

Unconventional Superconductivity in UTe₂ in Extreme Conditions

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Collaborators



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Institut Néel:

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ESRF, Grenoble

G. Gabarino, J.P. Sanchez, F. Wilhelm

ISIS, UK

P. Manuel

Topical review:

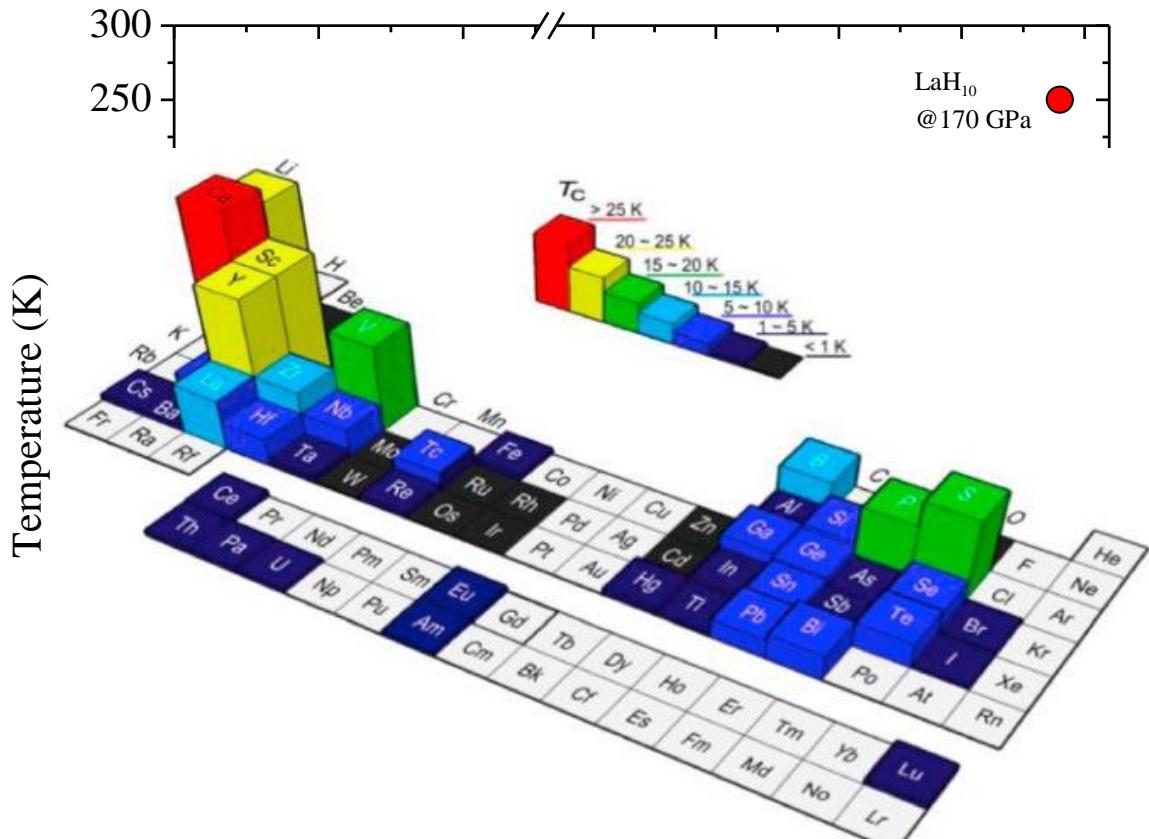
D. Aoki, J.-P. Brison, J. Flouquet, K. Ishida, G. Knebel, Y. Tokunaga, and Y. Yanase, *J. Phys.: Condens. Matter* **34** (2022) 243002

Progress report on high fields
S. K. Lewin, C.E. Frank, S. Ran, J. Paglione, N. Butch
Rep. Prog. Phys. **86** (2023) 114501

Thanks to : Y. Tokunaga, K. Ishida, J. Ishizuka, Y. Yanase, H. Harima

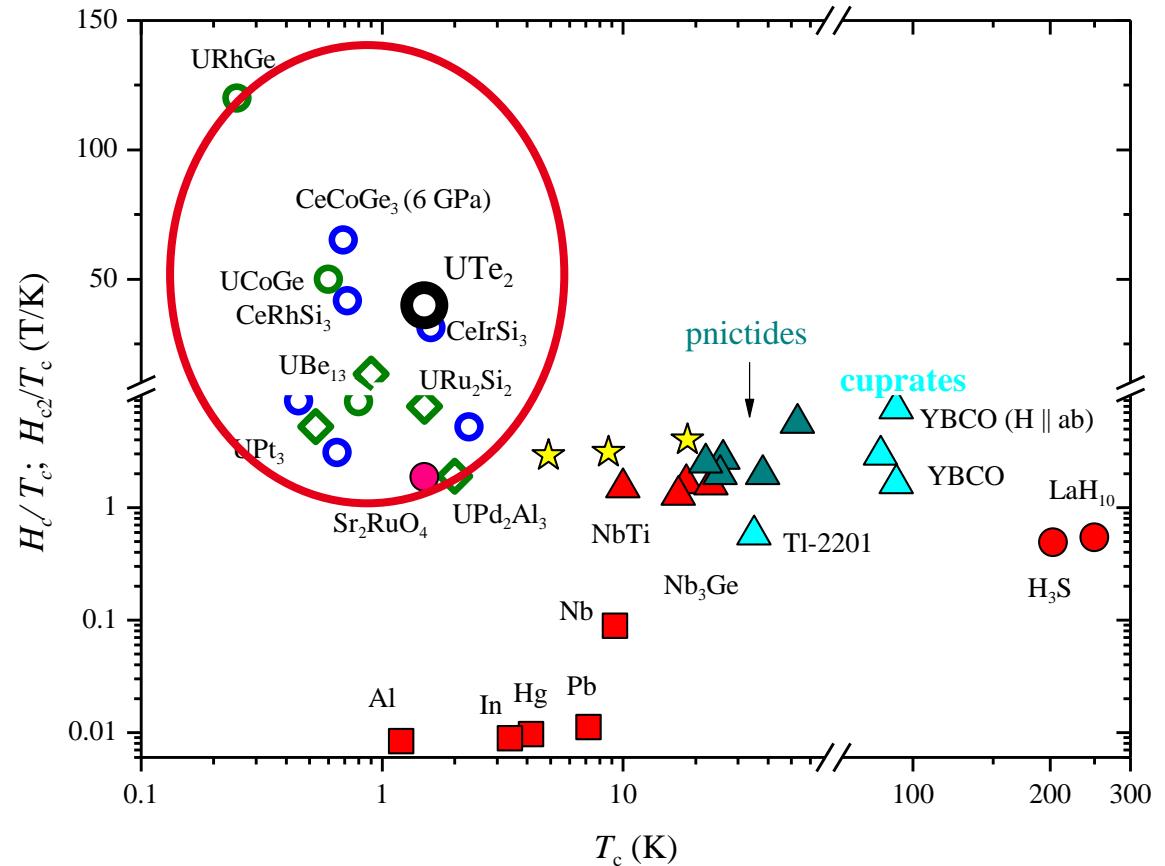


Superconductors



from K. Shimizu, Physica C, 552, 30 (2018)

magnetic field effect :



heavy fermion superconductors:

Cooper pairs formed by electrons with high effective masses \Rightarrow high critical fields



