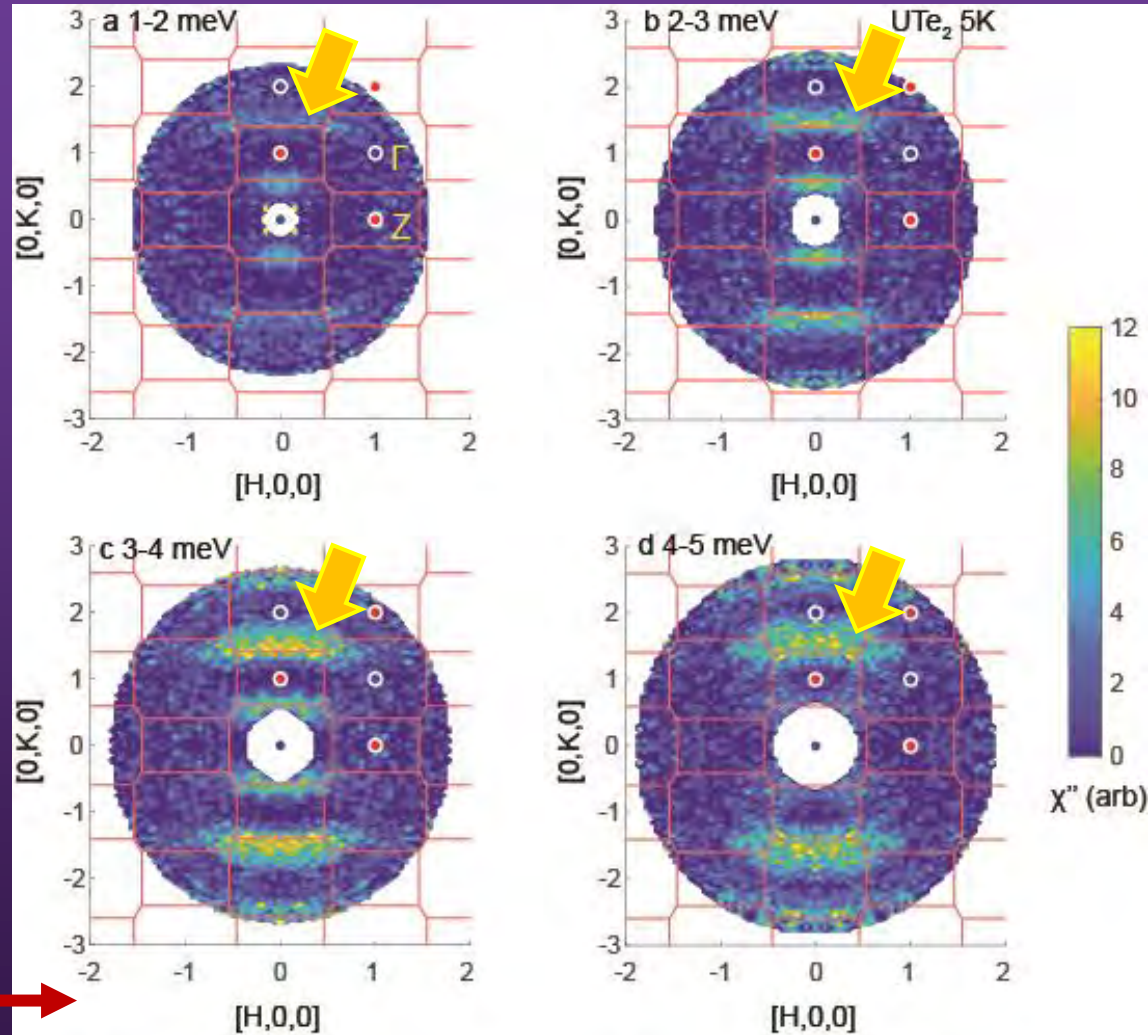
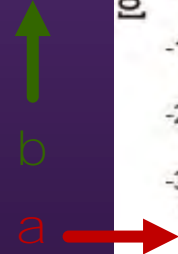
NOTE: THERE IS NO ANTIFERROMAGNETIC ORDER & THESE EXCITATIONS ARE NOT QUASIELASTIC

INELASTIC NEUTRON SCATTERING, A-B PLANE

Butch et al, npjQM 7, 39 (2022)

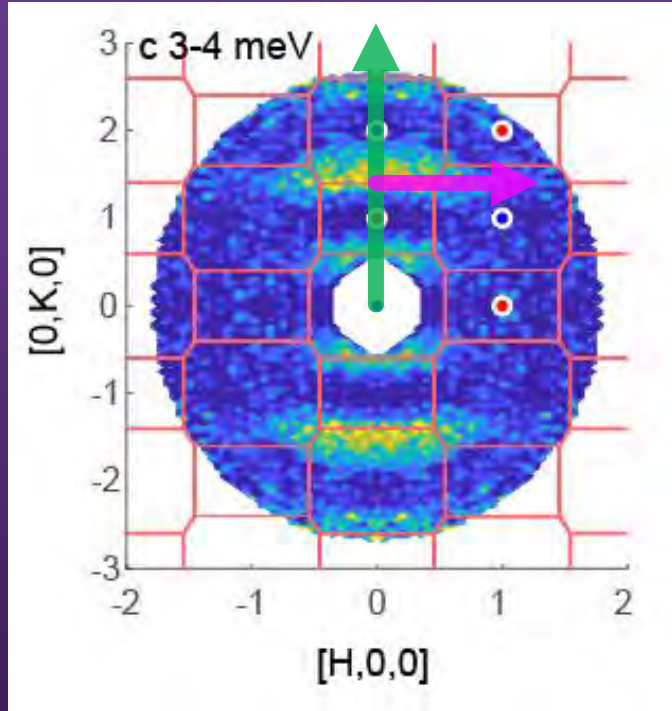


a is the magnetic easy axis
 b is perpendicular to "chains"



Scattering only at incommensurate Q along b -axis

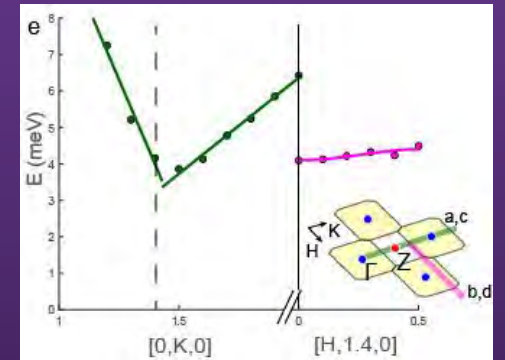
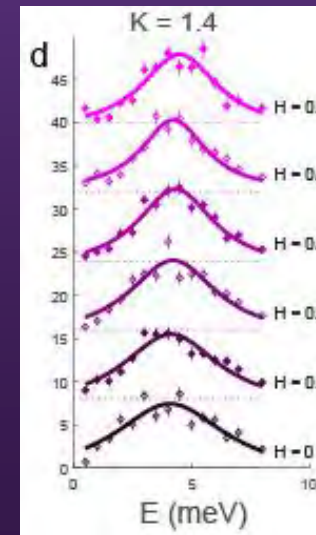
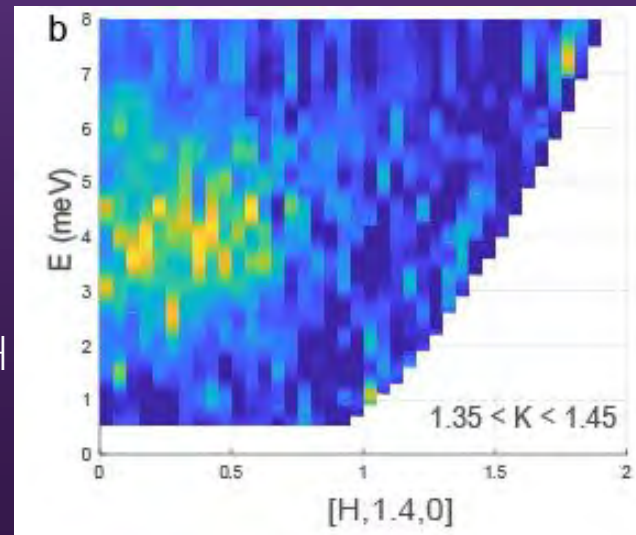
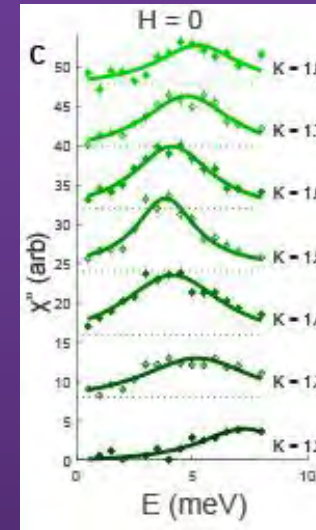
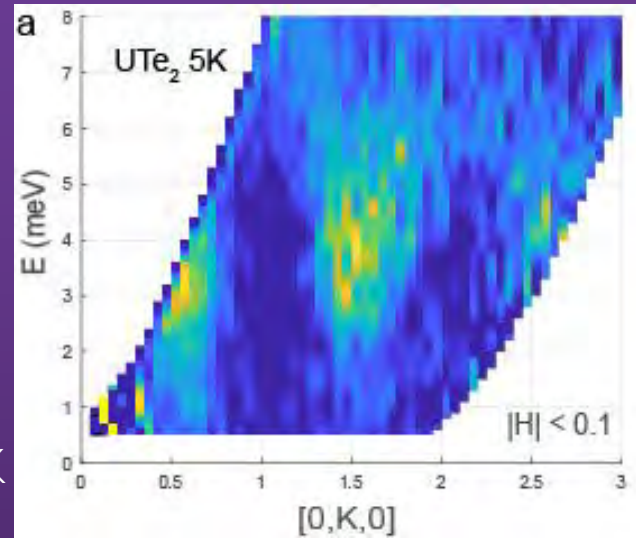
EXCITATIONS ARE DISPERSIVE AND ANISOTROPIC



Butch et al, npjQM 7, 39 (2022)

↑
Along K

→
Along H

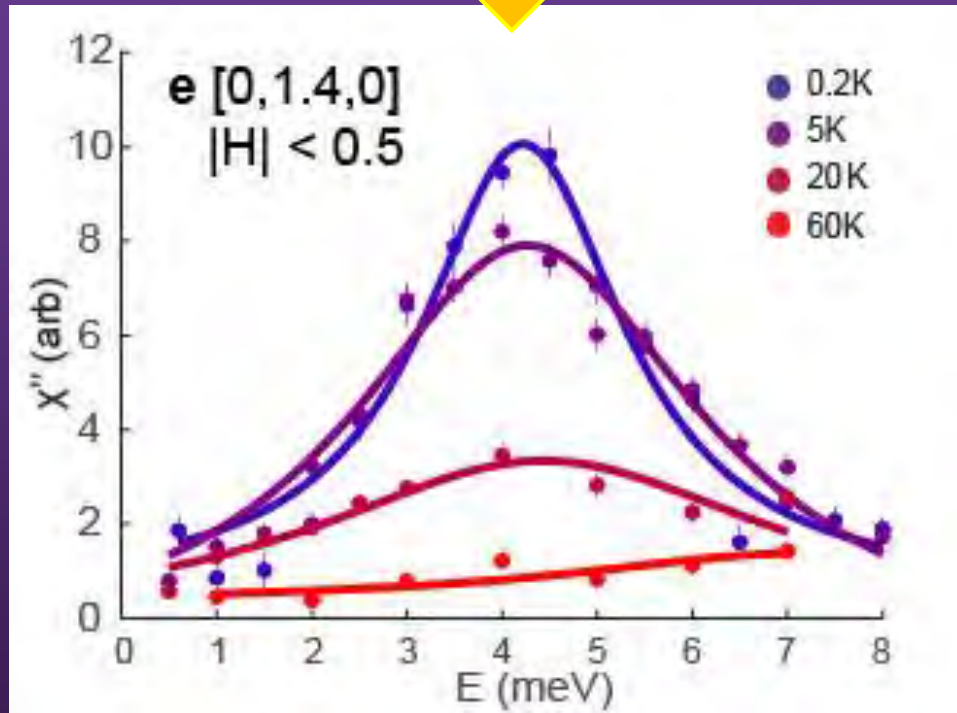


Anisotropic dispersion

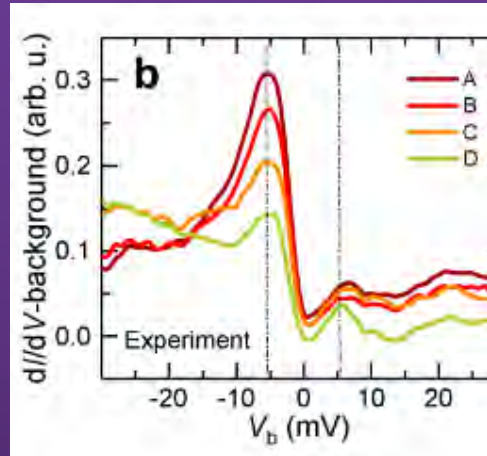
Not a typical property of paramagnetic excitations

COMPARE TO STM AND OPTICAL CONDUCTIVITY

Neutrons

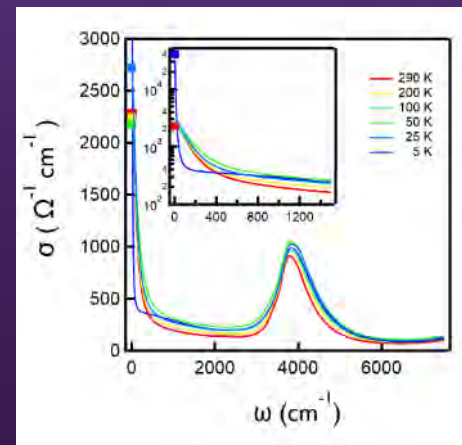


Butch et al, npjQM 7, 39 (2022)



Jiao et al, Nature 579, 523 (2020)

STM: 4 meV = 40 K hybridization gap



Mekonen PRB 106, 085125 (2022)

Optical conductivity:
Sharp Drude below 40K

THEORETICAL EXPLANATION: SCATTERING FROM ANDERSON LATTICE

Main feature: $S(Q,E)$ dispersive, inelastic peaks with energy minimum at BZ

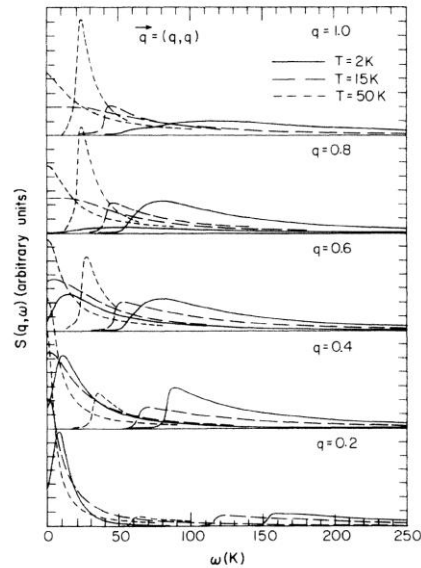


FIG. 3. $S(q, \omega)$ for square-lattice model, $\vec{q}=(q, q)$. Arbitrary units. Parameters similar to those of Fig. 1, as described in text. The contributions from χ_{++}^0 and χ_{-+}^0 are shown separately, as the left- and right-hand curves, respectively.

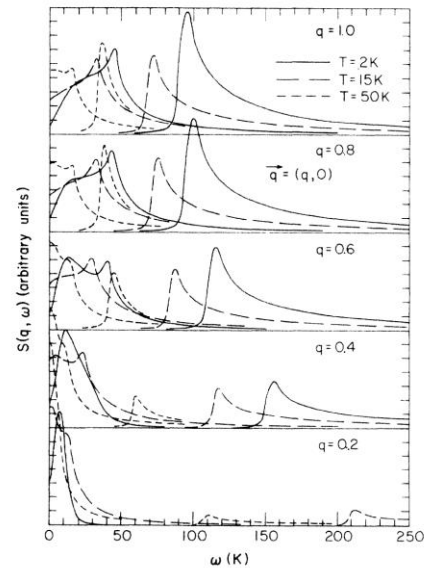


FIG. 4. $S(q, \omega)$ for $\vec{q}=(q, 0)$. Units and model parameters are the same as in Fig. 3.

Brandow PRB 37, 250 (1988)

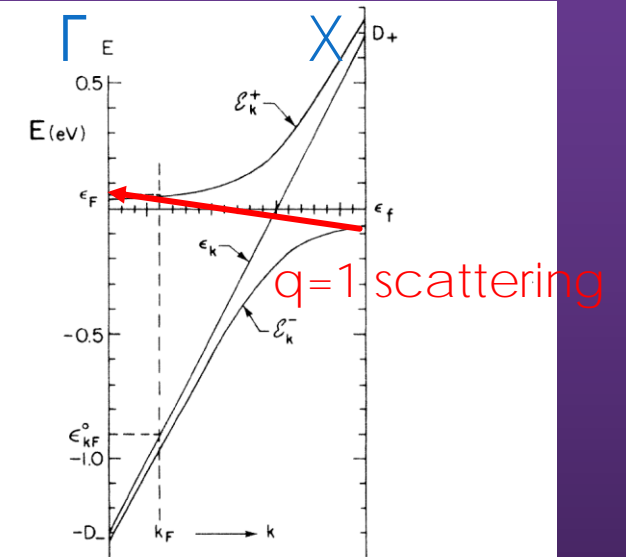
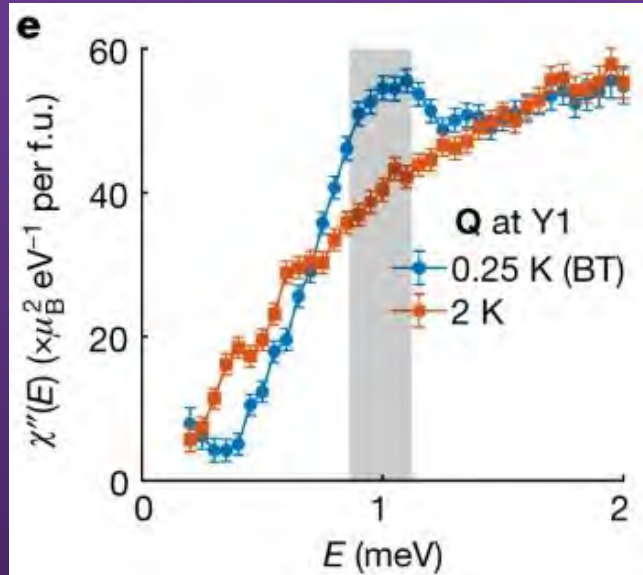


FIG. 1. Quasiparticle spectrum for $U=0$ case. Input parameters are $W=2.0$ eV, $V=-(0.5)^{1/2}$ eV, $D_- = 1.3$ eV, $\epsilon_{k_F}^0 = -0.9$ eV.