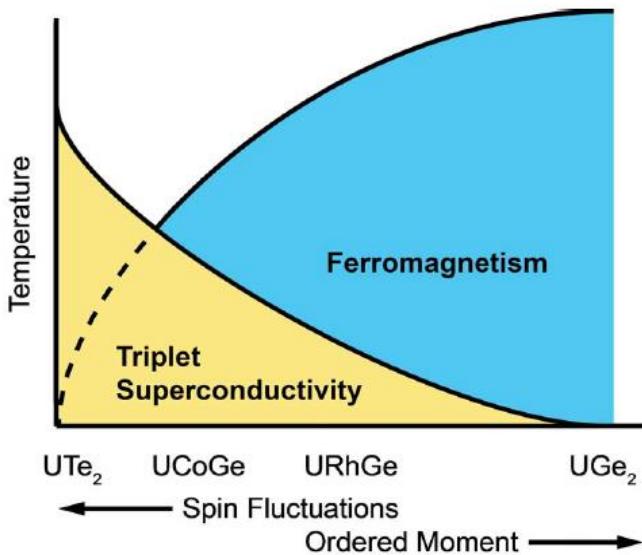
New heavy fermion superconductor: UTe₂

Science 365, 684–687 (2019) 16 August 2019

SUPERCONDUCTIVITY

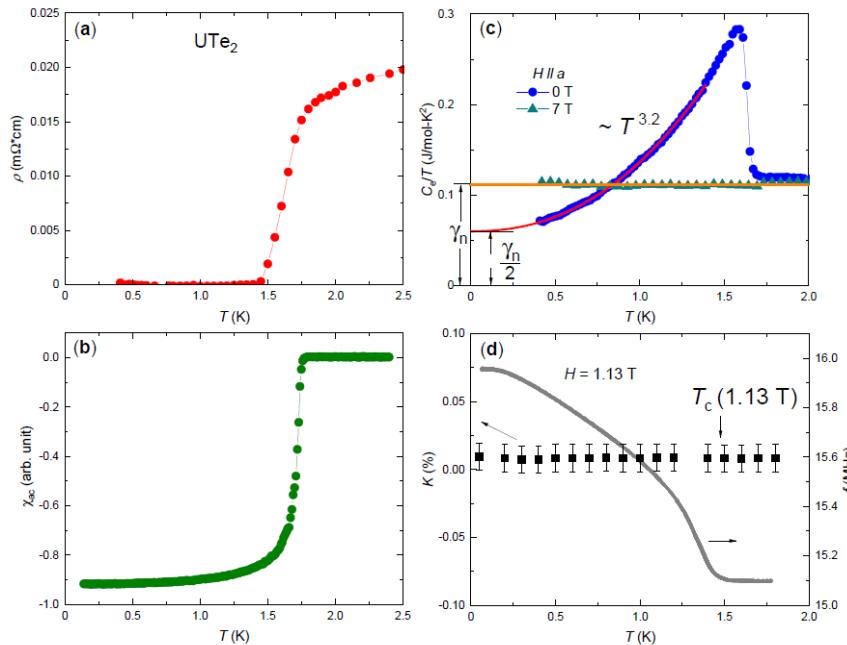
Nearly ferromagnetic spin-triplet superconductivity

Sheng Ran^{1,2*}, Chris Eckberg², Qing-Ping Ding³, Yuji Furukawa³, Tristin Metz², Shanta R. Saha^{1,2}, I-Lin Liu^{1,2,4}, Mark Zic², Hyunsoo Kim², Johnpierre Paglione^{1,2}, Nicholas P. Butch^{1,2*}

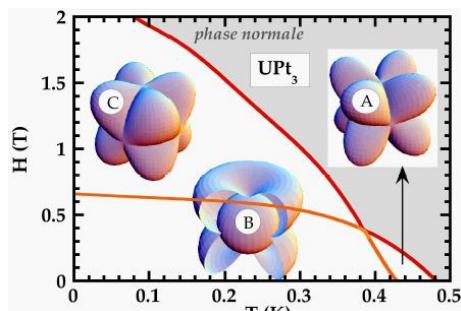


Spin – triplet superconductivity

- Cooper pair with spin $S = 1$, (p -wave)
- spin degree of freedom
⇒ multiple superconducting phases
- chiral p -wave states, topological superconductivity

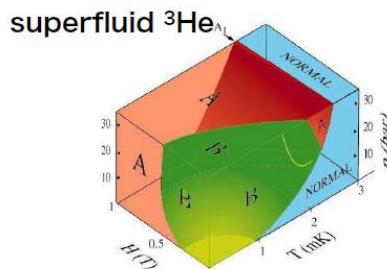


Multiple superconducting phases



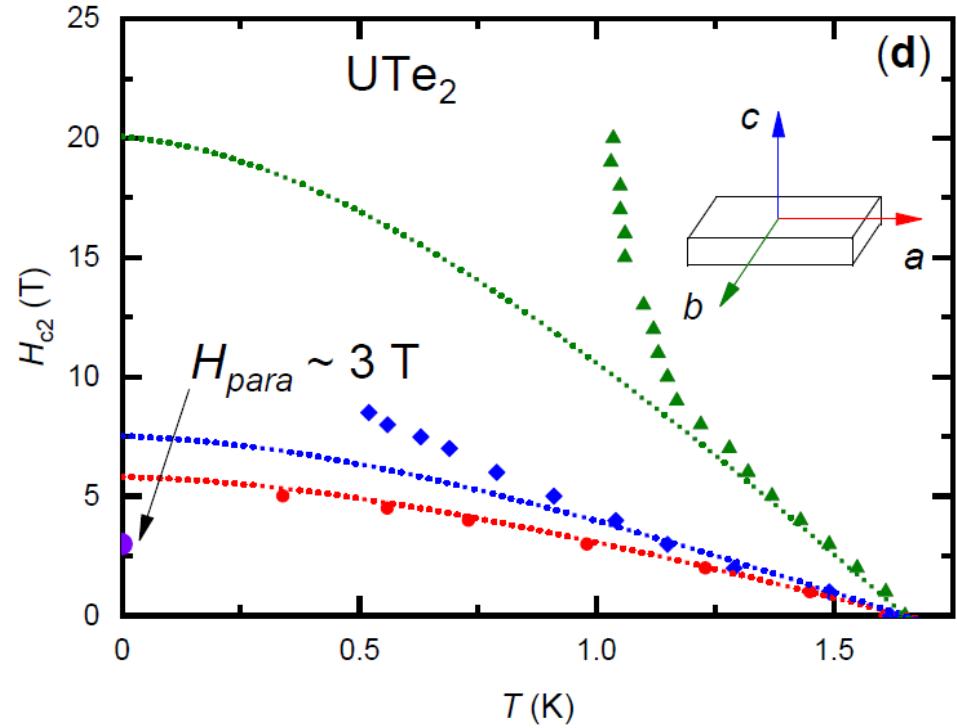
$$\begin{aligned} T_{sc} &= 1.6 \text{ K} \\ \gamma &= 117 \text{ mJ/molK}^2 \\ \gamma_0 &\approx 50 - 60 \text{ mJ/molK}^2 \\ C/T &\propto T^{3.2} \end{aligned}$$

- no change in Knight shift
- point nodes in the gap ?