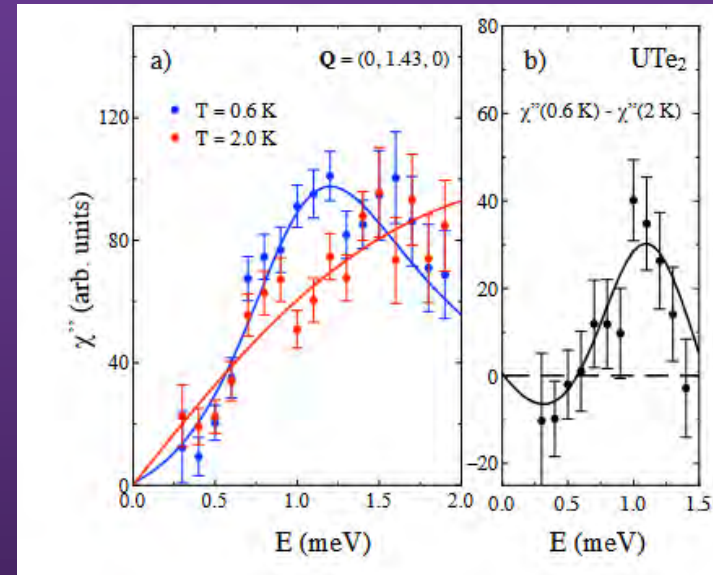
Brandow PRB 33, 215 (1986)

“One must conclude that this q dependence is not at all diagnostic for the presence of an antiferromagnetic interaction.” Brandow PRB 37, 250 (1988)

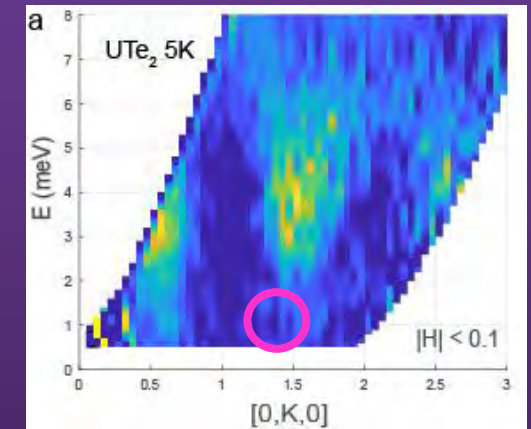
AFM CORRELATIONS, PART 2: SUPERCONDUCTIVITY



Duan et al, Nature 600, 636 (2021)



Raymond et al, JPSJ 90, 113706 (2021)



Butch et al, npjQM 7, 39 (2022)

Low-E tail of excitation

In SC state, a change near 1 meV at “AFM” momentum transfer

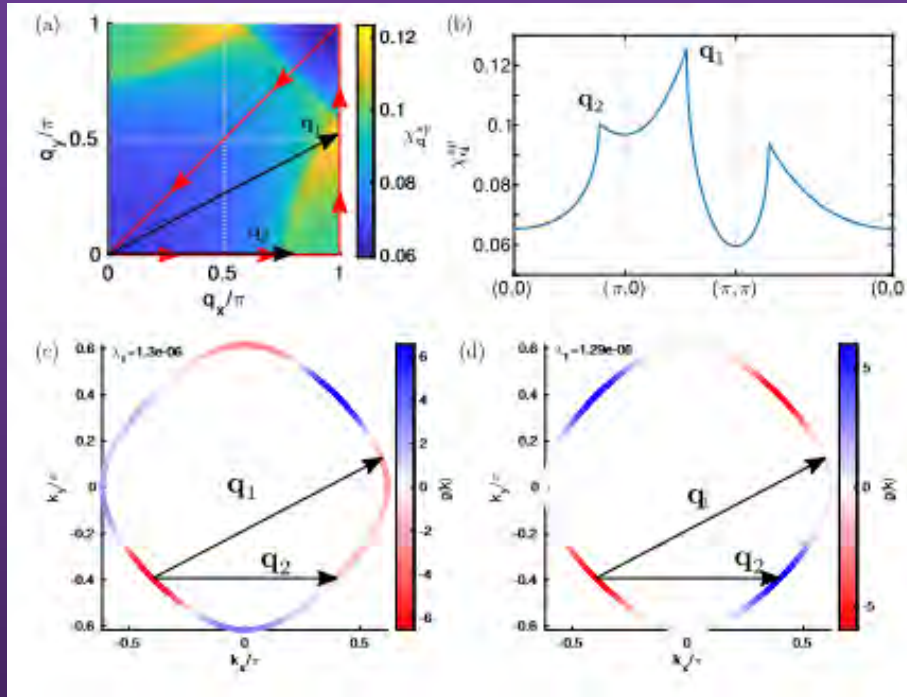


Why \gg gap energy?

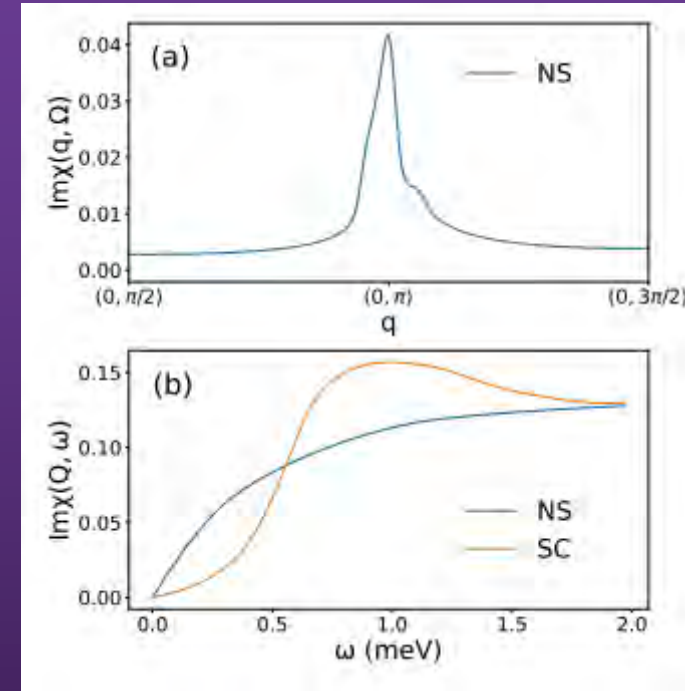


Opposite spin \rightarrow singlet?

DOMINANT FM SUSCEPTIBILITY IS NOT REQUIRED FOR SPIN-TRIPLET



Kreisel, Quan, Hirschfeld, PRB 105, 104507 (2022)

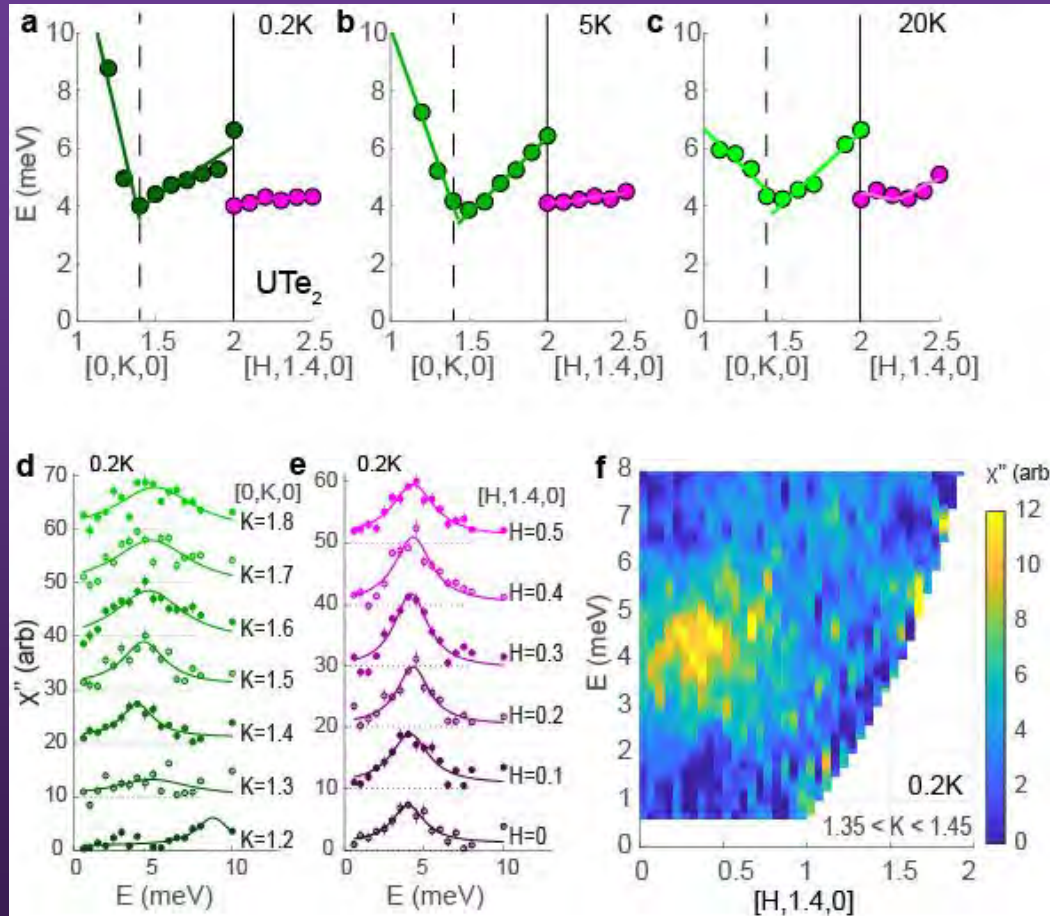


Chen et al, arXiv:2112.14750

Kreisel: “a strong peak at larger q in the magnetic susceptibility can drive [triplet pairing]”

Chen: “multiorbital spin-triplet pairing ... naturally yields a spin resonance at the antiferromagnetic wavevector”

THE DISPERSION IS SUBTLY TEMPERATURE-DEPENDENT



Butch et al, npjQM 7, 39 (2022)

- SMALL DIFFERENCES BETWEEN SC AND NORMAL STATE

- DISPERSION
- INTENSITY

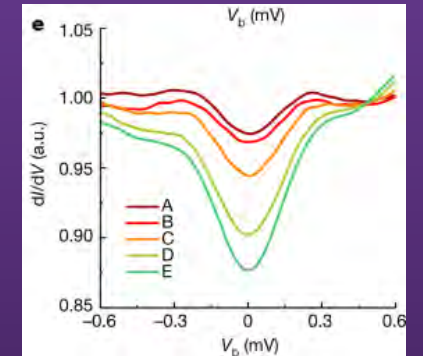
- ENERGY SCALE

- OF ORDER HYBRIDIZATION
- MUCH GREATER THAN SUPERCONDUCTIVITY

- DO NOT DETECT 1 meV RESONANCE

- $1 \text{ meV} \gg 0.25 \text{ meV SC GAP}$

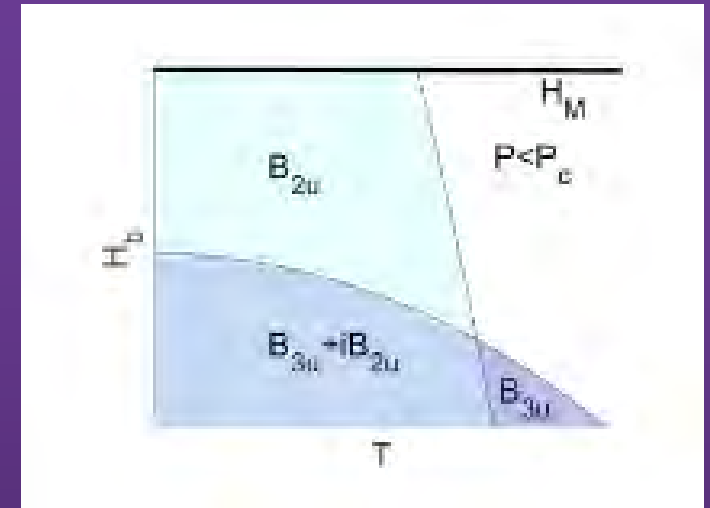
Jiao et al, Nature 579, 523 (2020)



EVIDENCE FOR UNCONVENTIONAL SUPERCONDUCTIVITY

ORDER PARAMETER THEORY FOR UTe_2

- $B_{3U}+iB_{2U}$ (SHISHIDOU, HAYES) WEYL POINTS FOR $k_x=0$ OR $k_y=0$
- $B_{1U}+iB_{3U}$ (NEVIDOMSKYY)
- B_{3U} OR A_U (ISHIZUKA, YANASE)
- B_{3U} (NAKAMINE 2021)
- EQUAL SPIN PAIRING (YARZHEMSKY, TEPLYAKOV)
- $B_{3U}+iA_U$ (ISHIHARA 2021)



Shishidou PRB 103, 104504 (2021)

Irrep	E	C_{2z}	C_{2y}	C_{2x}	linear	quadratic $[\psi(\mathbf{k})]$	$\vec{d}(\mathbf{k})$	nodes
A_{1g}	1	1	1	1	-	k_x^2, k_y^2, k_z^2	-	-
B_{1g}	1	1	-1	-1	H_z	$k_x k_y$	-	line
B_{2g}	1	-1	1	-1	H_y	$k_x k_z$	-	line
B_{3g}	1	-1	-1	1	H_x	$k_y k_z$	-	line
A_u	1	1	1	1	-	-	$\hat{x}k_x, \hat{y}k_y, \hat{z}k_z$	-
B_{1u}	1	1	-1	-1	k_z	-	$\hat{x}k_y, \hat{y}k_x, \hat{z}k_x k_y k_z$	point
B_{2u}	1	-1	1	-1	k_y	-	$\hat{x}k_z, \hat{y}k_x k_y k_z, \hat{z}k_x$	point
B_{3u}	1	-1	-1	1	k_x	-	$\hat{x}k_x k_y k_z, \hat{y}k_z, \hat{z}k_y$	point

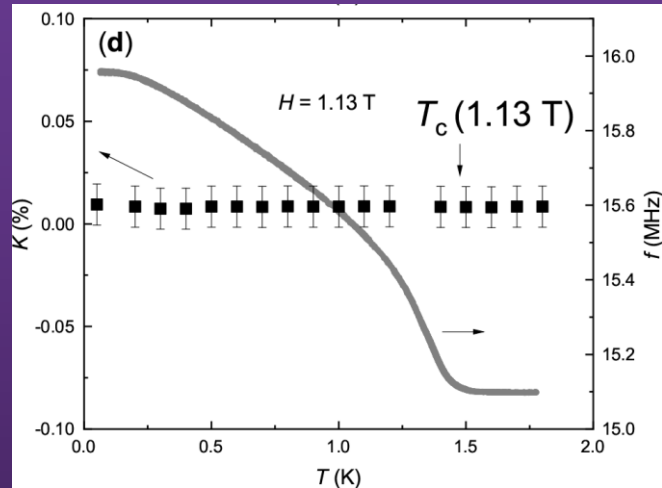
Table I. Irreducible representations and representative functions for point group D_{2h} .

Hayes et al, Science 373, 797 (2021)

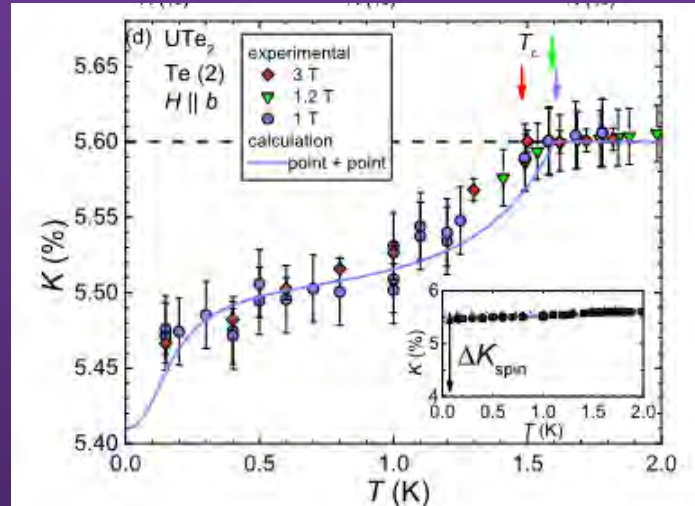
UTe₂ ¹²⁵Te NMR EVIDENCE FOR SPIN TRIPLET PAIRING

NMR Knight shift: spin susceptibility of the electrons forming Cooper pairs

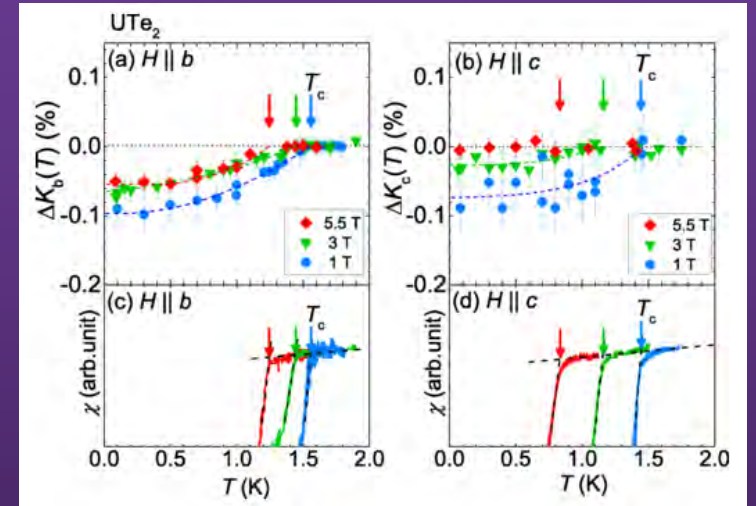
Ding & Furukawa, Ames Lab



Ran Science 365, 684 (2019)



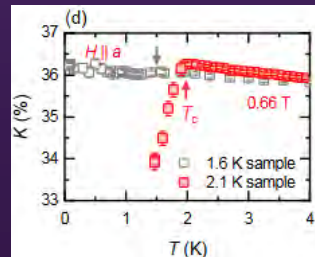
Nakamine JPSJ 88, 113703 (2019)



Nakamine PRB 103, L100503 (2021)

Powder NMR: no change in Knight shift through T_c triplet pairing & $1/T_1 \sim T^6$ (nodes)

Single crystal NMR: tiny change in Knight shift through T_c , consistent with triplet

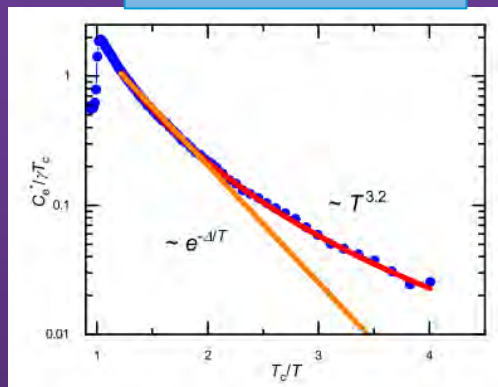


Recent (2.1K Tc): larger a-axis Knight shift → nodeless

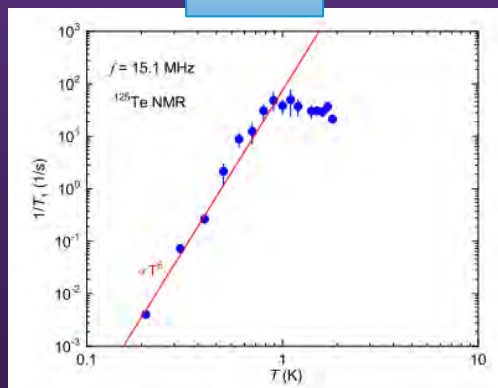
Matsumura JPSJ 92, 063701 (2023)

EVIDENCE FOR SC GAP POINT NODES

Heat capacity

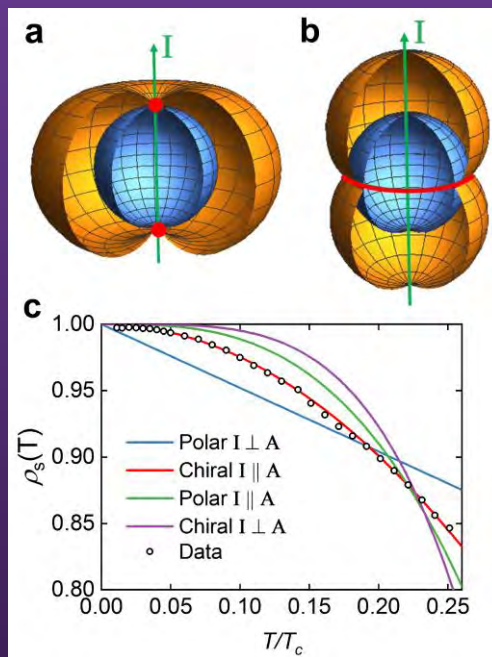


NMR

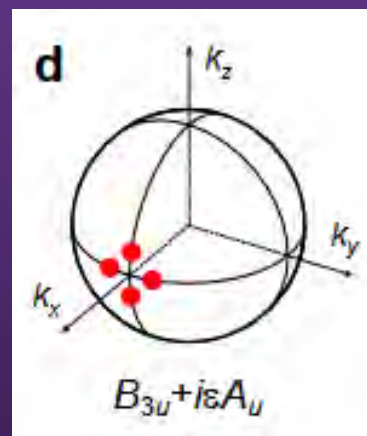


Ran Science 365, 684 (2019)

Penetration depth

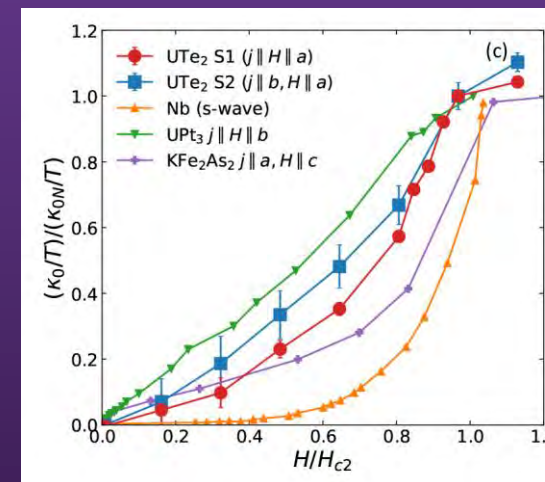


Bae Nature Comm. 12, 2644 (2021)



Ishihara Nature Comm 14, 2966 (2023)

Thermal conductivity

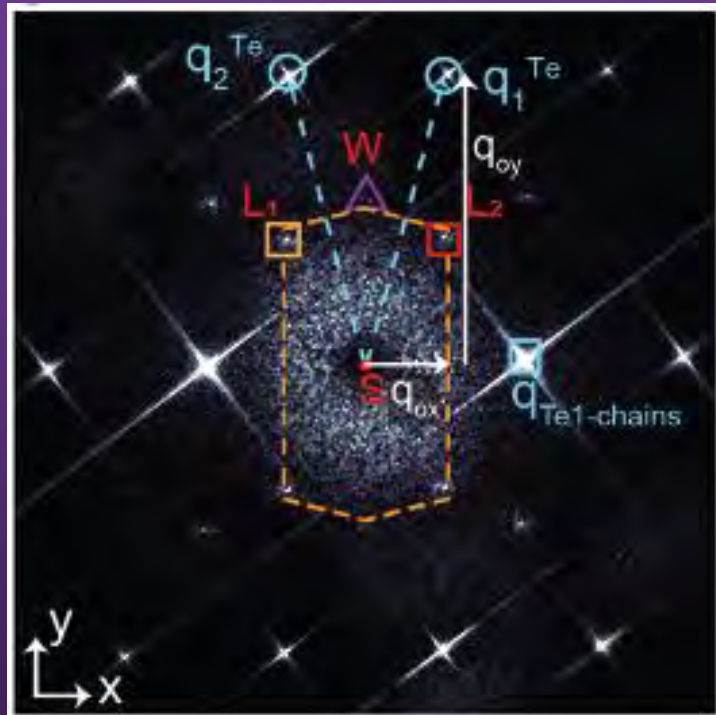


Metz Phys. Rev. B 100, 220504(R) (2019)

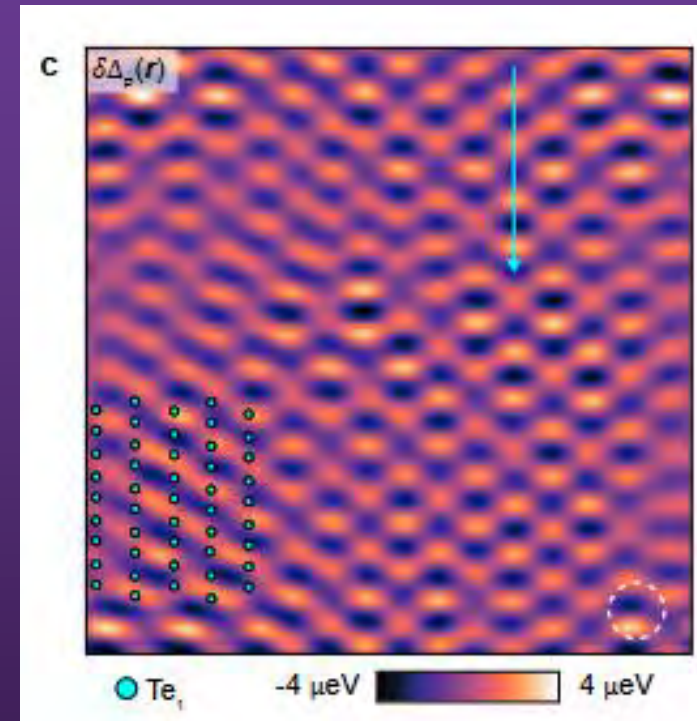
STM: RECENT SIGHTINGS OF PAIR DENSITY WAVE

PDW = superconductor with periodic spatial variation of order parameter and zero average

See Agterberg et al, Annual Reviews of CMP 11, 231 (2020)



Aishawara Nature 618, 928 (2023)



Gu Nature 618, 921 (2023)

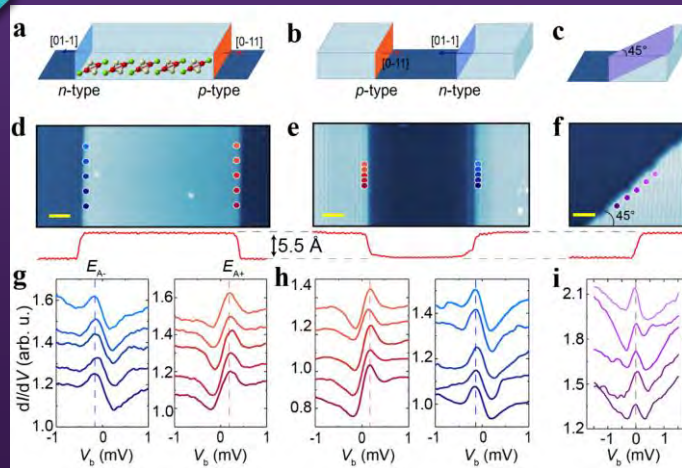
STM: Charge Density Wave in normal state
Pair Density Wave in superconducting state
PDW demonstrated in helium-3 and in cuprates

IS IT A TOPOLOGICAL SUPERCONDUCTOR?

For topological superconductivity (nodal spin triplet)

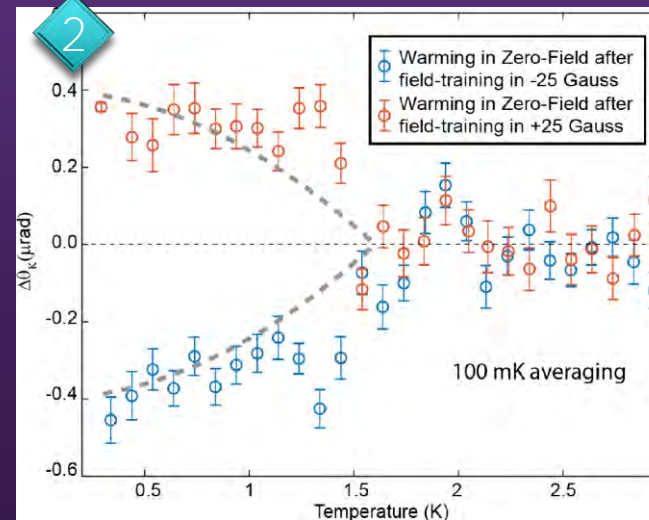
1. CHIRAL SURFACE STATES (MAJORANA)
2. TIME REVERSAL SYMMETRY BREAKING (SPONTANEOUS MOMENT)
3. 2-COMPONENT, COMPLEX ORDER PARAMETER

1



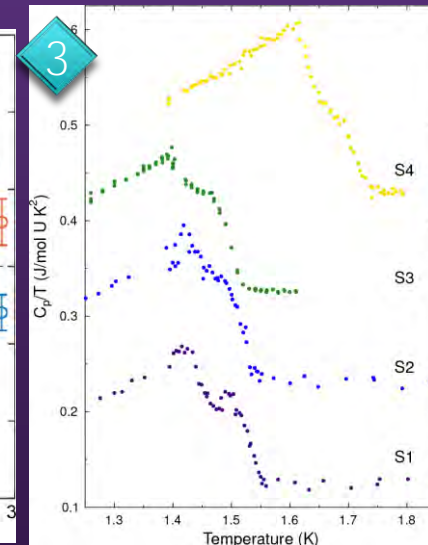
Jiao et al, Nature 579, 523 (2020)

2



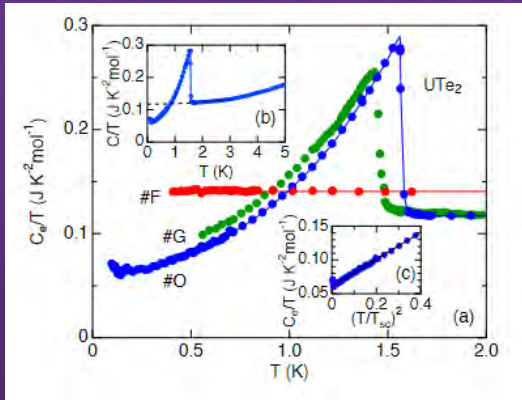
Hayes et al, Science 373, 797 (2021)

3

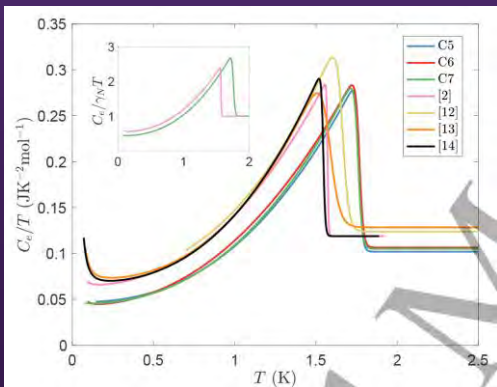


ARGUMENTS AGAINST 2 TRANSITIONS

Variations in T_c –
Not all samples superconduct

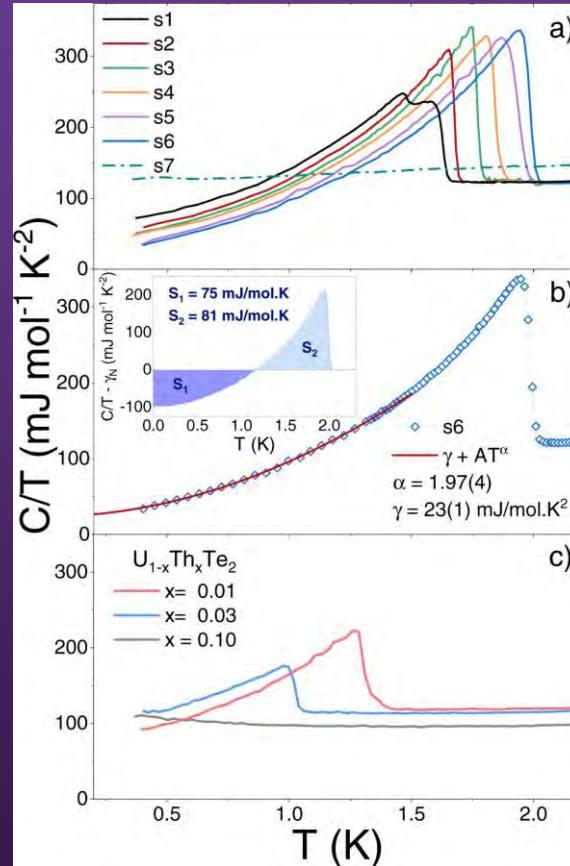


Aoki JPSJ 88, 043702 (2019)



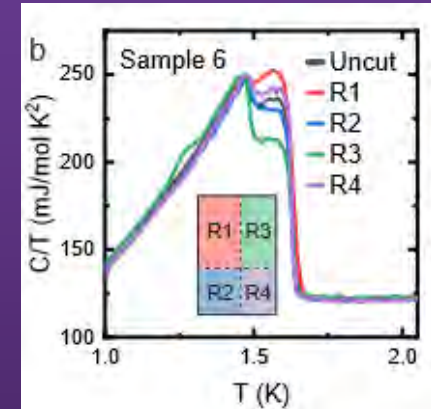
Cairns JPCM 32 415602 (2020)

Higher T_c - only one transition

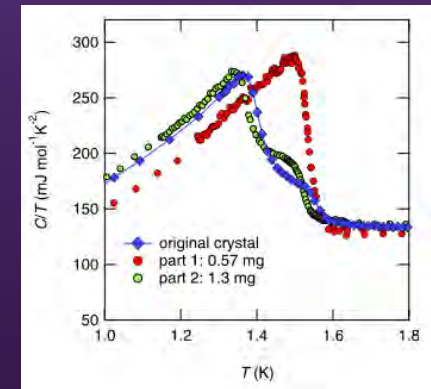


Rosa Commun Mater 3, 33 (2022)

Inhomogeneous superconductivity



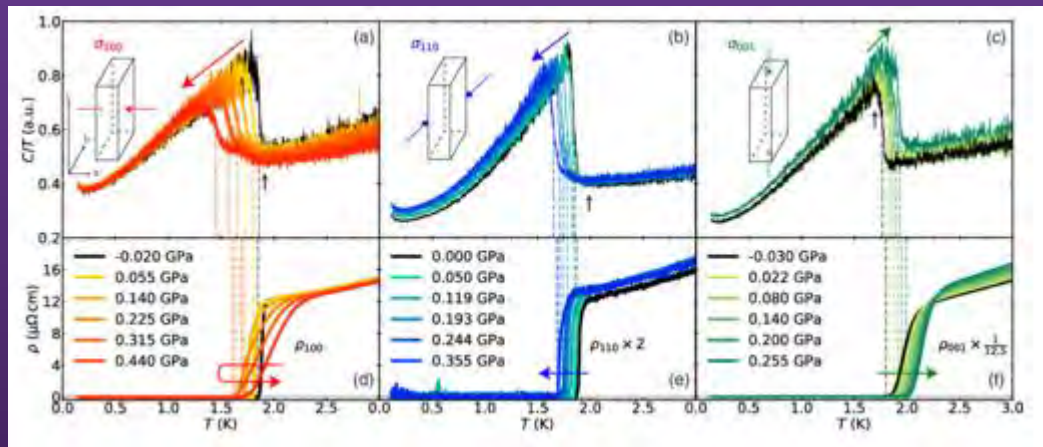
Thomas PRB 104, 224501 (2021)



Aoki JPCM 34, 243002 (2022)

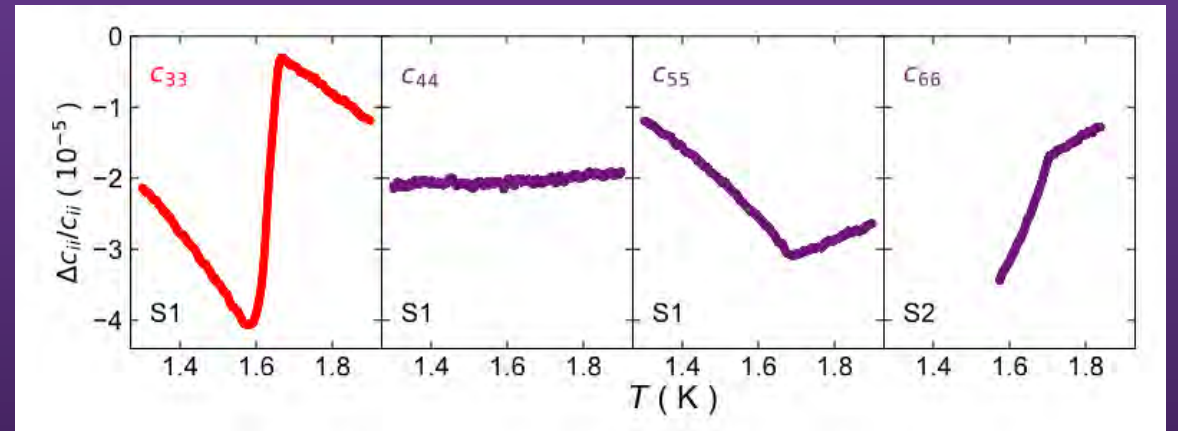
MORE ARGUMENTS FOR SINGLE ORDER PARAMETER

110 (shear) stress – no splitting



Girod PRB 106, L121101 (2022)

Echo ultrasound – weak anomalies in shear modes (vs compression)

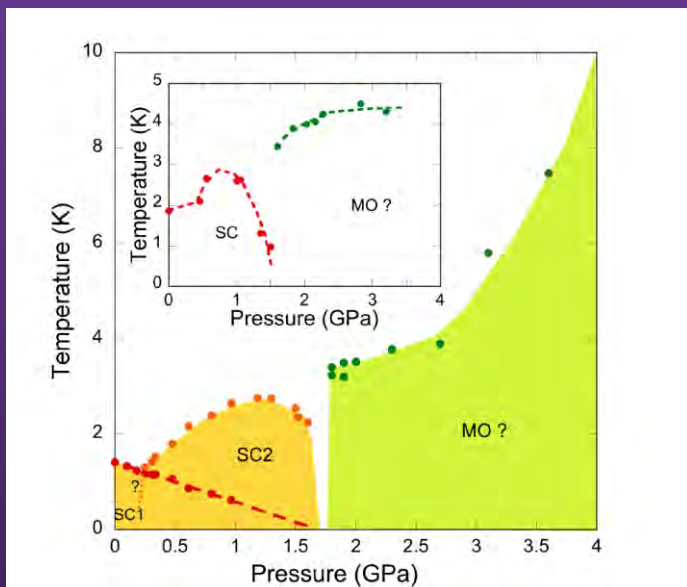


Theuss arXiv:2307.10938

Spontaneous Time Reversal Symmetry breaking??
It requires a 2-component order parameter