Extremely high superconducting upper critical field



Ran et al. Science 365, 684 (2019)

Pauli – limit : polarization = condensation energy

$$H_P = \sqrt{2}\Delta/g\mu_B$$
$$H_{c2}^P = 1.86 T_c = 3 \text{ T}$$

orbital – limit : Lorentz force on Cooper pairs

$$H_{\text{orb}}(T) = \Phi_0/2\pi\xi^2(T)$$

$$H'_{c2} = (dH_{c2}/dT)_{T=T_c} \approx T_c/v_F^2$$

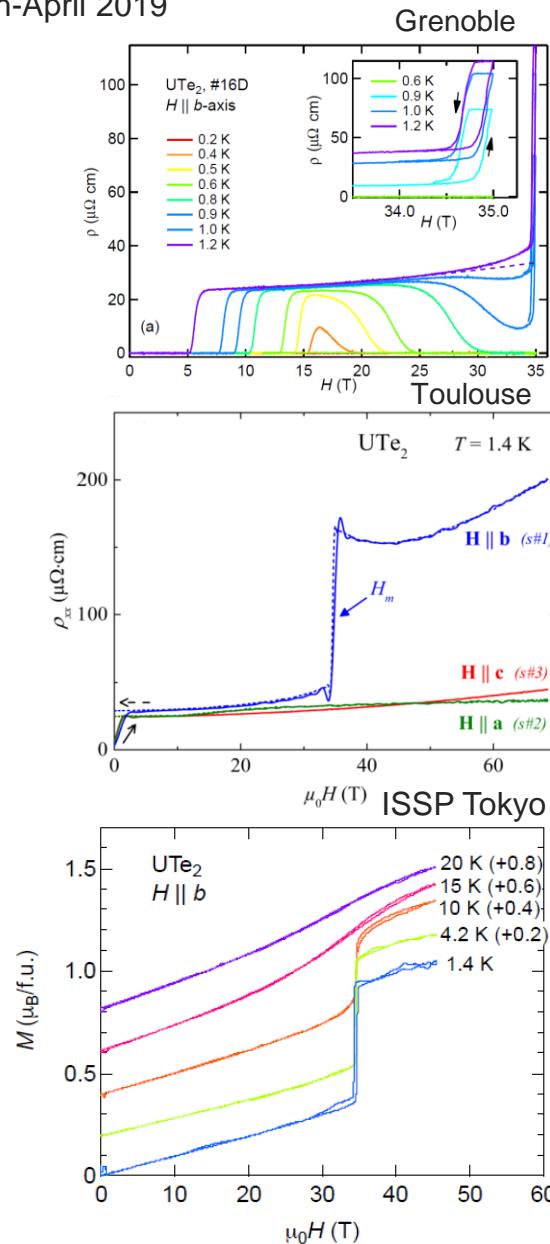
$$H_{c2}^{orb} = -0.7 \frac{dH_{c2}}{dT}|_{T_c} T_c$$
$$\approx 20 \text{ T} \quad (H \parallel b)$$

- ⇒ very large upper critical field
- ⇒ spin triplet superconductivity
- ⇒ mechanism enhancing superconductivity under field

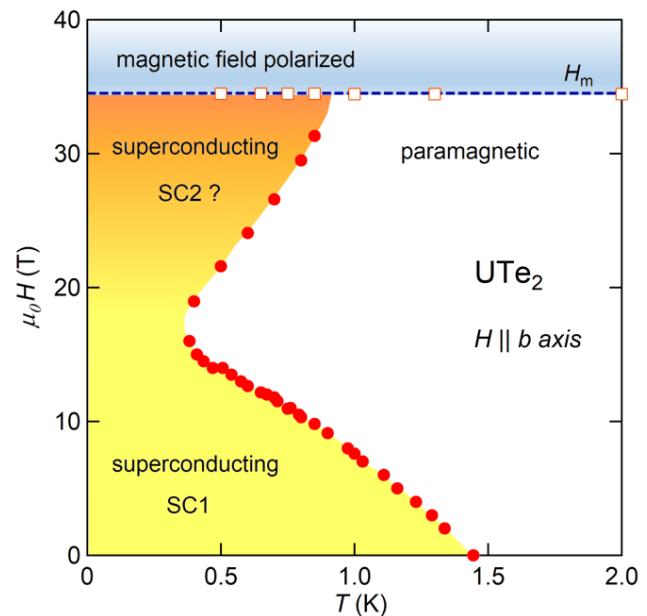


Phase diagram $H \parallel b$ axis at zero pressure

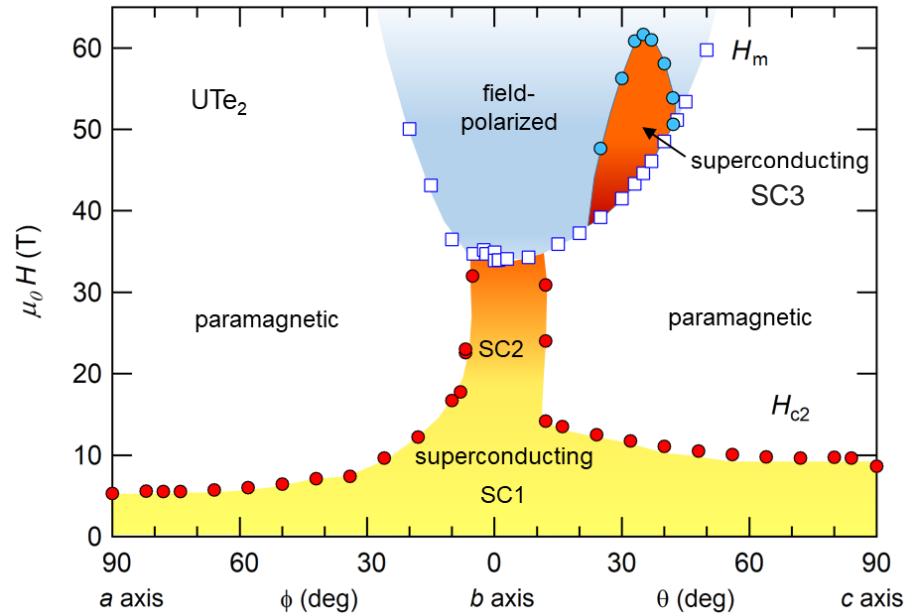
March-April 2019



upper critical field $H \parallel b$ axis



angular dependence of upper critical field



Ran et al. Nat. Phys. 2019

Phase diagram only determined by transport, no thermodynamic measurement !