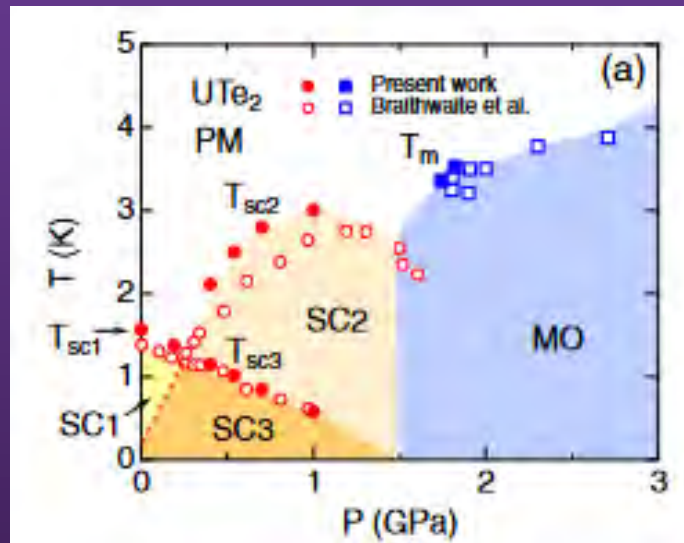
BUT... 2 (OR MORE) SC TRANSITIONS UNDER PRESSURE

2 SC phases



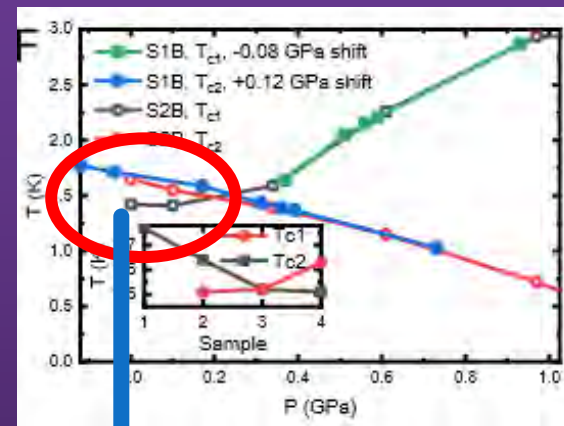
Braithwaite Comm Phys 2, 147 (2019)

More than 2 SC phases

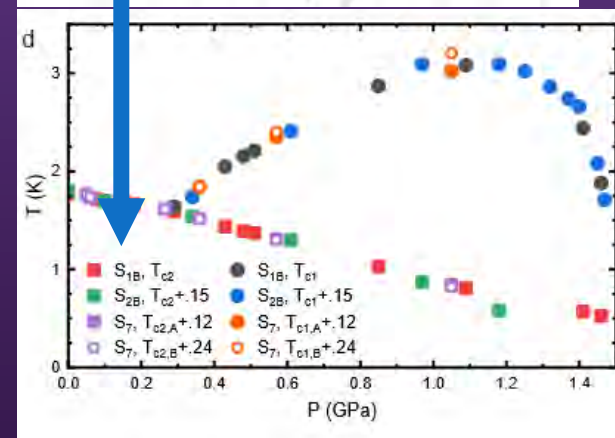


Aoki, JPSJ 89, 053705 (2020)

Are there 2 phases at low pressure?



v2



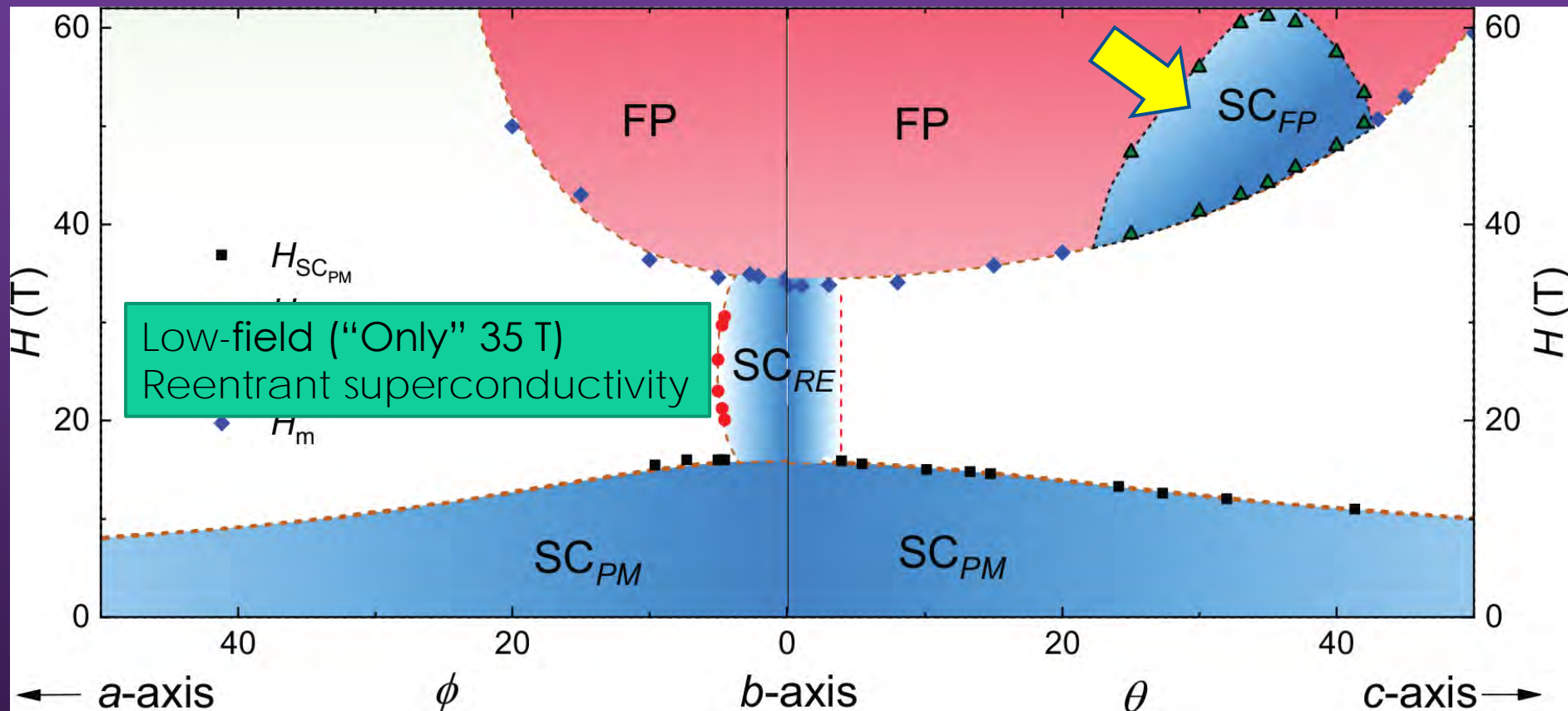
v3

Thomas PRB 104, 224501 (2021)

HIGH FIELD

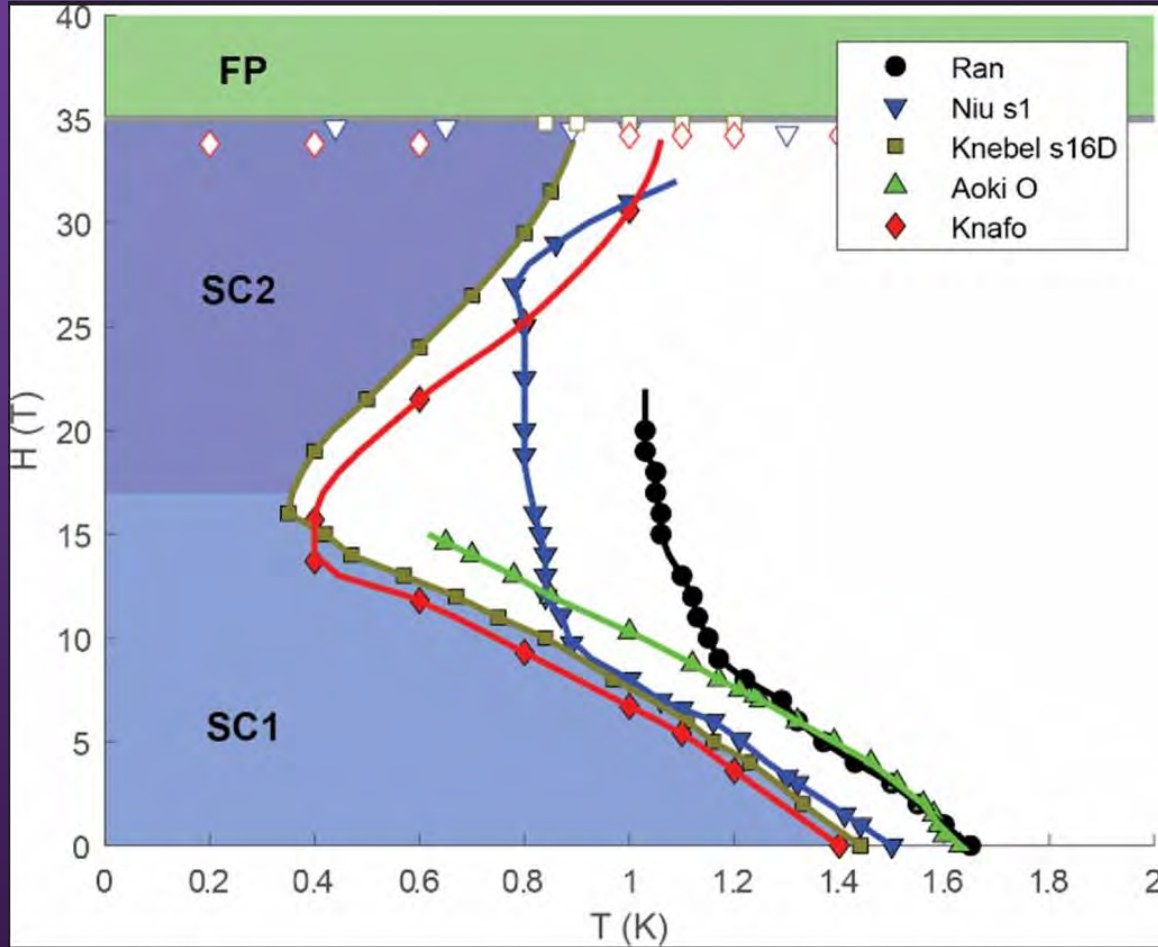
SUPERCONDUCTIVITY BEYOND 60 T!

Lazarus superconductivity- Highest field reentrant phase



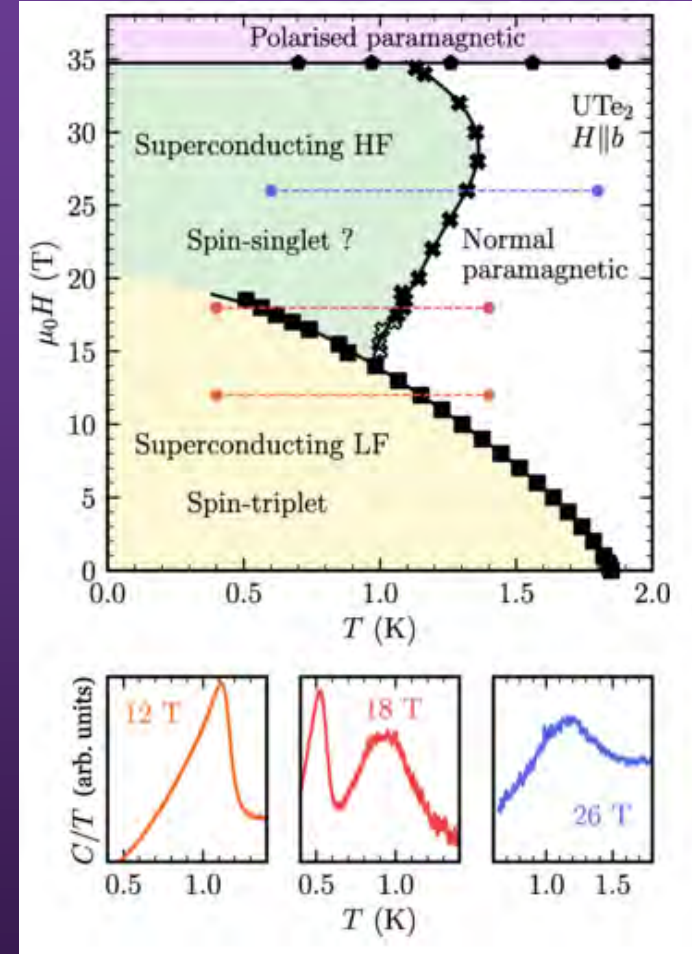
Ran Nature Physics 15, 1250 (2019)

B-AXIS SUPERCONDUCTIVITY: 2 PHASES



Lewin, Rep. Prog. Phys. 86 114501 (2023)

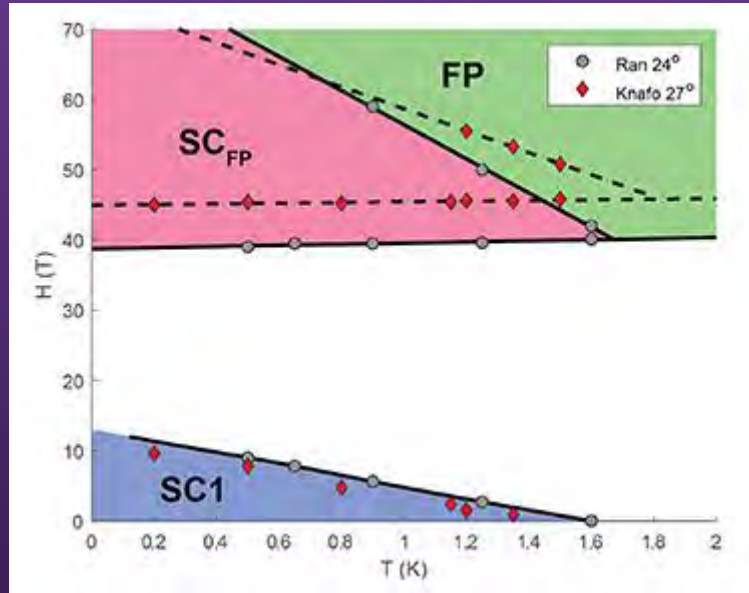
Anomalies in heat capacity



Rosuel PRX 13, 011022 (2023)

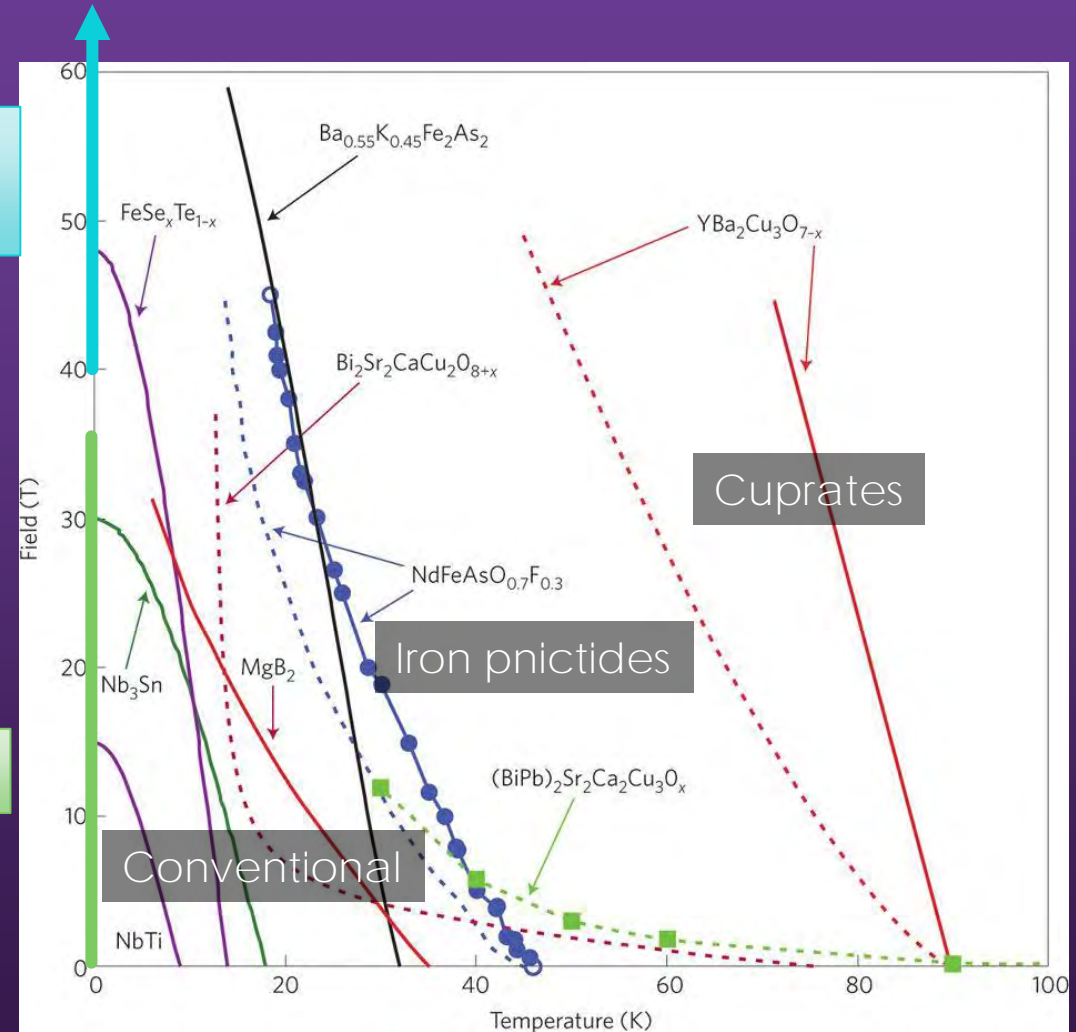
UTe₂ HIGH FIELD SUPERCONDUCTIVITY IS DIFFERENT

UTe₂
Lazarus phase



Lewin, Rep. Prog. Phys. 86 114501 (2023)

UTe₂

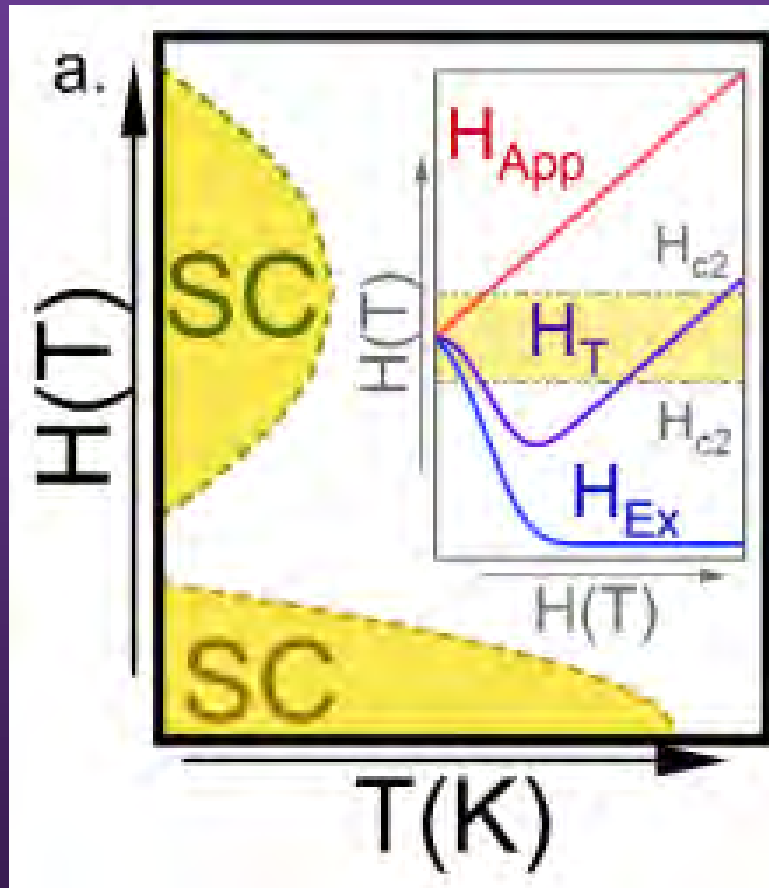


Gurevich, Nature Mater. 10, 255 (2011)

MECHANISMS FOR SC AT HIGH MAGNETIC FIELDS

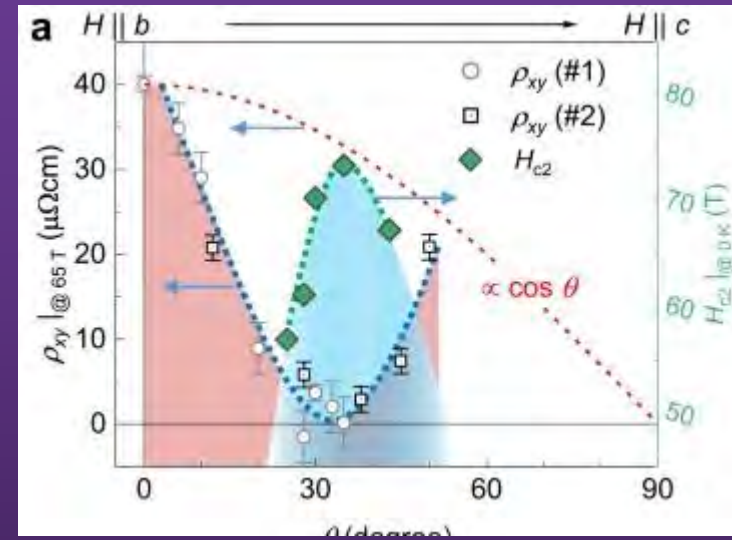
- MINIMIZE ZEEMAN COUPLING
 - REDUCED G-FACTOR
 - SPIN TRIPLET
- COMPENSATE APPLIED FIELD
 - INTERNAL EXCHANGE FIELD (JACCARINO PETER EFFECT)
- WEAKEN ORBITAL DEPAIRING
 - LOW DIMENSIONALITY - ELIMINATES ORBITAL MOTION
 - LANDAU-LEVEL ENHANCEMENT
- STRENGTHEN PAIRING
 - MAGNETIC FLUCTUATIONS AT QUANTUM CRITICAL POINT

MAGNETIC FIELD COMPENSATION (JACCARINO-PETER)



Frank arXiv:2304.12392

Argue for JP mechanism based on Hall data

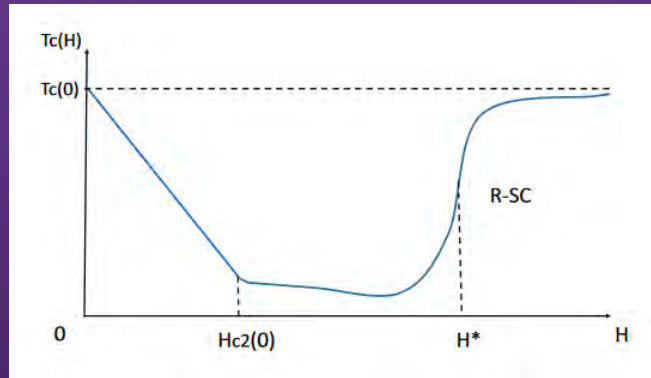


Helm Nature Comm 15, 37 (2024)

Requires local moments
 Exchange field opposes applied field
 Acts on the spins, not the orbits
 Rather sensitive to angle (organic SCs)

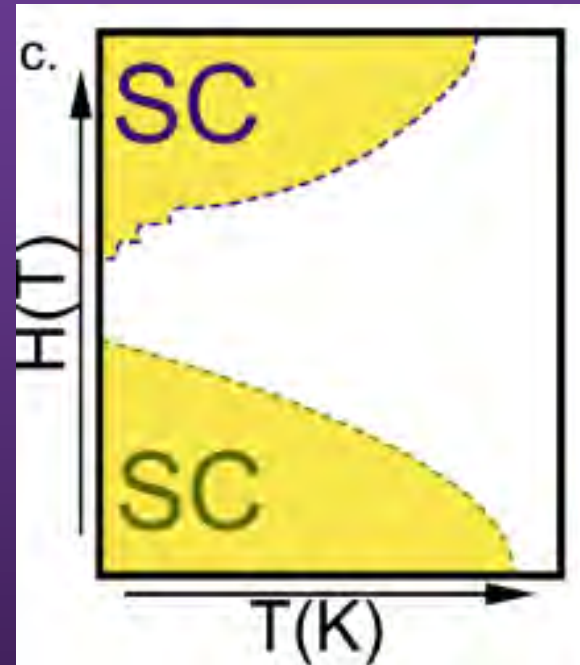
LOW-DIMENSIONALITY / LANDAU LEVEL

Low D
e amplitude < interplane distance



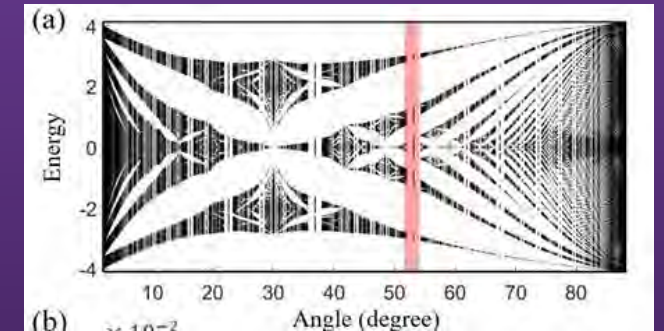
Lebed Mod. Phys. Lett. B 34 2030007 (2020)

Landau Level
LL Quantization enhances H_{c2}



Frank arXiv:2304.12392
After Rasolt & Tesanovic

Field-induced Hofstadter Butterfly
Tilted magnetic fields enhance T_c



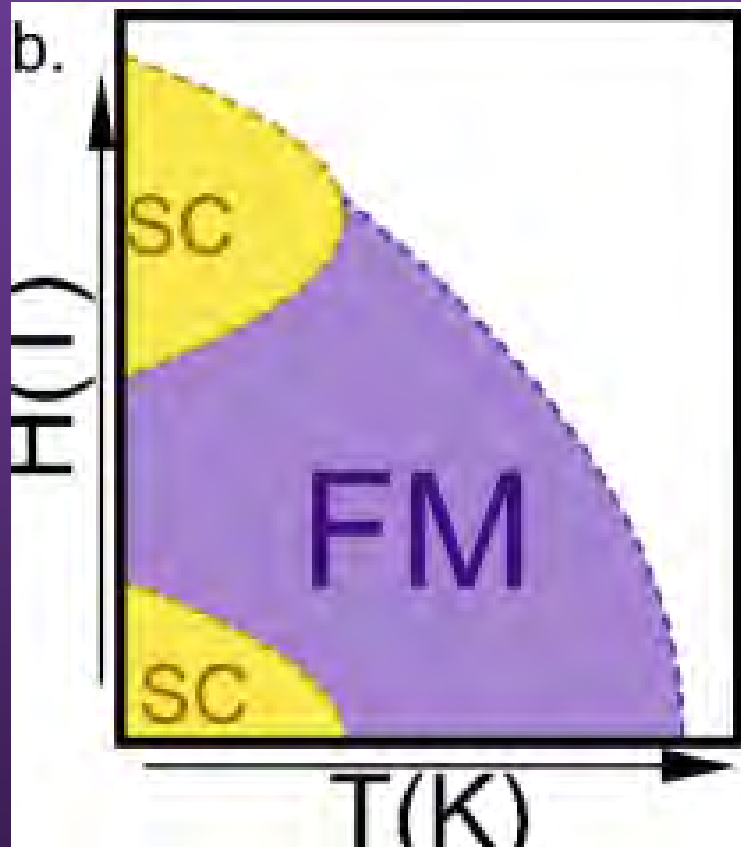
Park arXiv:2007.16205

Is UTe_2 electronic structure 2D (enough)?

Oscillatory upper critical field?

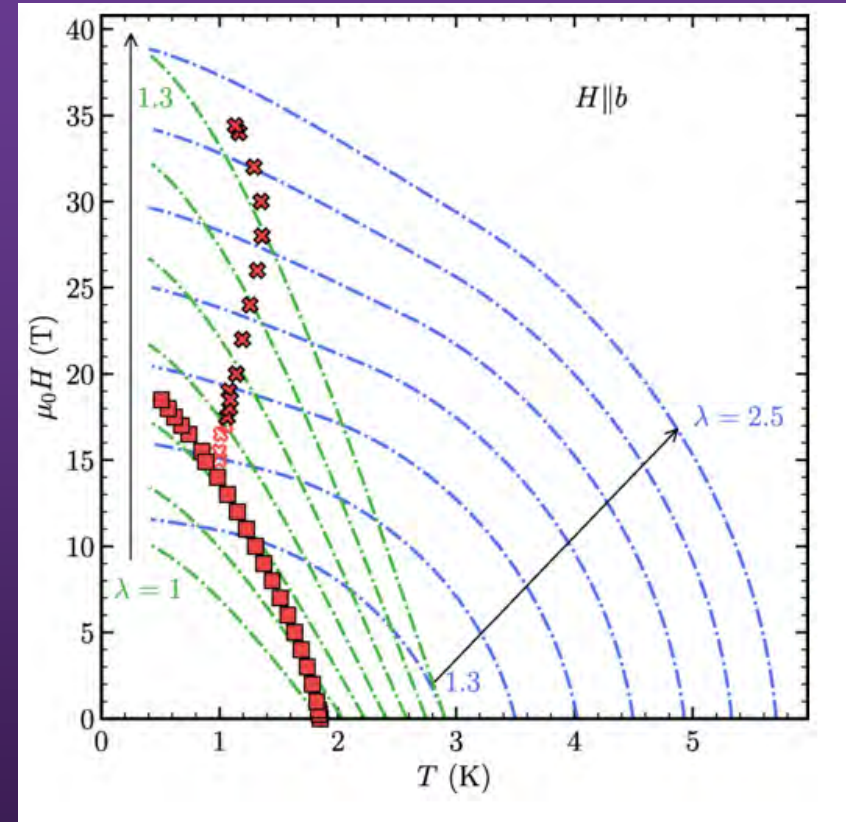
Proposed to explain Lazarus

PAIRING ENHANCEMENT



Frank arXiv:2304.12392

Series of H_{c2} curves with different coupling

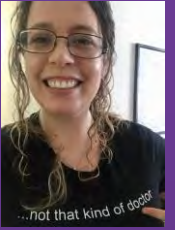


Rosuel PRX 13, 011022 (2023)

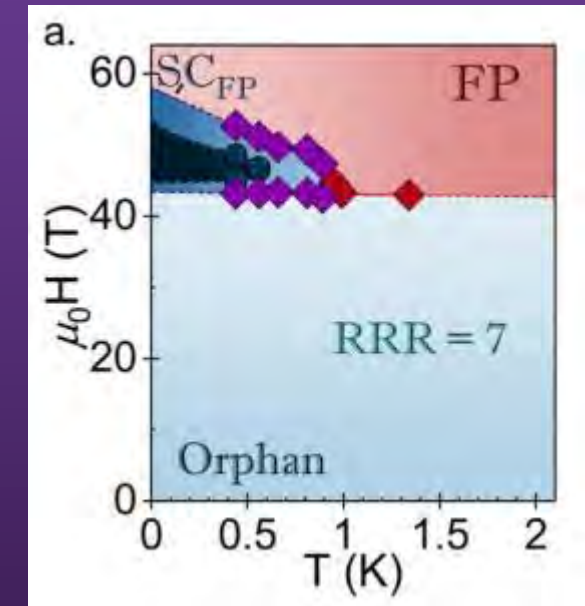
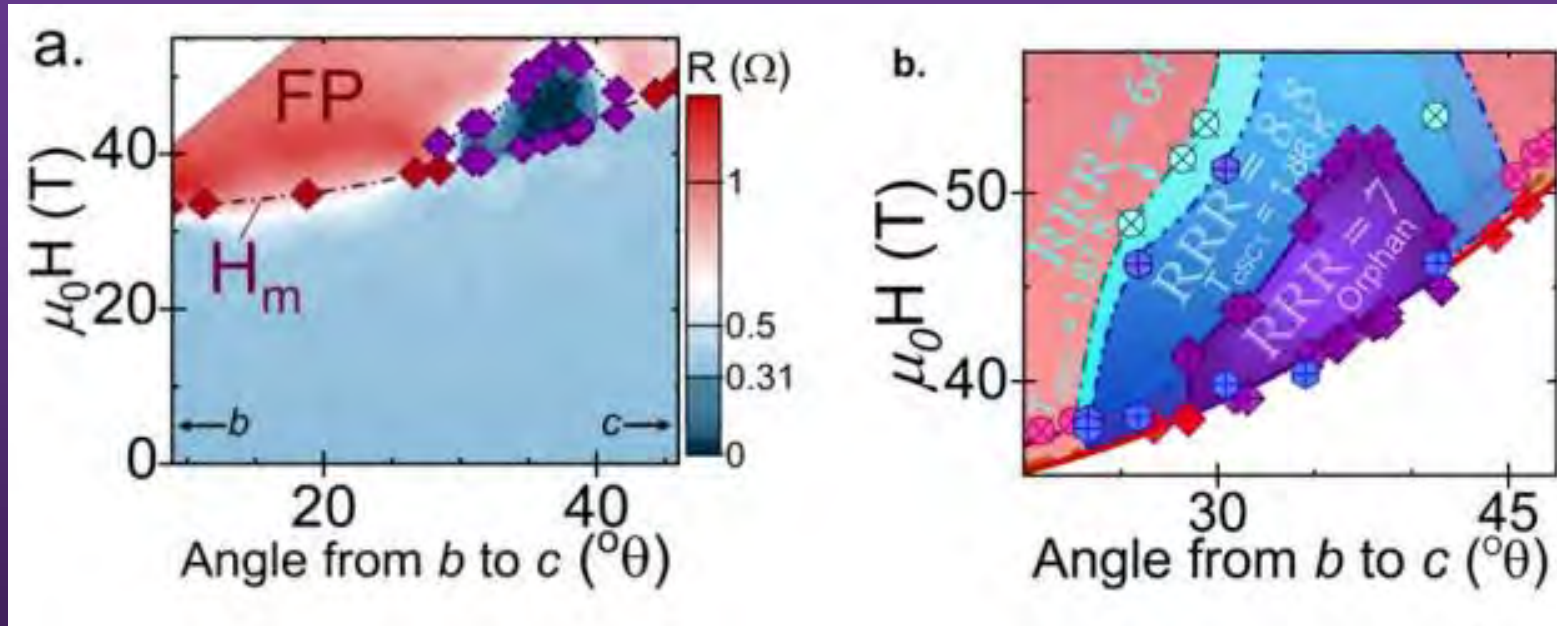
Believed relevant for FM SCs like URhGe, but no similar QCPs in UTe_2

LAZARUS BECOMES AN ORPHAN

High field SC is more stable than zero-field SC



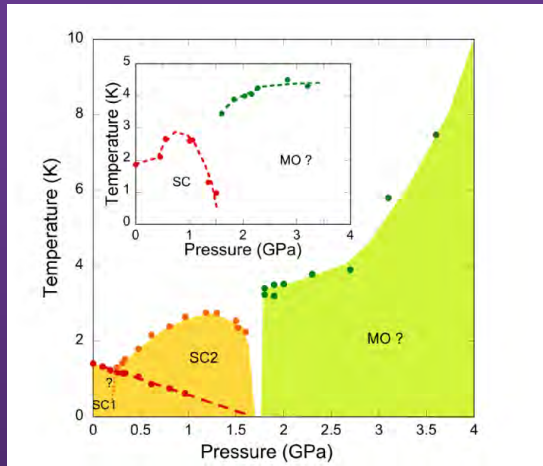
Frank arXiv:2304.12392



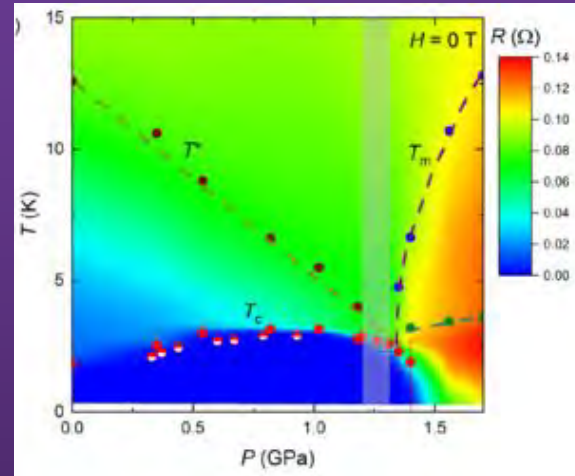
Disorder can kill low-field parent superconductivity
 But in a certain range of disorder, Lazarus phase survives! Why is it more robust?