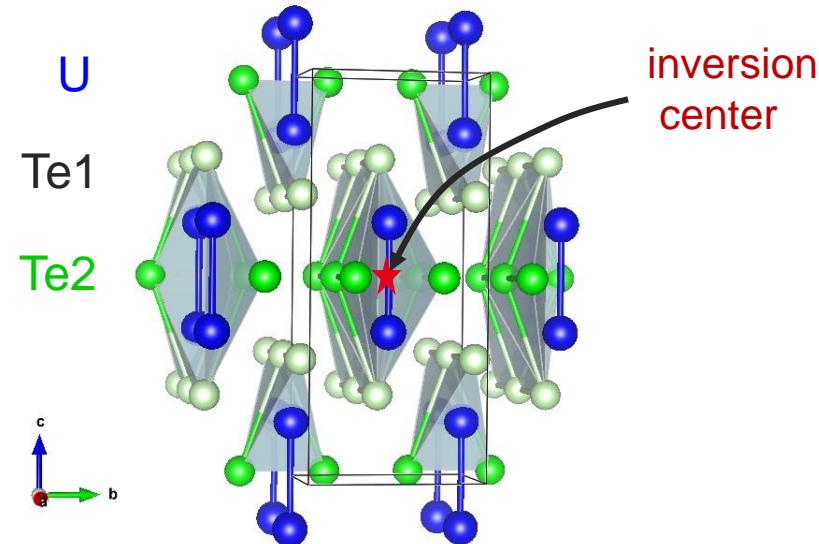
Knebel et al. JPSJ 2019, W. Knafo et al. JPSJ **88**, 063705 (2019), A. Miyake et al. JPSJ **88**, 063705 (2019)
Ran et al. Nat. Phys. 2019, Knafo et al. Commun. Phys. 2021, Helm et al. Nature Communications | (2024)15, 37



UTe₂ crystal structure: local inversion symmetry broken

body centered orthorhombic
space group Immm (#71, D_{2h}^{25})

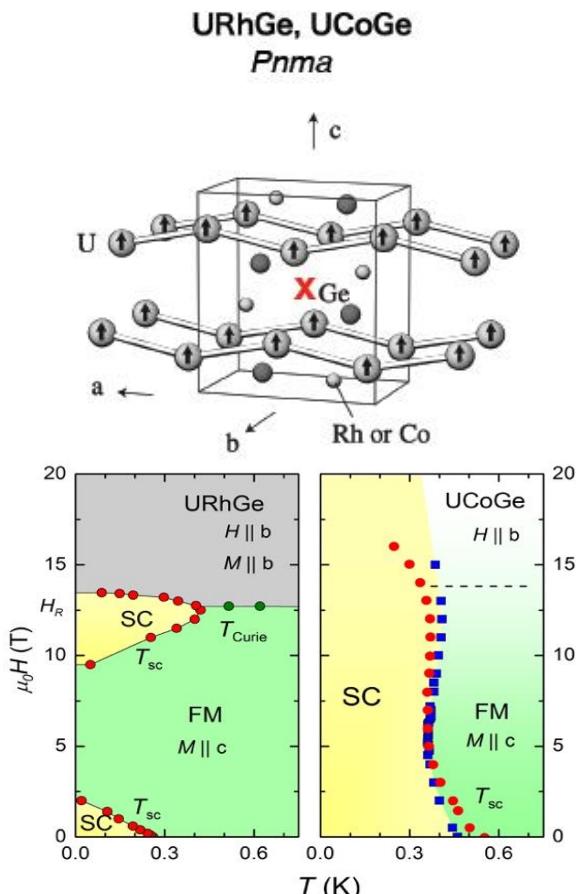
- two leg ladder structure of U atoms
- Te2 chains along the *b* axis
- local inversion symmetry broken



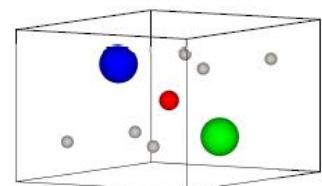
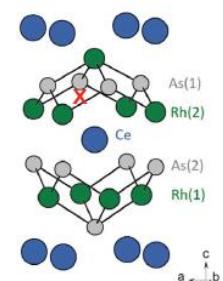
$a=4.161 \text{ \AA}$, $b=6.122 \text{ \AA}$, $c = 13.955 \text{ \AA}$
 $d_{\text{U-U}} = 3.78 \text{ \AA}$ on ladder rung > Hill – limit
(U localized, no superconductivity)

P.W. Anderson PRB 32, 499 (1985)

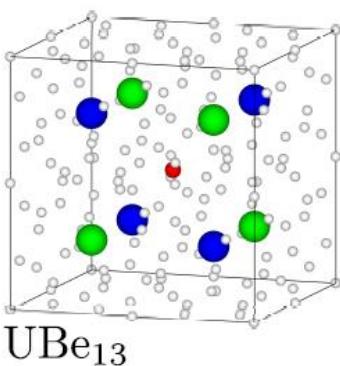
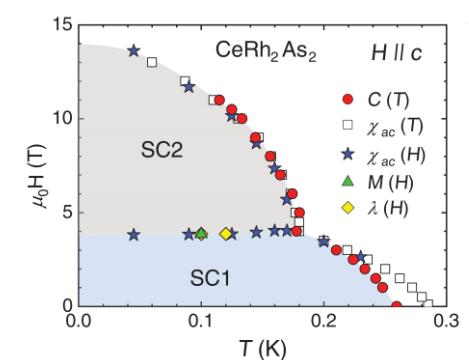
The energy-gap symmetry is severely constrained by the physical assumption that the attractive interaction involves *f*-electron pairing in the *f*-shell heavy-electron superconductors. This suggests that odd-parity (“triplet”) superconductors must have at least two *f*-shell atoms per unit cell.