PRESSURE AND HIGH FIELD

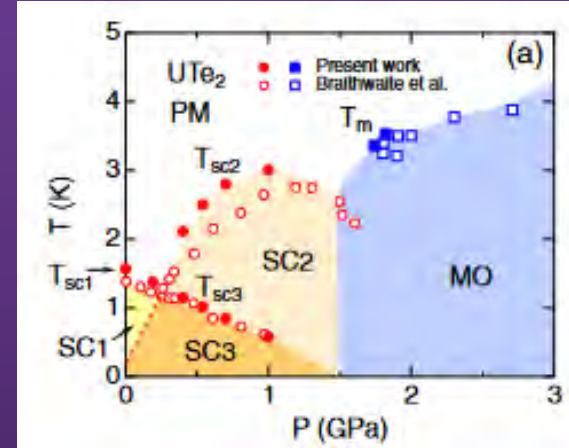
UTe₂ UNDER PRESSURE



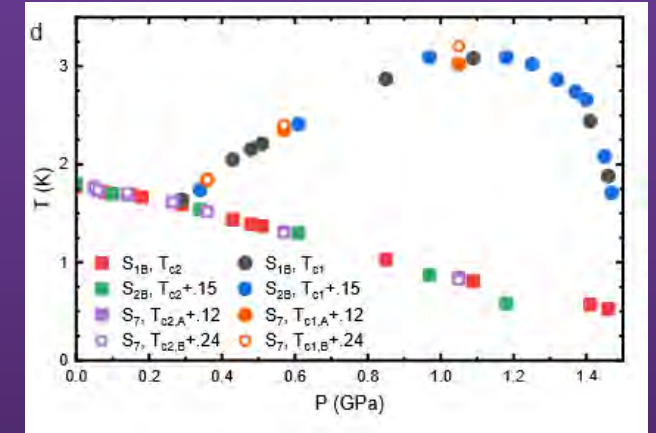
Braithwaite Commun Phys 2, 147 (2019)



Ran et al, PRB 101, 140503(R) (2020)



Aoki, JPSJ 89, 053705 (2020)

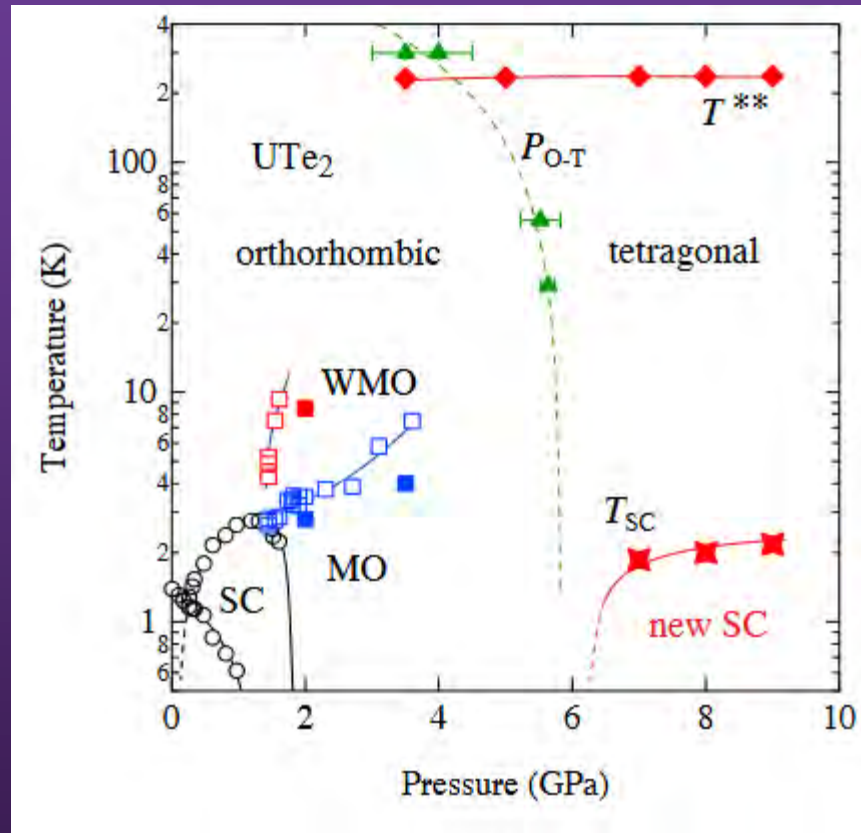


Thomas PRB 104, 224501 (2021)

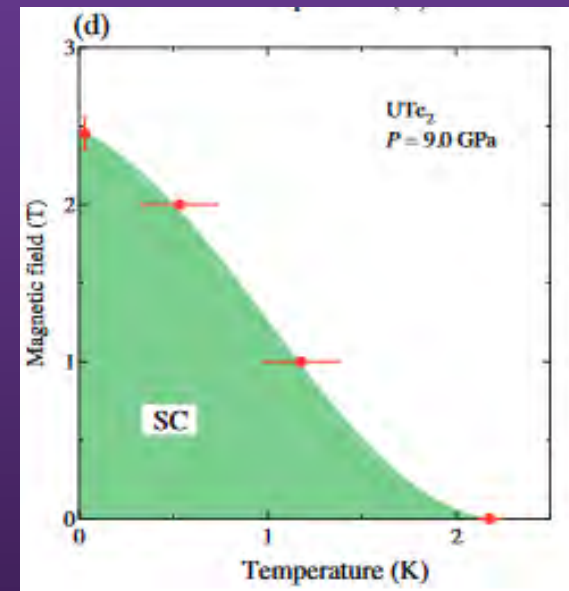
Superconducting critical temperature increases

Magnetic order above critical pressure ~ 1.5 GPa

SOMEWHAT HIGHER PRESSURES

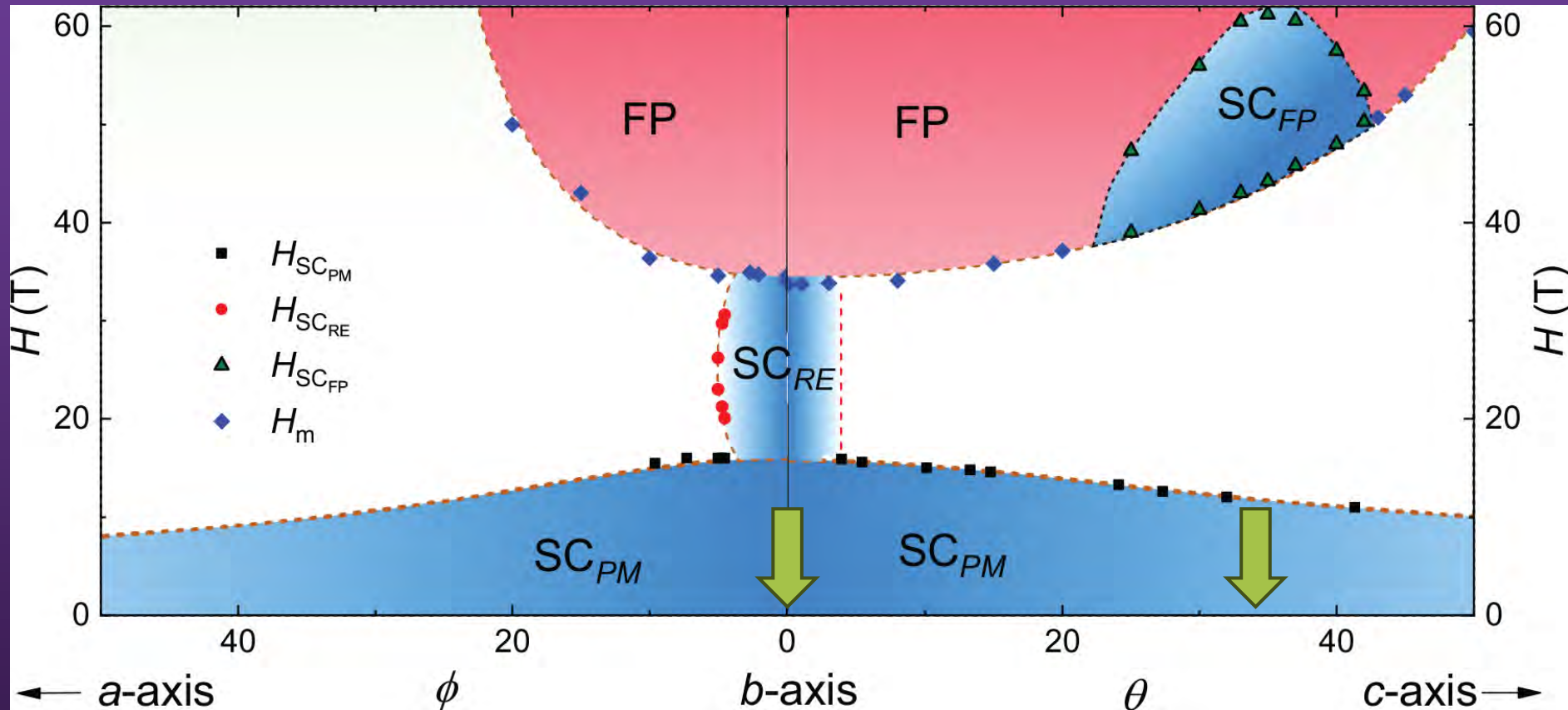


Honda JPSJ 92, 044702 (2023)



Superconductivity but smaller H_{c2}
“conventional”

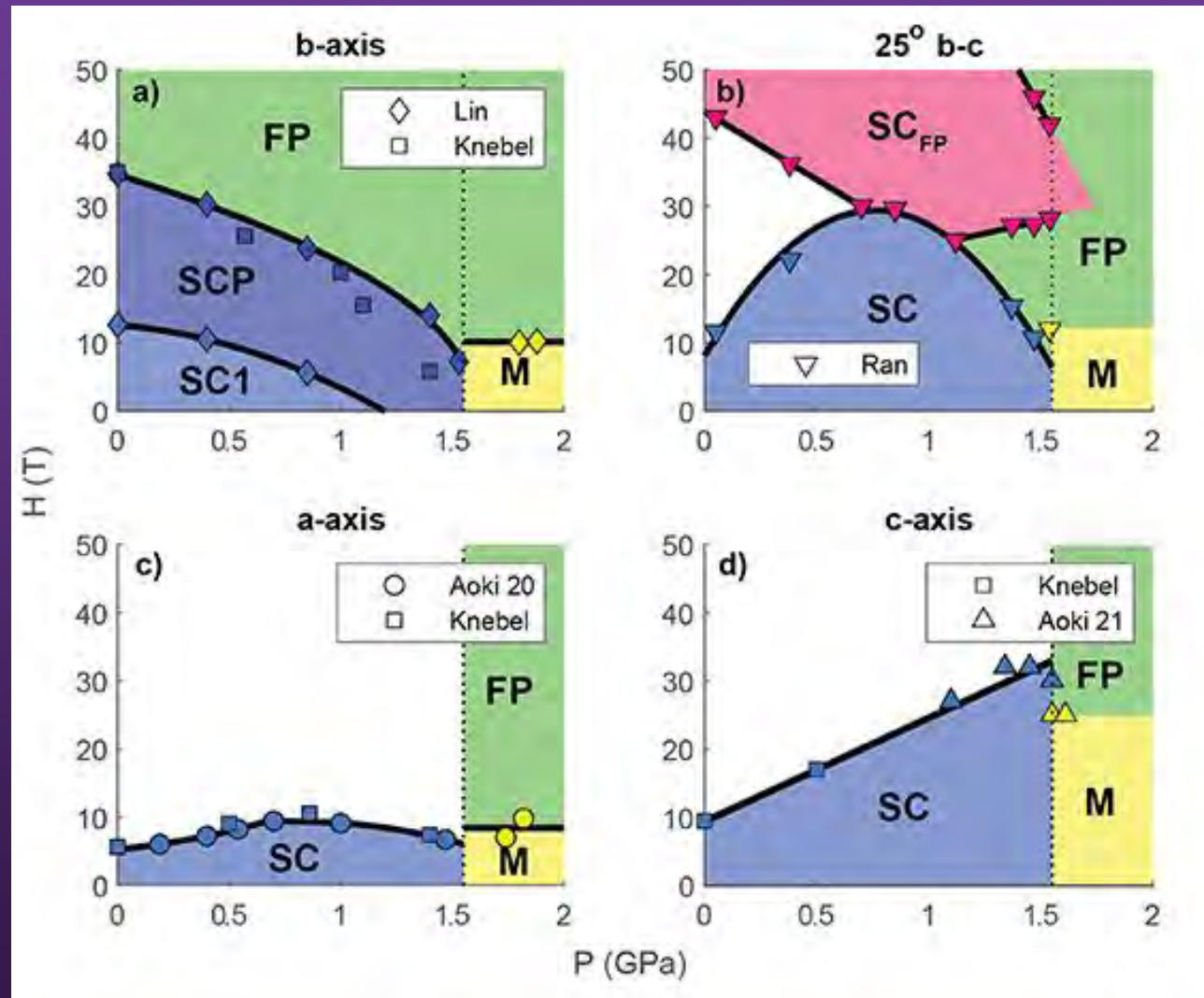
MAGNETIC FIELD ANGLE DEPENDENCE



Ran Nature Physics 15, 1250 (2019)

PRESSURE PHASE DIAGRAM, FIELD ORIENTATIONS

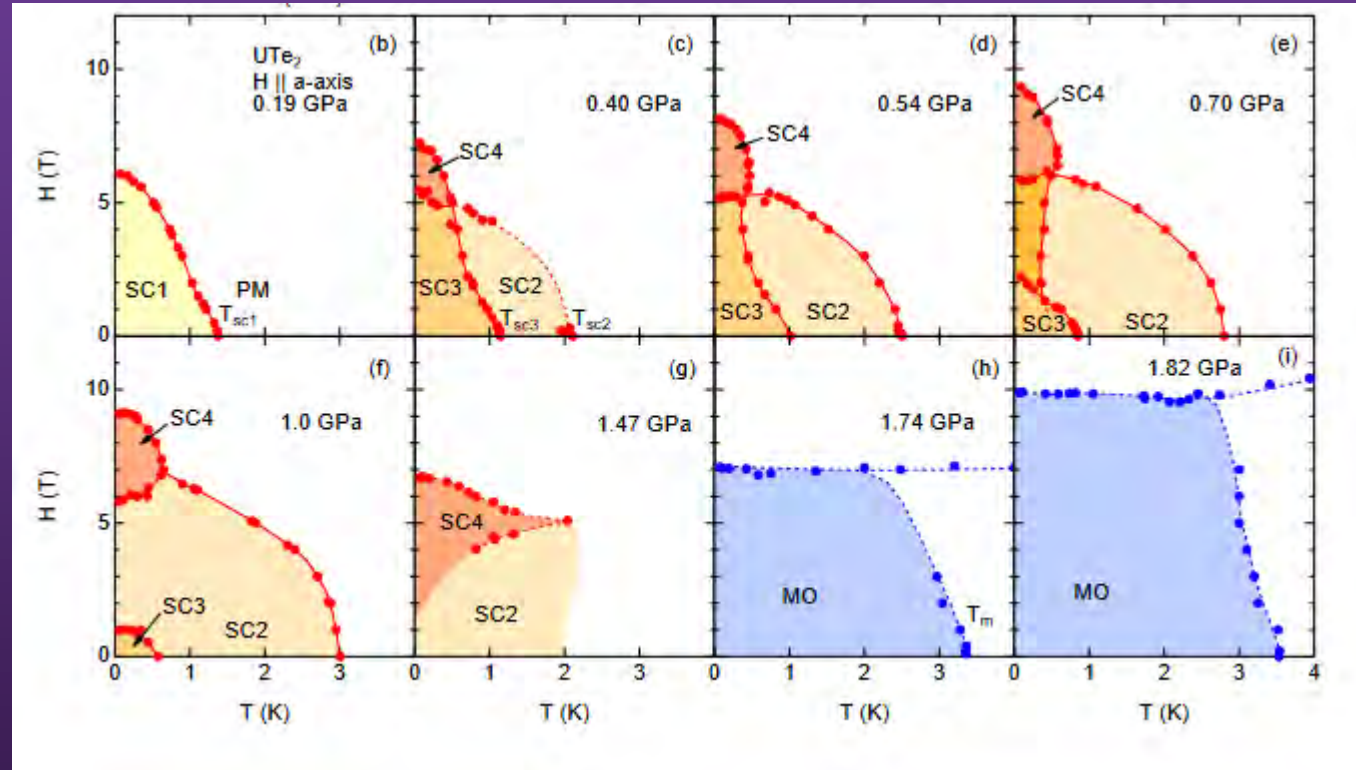
High field & Lazarus



Lower field

PRESSURE + MAGNETIC FIELD | | A

Multiple low-field superconducting phases

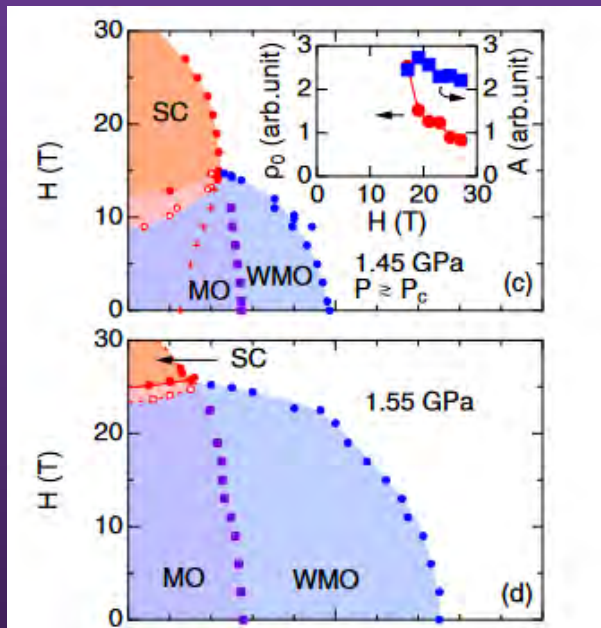


Aoki, JPSJ 89, 053705 (2020)

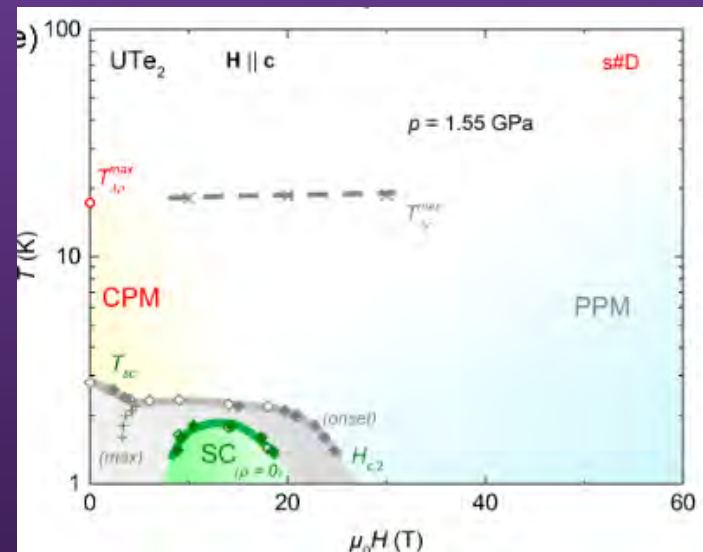
PRESSURE + MAGNETIC FIELD | | C

Reentrant superconductivity near critical pressure

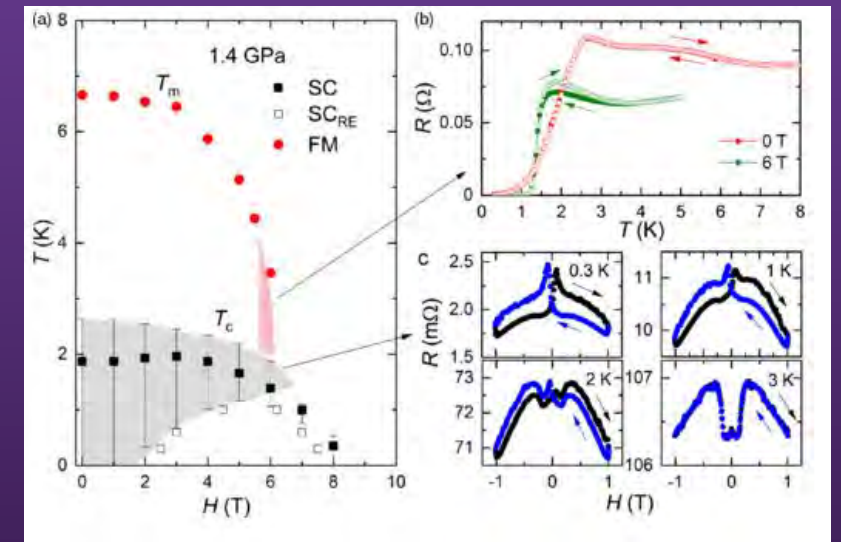
Also found at other field angles



Aoki et al, JPSJ 90, 074705 (2021)