CeRh₂As₂
P4/nmm

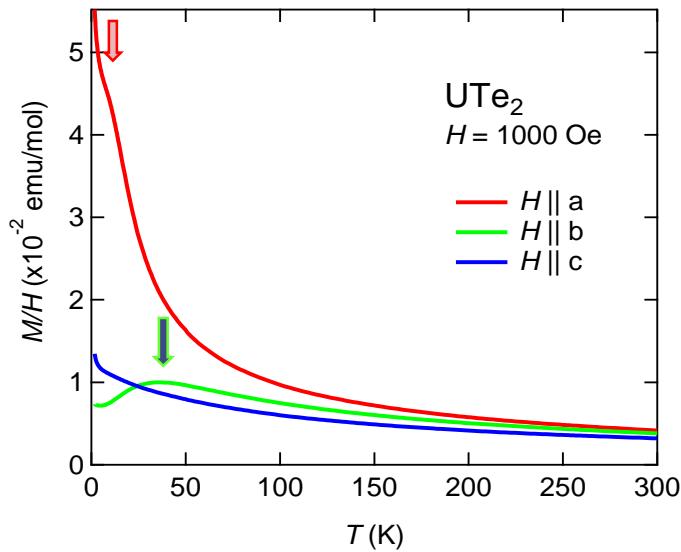


UPt₃

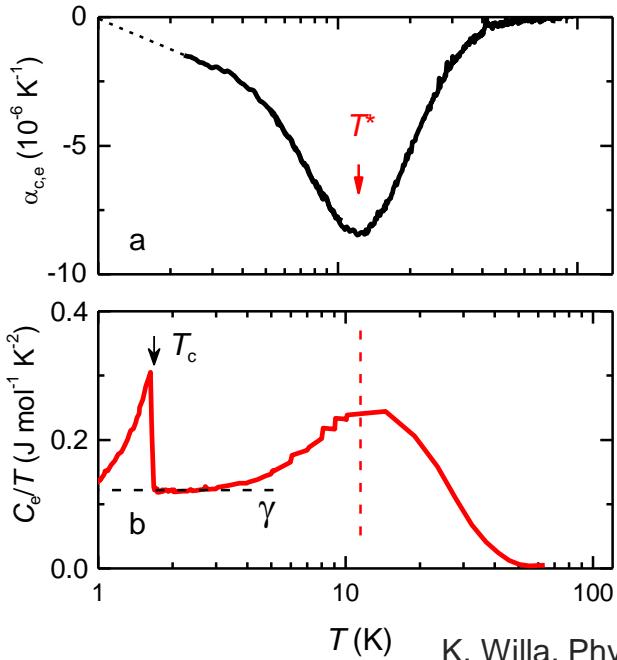


UBe₁₃

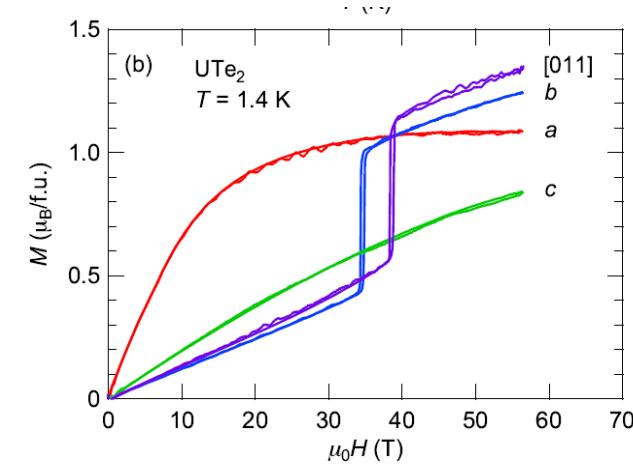
UTe₂: normal state properties



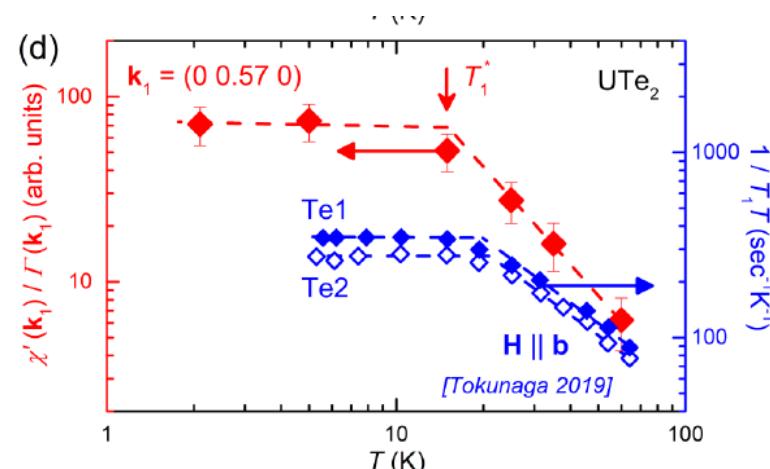
- paramagnet
- strong magnetic anisotropy, a -axis easy axis
- maximum in M/H near 35 T for $H \parallel b$
- 1st order metamagnetic transition at 35 T
- negative Curie-Weiss temperatures \Rightarrow antiferromagnetic exchange at high temperatures



- “Schottky”-like anomaly at $T^* \sim 12$ K in thermodynamics and transport properties
- $T^* \sim 12 - 15$ K characteristic energy scale in microscopic experiments.

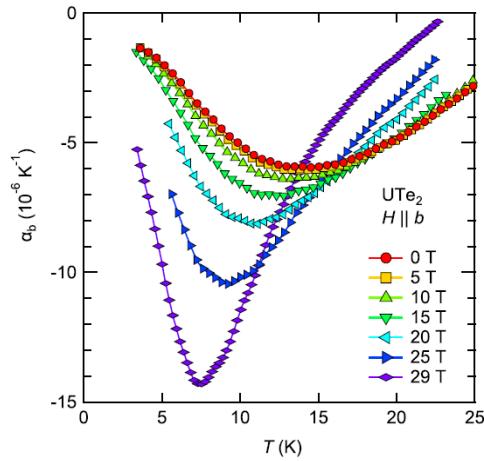
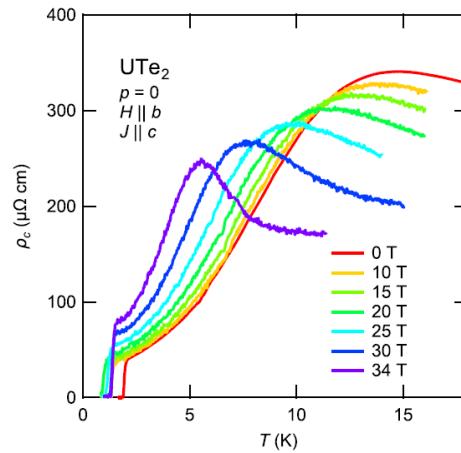


A. Miyake et al. JPSJ 2021

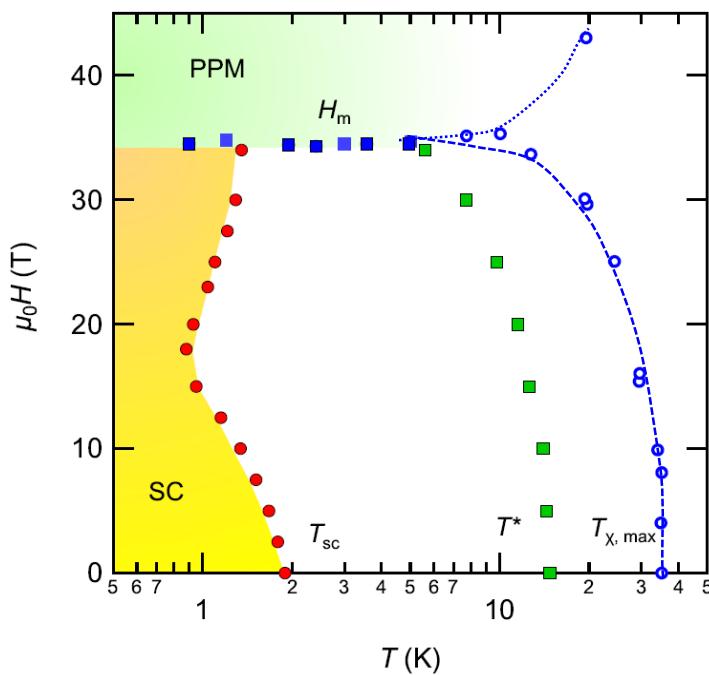




Characteristic energy scales under field



- 1st order metamagnetic transition
- critical point of the 1st order transition
- superconductivity suppressed at metamagnetic transition H_m



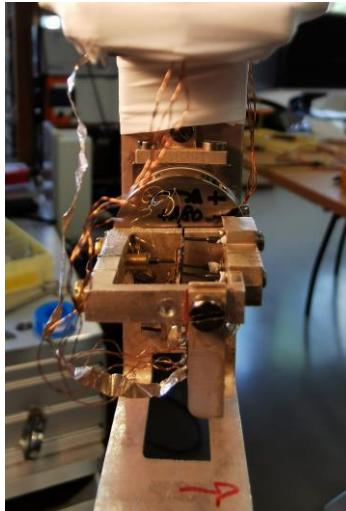
Mechanism for field enhancement of superconductivity ?

- Enhancement bulk property ?
- Requires increase of pairing strength !
 - Noted by experimentalist (Ran et al. Supp. Science 2019, **365**, 684)
 - Ignored by theoretical models !
- different superconducting phases ?
 - symmetry OP, change of OP with magnetic field ? Rotation of **d** vector?
 - Connection with (p, T) phase diagrams ?

Thermodynamic probes under (static) high fields

❑ Specific heat – semi-adiabatic (10mg UTe₂) CEA

- two axes rotation
- 0.06mK / 15T (A. Rosuel, J.-P. Brison)



❑ Specific heat – ac (10µg UTe₂)

- 0.4K / 36T (A. Rosuel, C. Marcenat, T. Klein)

❑ Thermal dilatation/magnetostriction

$$\alpha = \frac{1}{V} \frac{\partial V}{\partial T} |_{p,B} \Leftrightarrow -\frac{1}{V} \frac{\partial S}{\partial p} |_{T,B} \quad \lambda = \frac{1}{V} \frac{\partial V}{\partial B} |_{p,T} \Leftrightarrow -\frac{1}{V} \frac{\partial M}{\partial p} |_{T,B}$$

⇒ phase transition
kink in $\Delta L/L$
discontinuity in $\alpha(T)$

⇒ magnetic properties
irreversibilities
vortex pinning



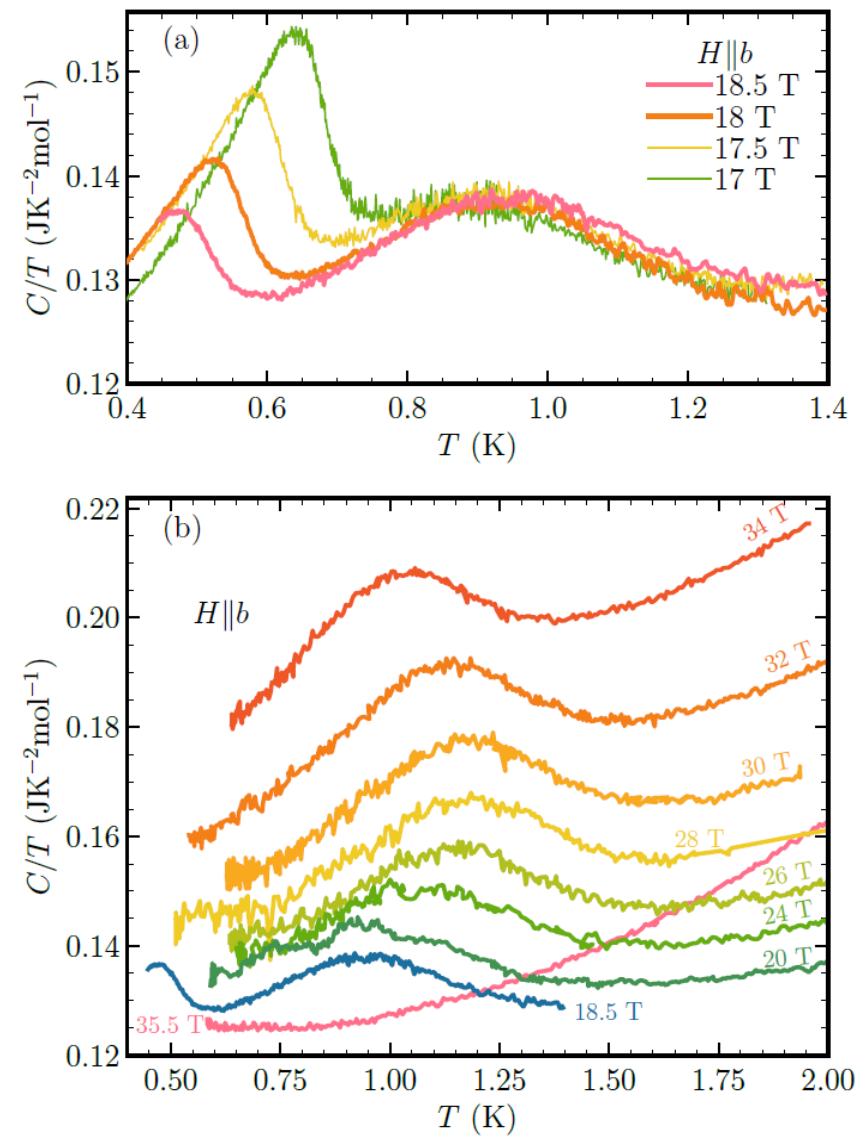
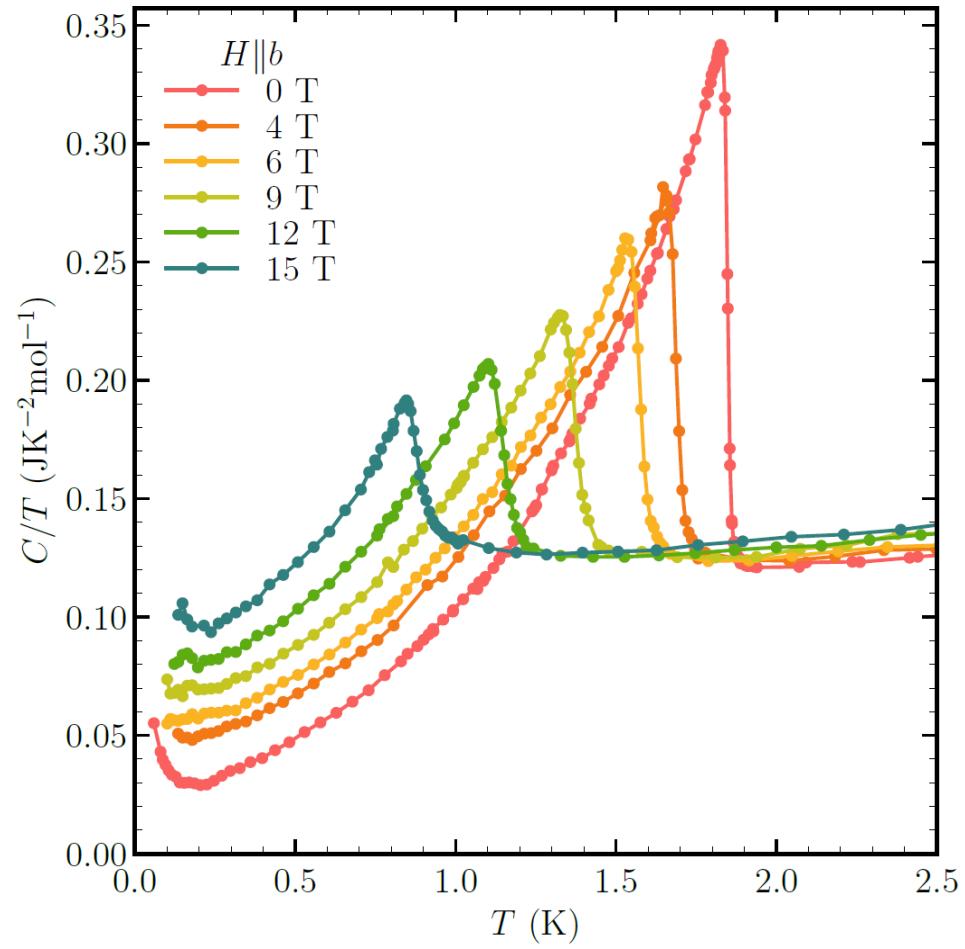
Ac calorimetry

capacitive dilatometer

$$: \frac{\Delta L_i}{L_i} = \frac{1}{L_i} \epsilon_0 \epsilon_r A \frac{c - c_i}{c \cdot c_i}$$