Valiska et al, PRB 104, 214507 (2021)



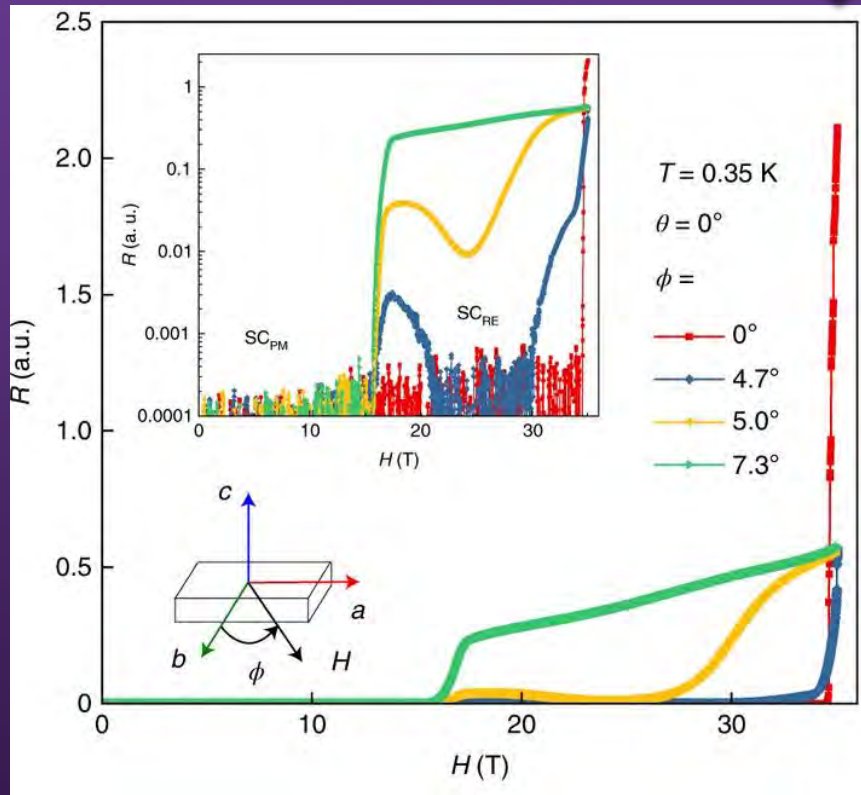
Ran et al, PRB 101, 140503(R) (2020)

FIELD POLARIZED (B-AXIS) UNDER PRESSURE

Metamagnetic transition suppressed

Ambient pressure

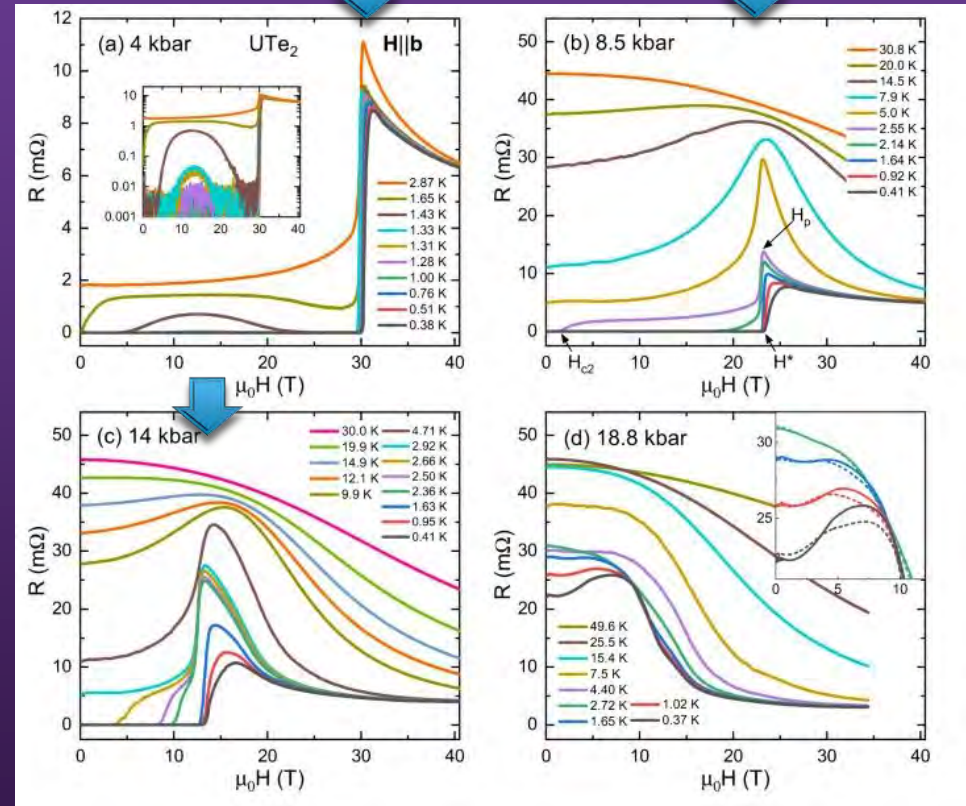
35T



Ran Nature Physics 15, 1250 (2019)

30T

24T

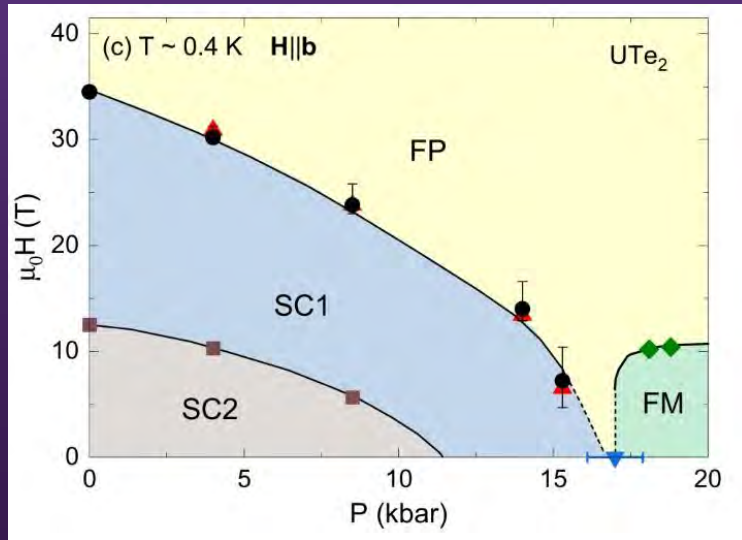
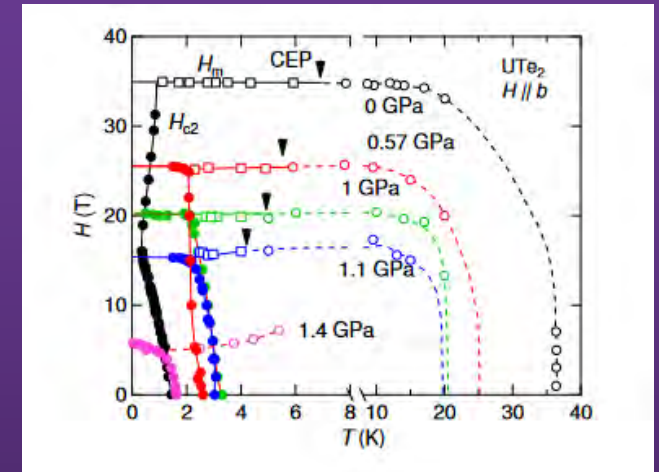
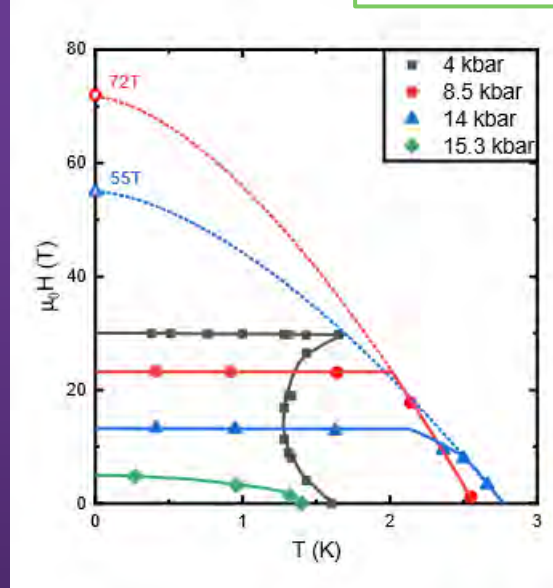
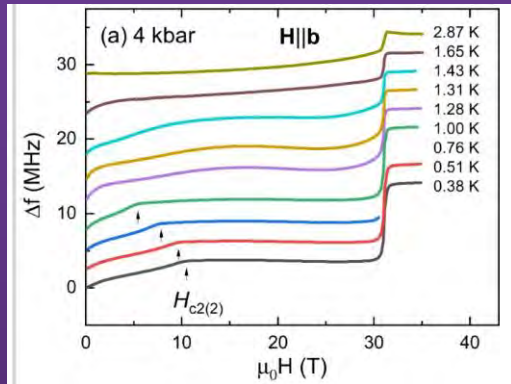


WC Lin et al, npjQM 5, 68 (2020)

MAGNETIC CONFINEMENT OF SC PHASE

Very large extrapolated $H_{c2}(0)$

Second SC seen in TDO measurements



WC Lin et al, npjQM 5, 68 (2020)

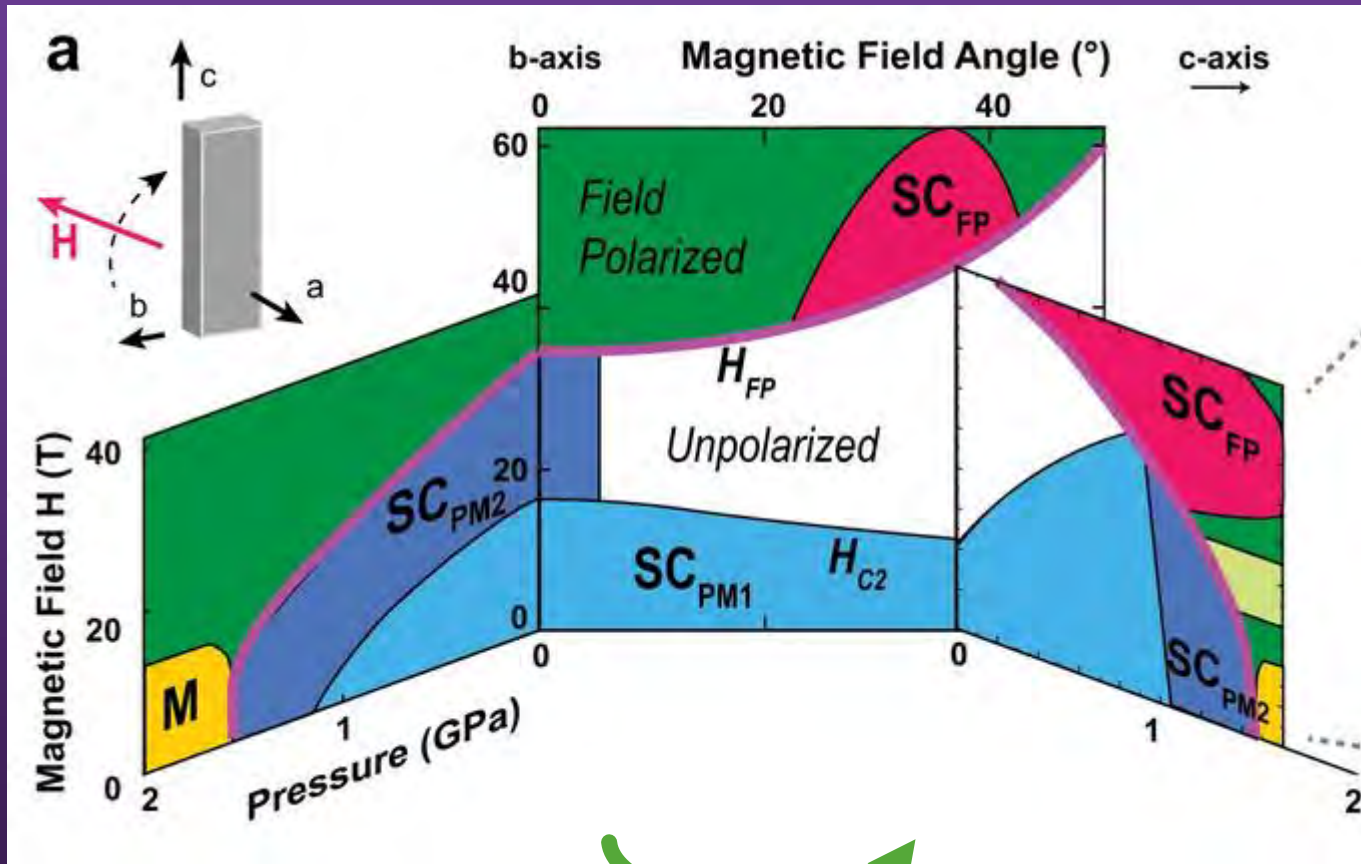
WC Lin et al, npjQM 5, 68 (2020)

Knebel et al, JSPJ 89, 053707 (2020)

PRESSURE DECREASES FP FIELD
 FP ALWAYS UPPER BOUND FOR SC
 MAGNETISM WHEN FP \rightarrow 0 FIELD

PRESSURE + MAGNETIC FIELD, LAZARUS PHASE

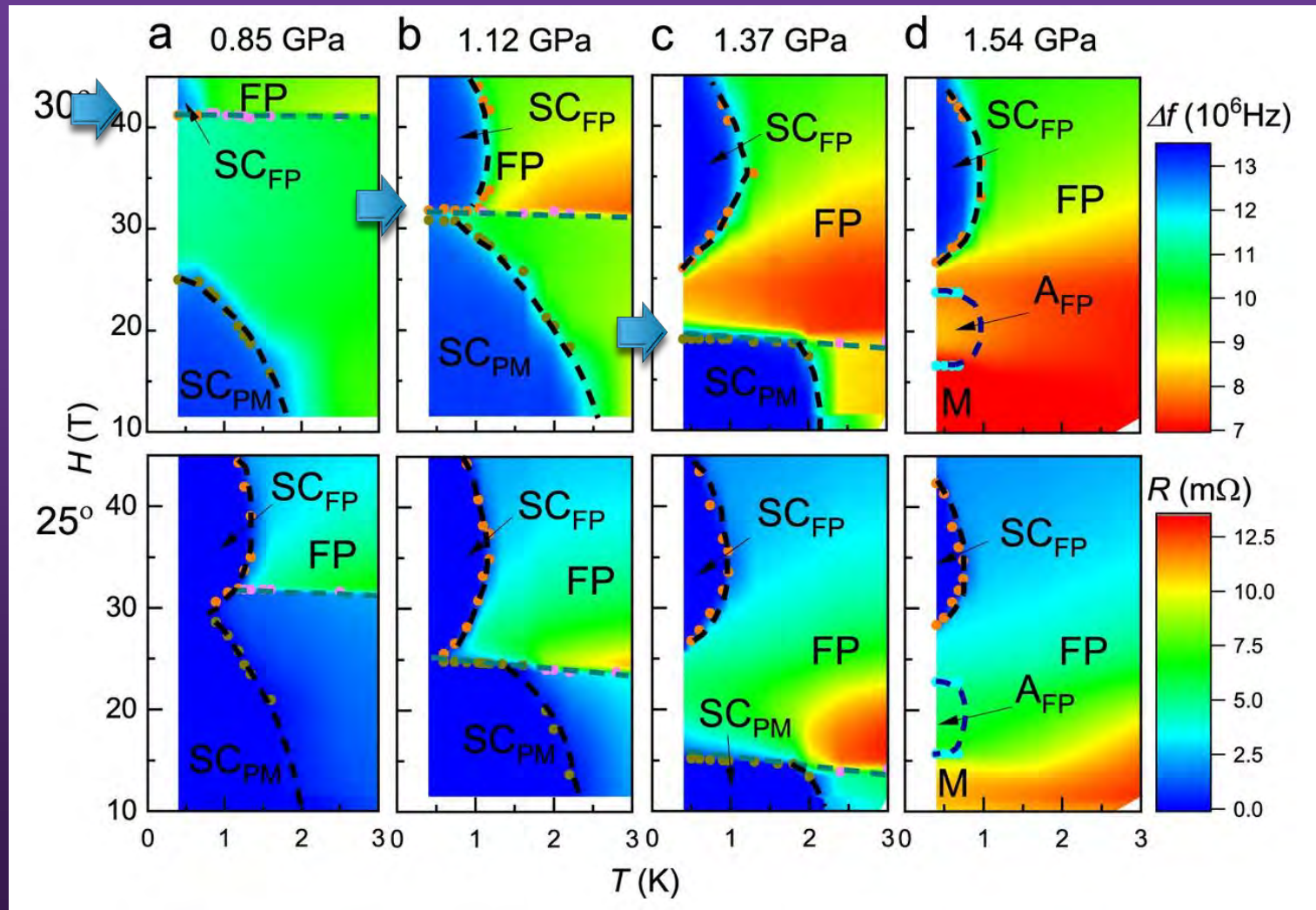
S Ran et al, npj Quantum Mater 6, 75 (2021)



Measure in 45 T DC hybrid magnet
Just before COVID pandemic shutdown

Rotate field from b towards c

SUPPRESSION OF FP, BUT ENHANCEMENT OF SC!