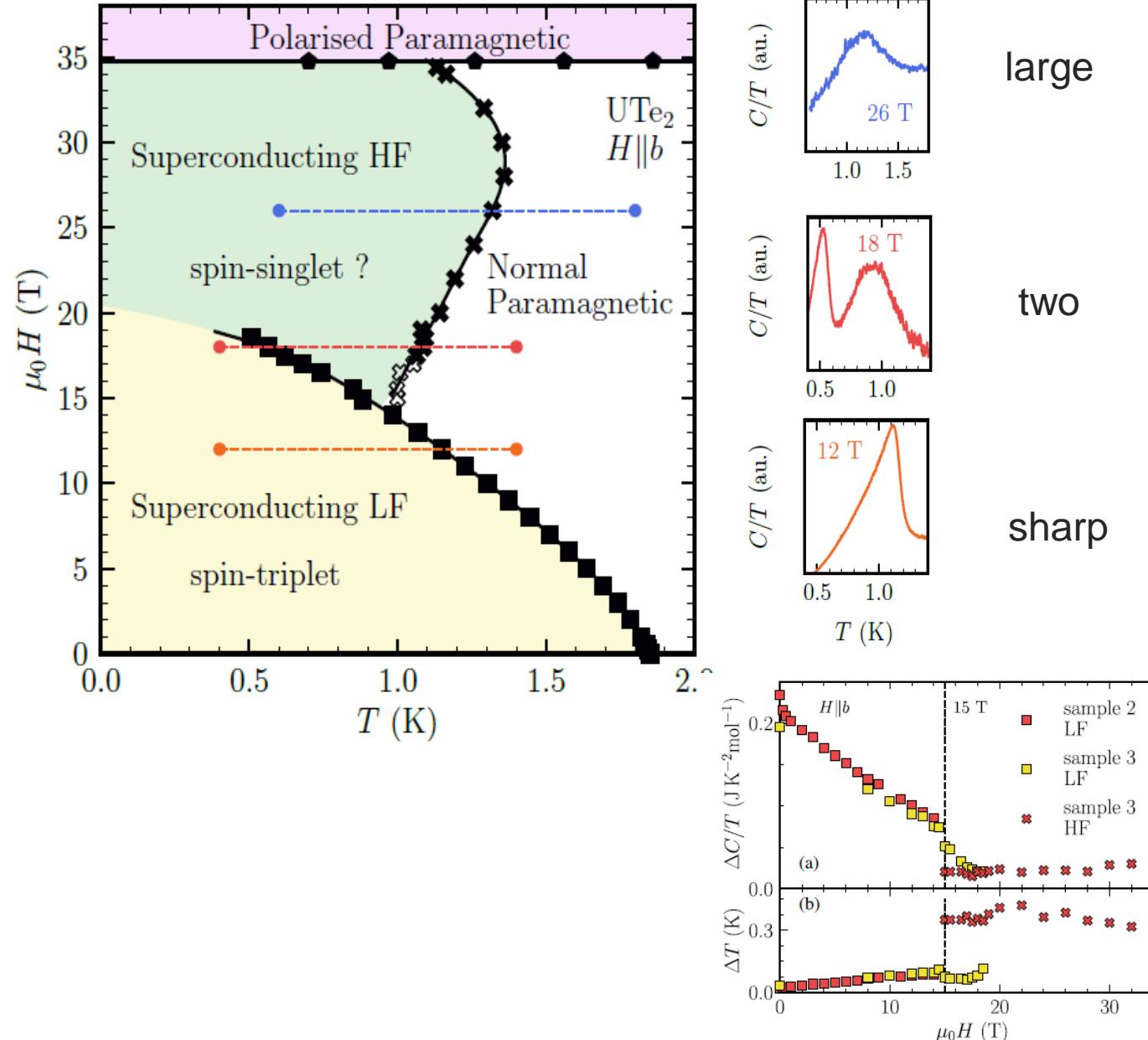


Determination of phase diagram by specific heat





Two distinct superconducting phases !



Two distinct superconducting phases with different pairing mechanisms:

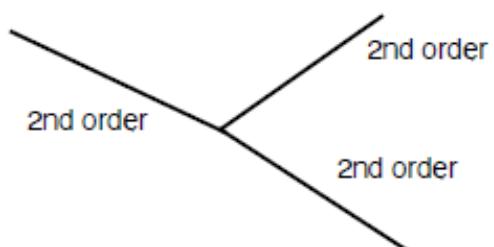
proposal :

spin-triplet at low field

new pairing mechanism (spin singlet) at high field, driven by critical fluctuations related to the metamagnetic transition ?

field enhancement of pairing mechanism

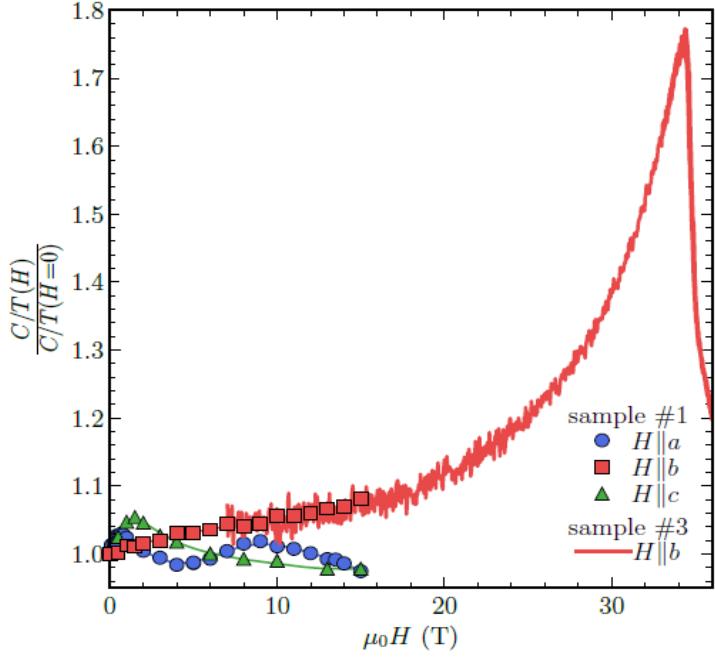
Problem :