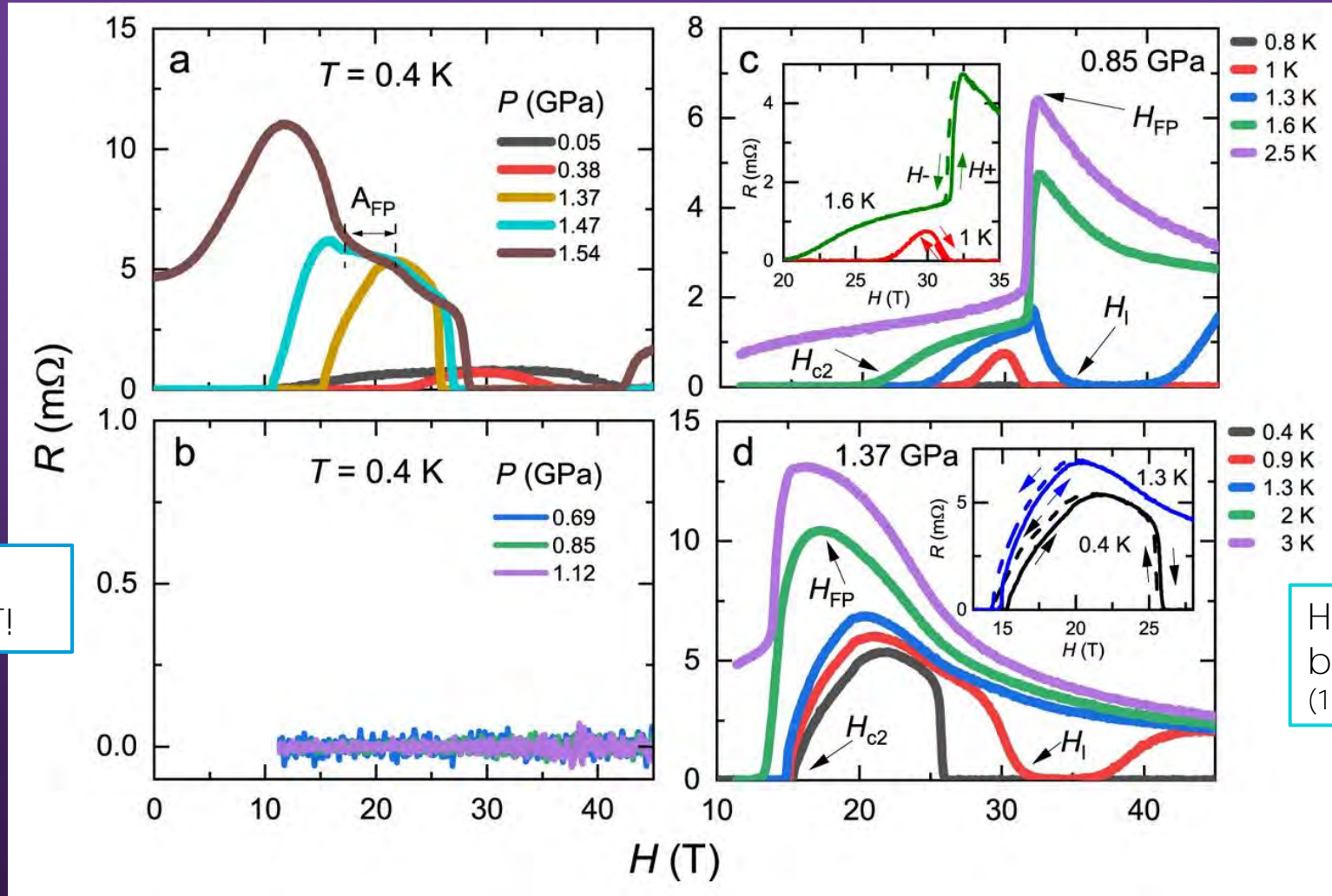
TDO

Resistance

- As pressure increases
1. FP field decreases
 2. SC_{FP} dome exposed
 3. SC_{PM} grows
 4. FP cuts off both SC_{FP} and SC_{PM}
 5. New anomalies A_{FP} at high pressure

S Ran et al, npj Quantum Mater 6, 75 (2021)

HIGHLIGHTS

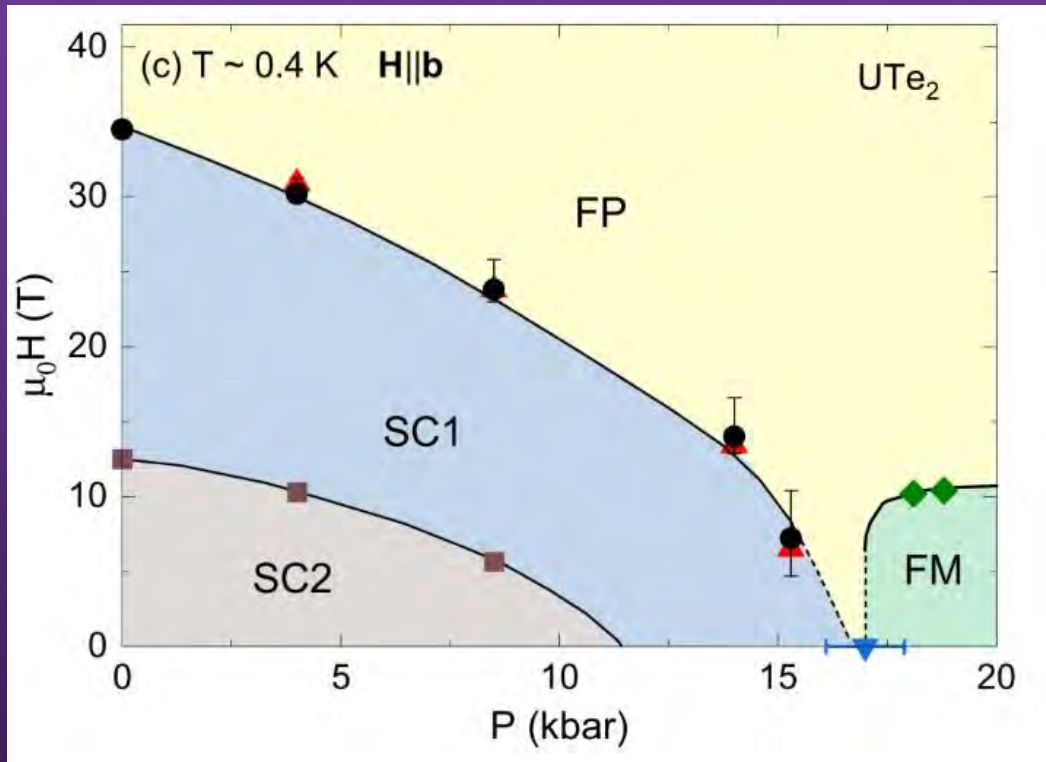


At intermediate pressures, $R=0$ to 45 T!

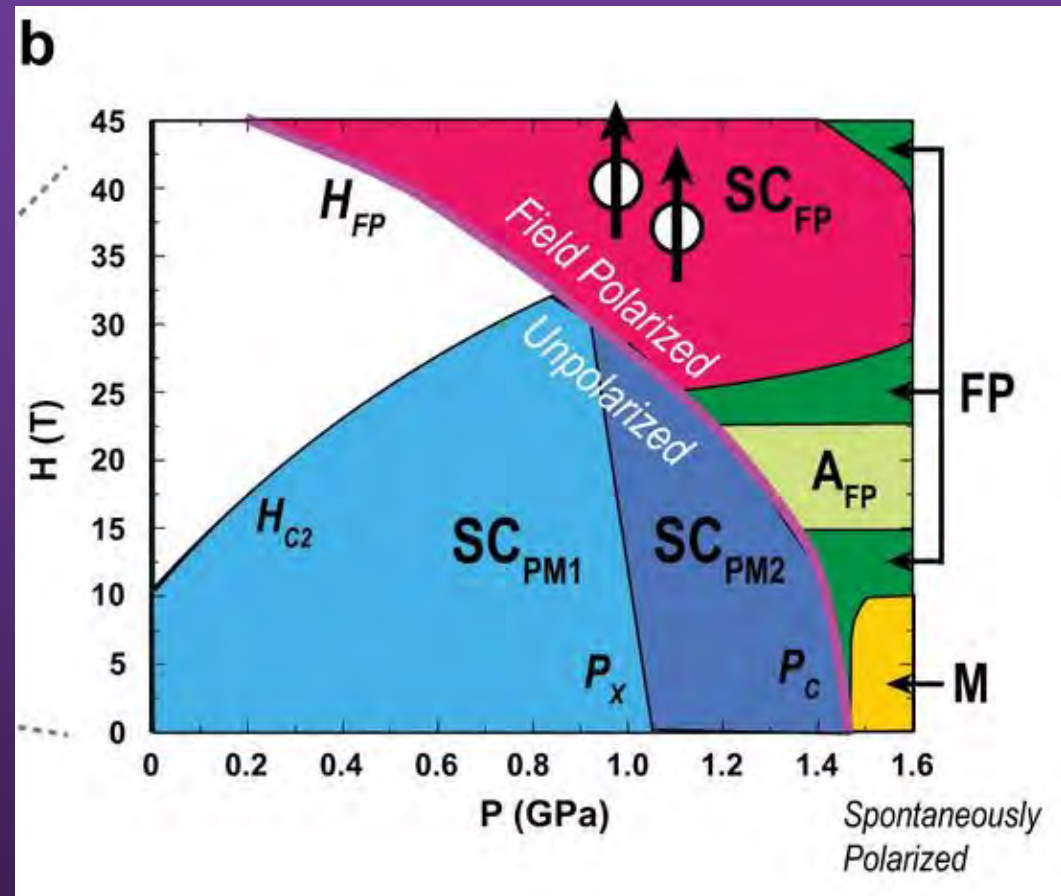
Hysteresis at boundaries (1st order transitions)

S Ran et al, npj Quantum Mater 6, 75 (2021)

THE FP TRANSITION GENERALLY LIMITS SC



WC Lin et al, npjQM 5, 68 (2020)

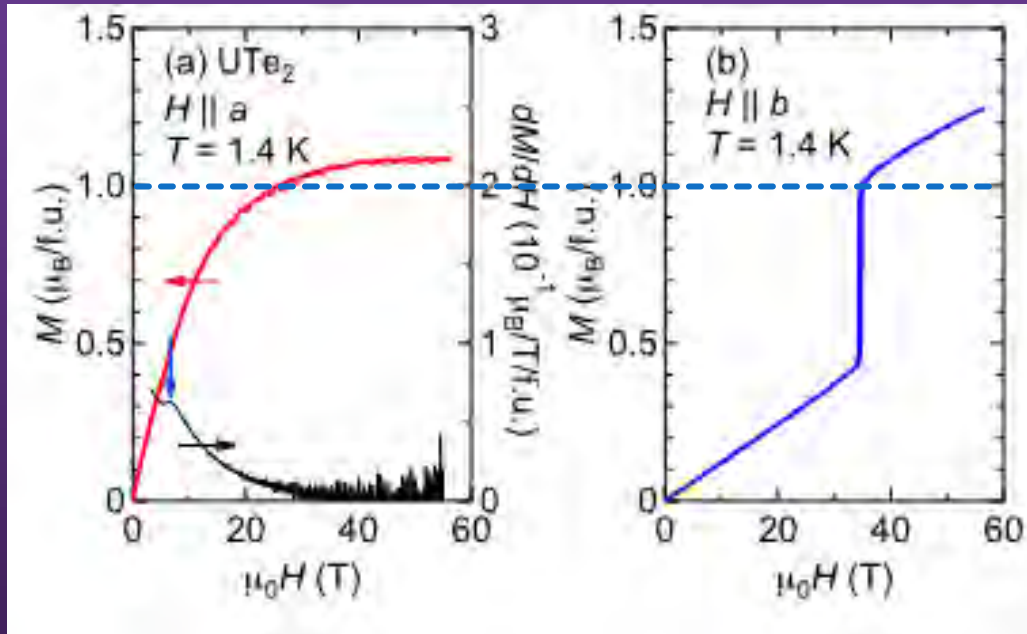


S Ran et al, npjQM 6, 75 (2021)

1. FP transition always cuts off SC, whether it is enhanced or suppressed by pressure
2. FP transition also cuts off high-field SC, although it survives beyond the critical pressure

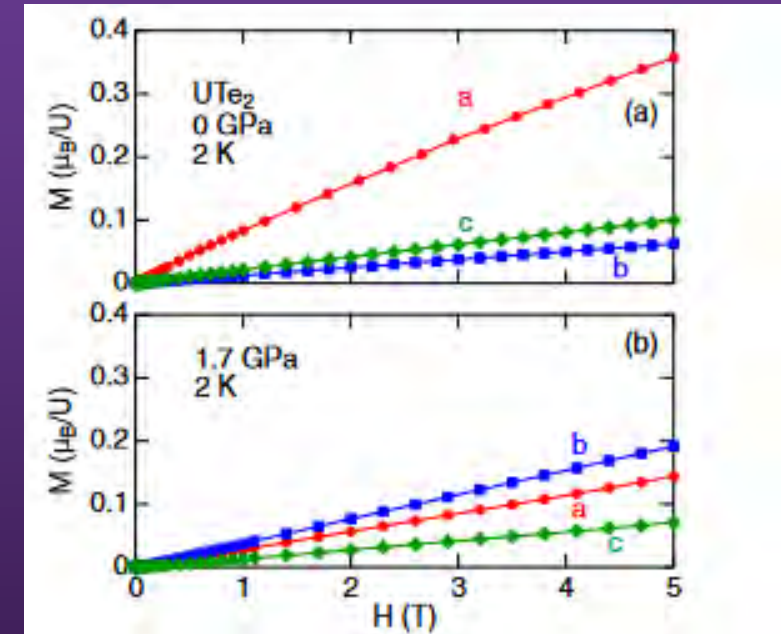
BOTH FIELD AND PRESSURE: B-AXIS BECOMES EASY AXIS

At high fields

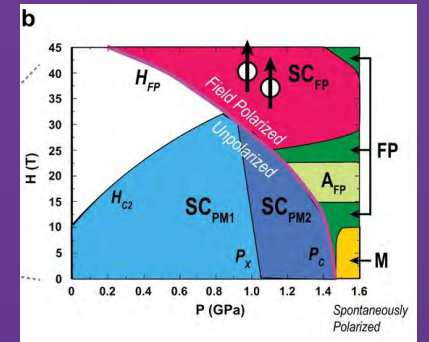


Miyake et al, JPSJ 88, 063706 (2019)

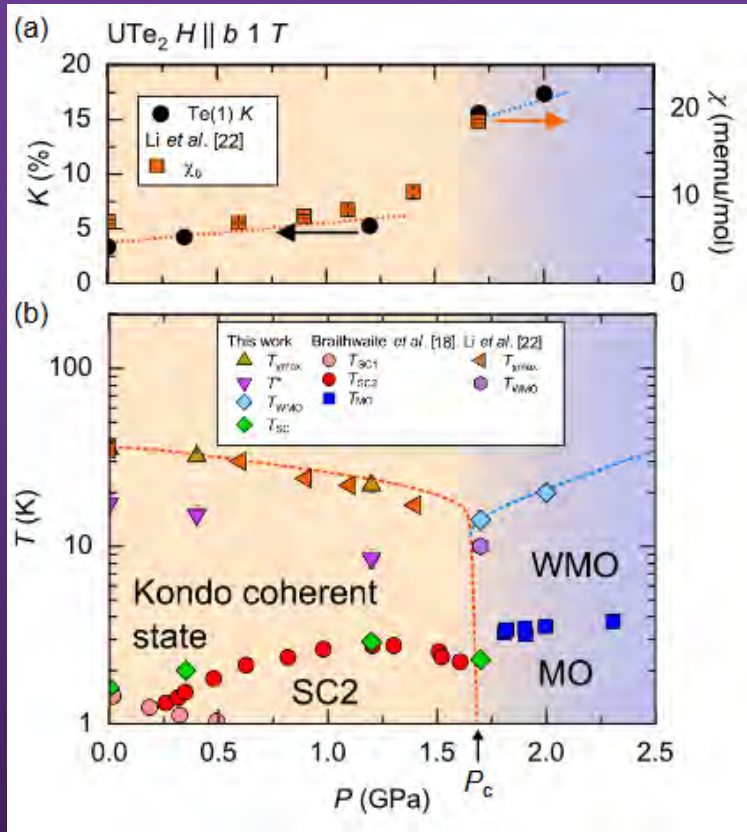
Under pressure



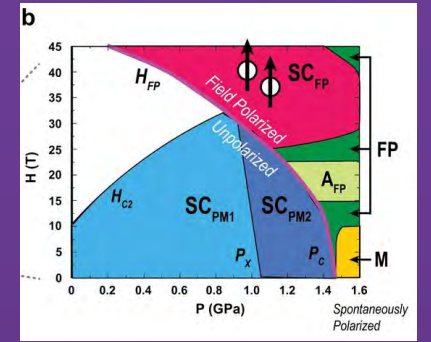
Li et al, JPSJ 90, 073703 (2021)



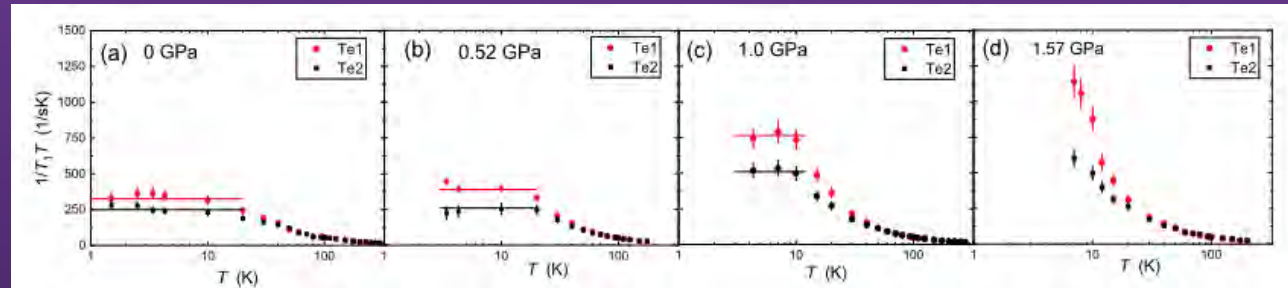
NMR UNDER PRESSURE



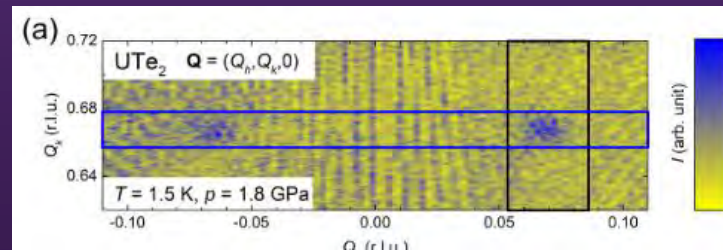
Kinjo PRB 105, L140502 (2022)



Ground state changes from Fermi liquid (heavy fermion) to magnetic order above the critical pressure



Ambika PRB 105, L220403 (2022)



Knafo arXiv:2311.05455

Neutron under pressure – Incommensurate AFM magnetic order

CONCLUSIONS

- SPIN-TRIPLET, NODAL “LOW FIELD” SUPERCONDUCTIVITY
 - IDENTIFICATION OF ORDER PARAMETER(S) CONTINUES
 - TIME REVERSAL SYMMETRY AND SURFACE STATES?
- UNPRECEDENTED HIGH FIELD SUPERCONDUCTIVITY
 - COMPLICATED EVOLUTION WITH PRESSURE
 - METAMAGNETIC FIELD AND CRITICAL PRESSURE CONNECTED
 - MULTIPLE SUPERCONDUCTING PHASES

THANK YOU FOR YOUR ATTENTION ...