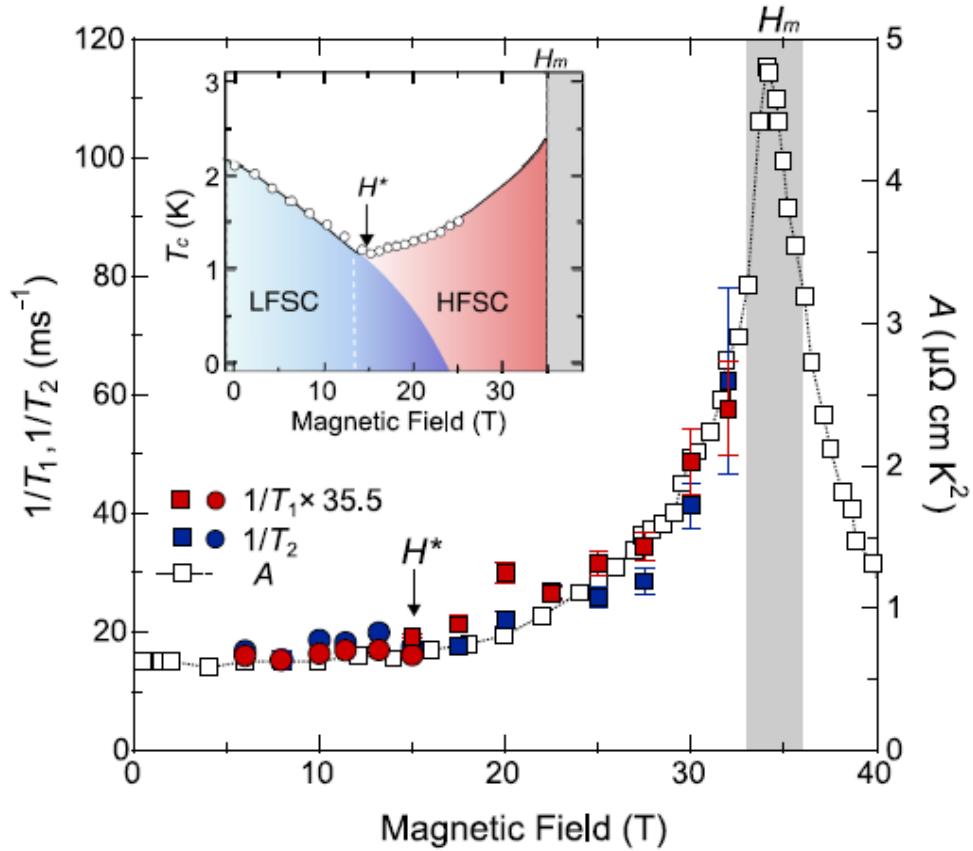
thermodynamically forbidden



Enhancement of fluctuations approaching the metamagnetic transition

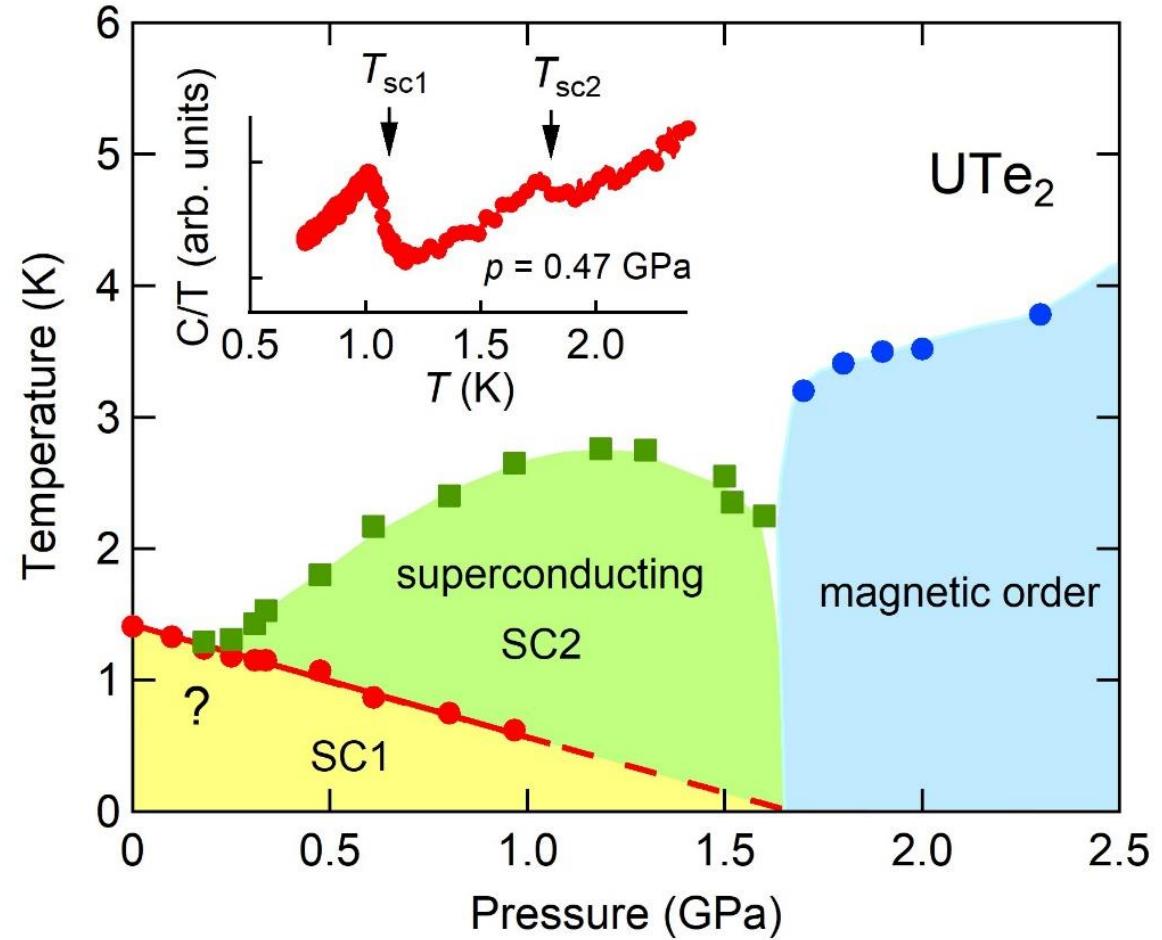


Strong enhancement of the γ coefficient on approaching H_m
⇒ strong enhancement of magnetic fluctuations



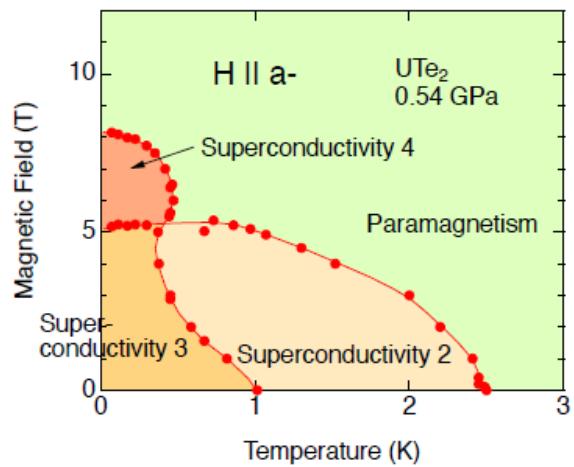


High pressure phase diagram



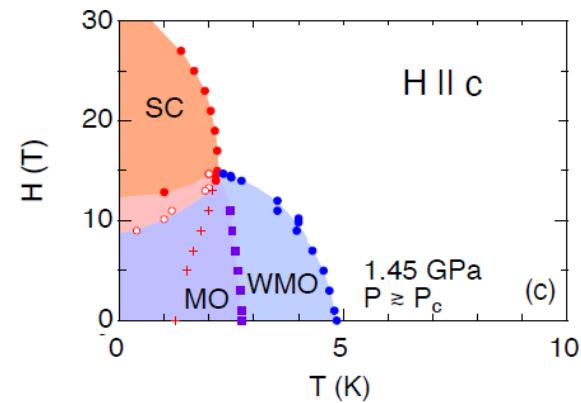
- from ac calorimetry :

for $H \parallel a$: multiple superconducting phases



Aoki et al. JPSJ **89**, 053705 (2020)
Aoki et al. JPSJ **90**, 074705 (2021)

for $H \parallel c$: field induced SC

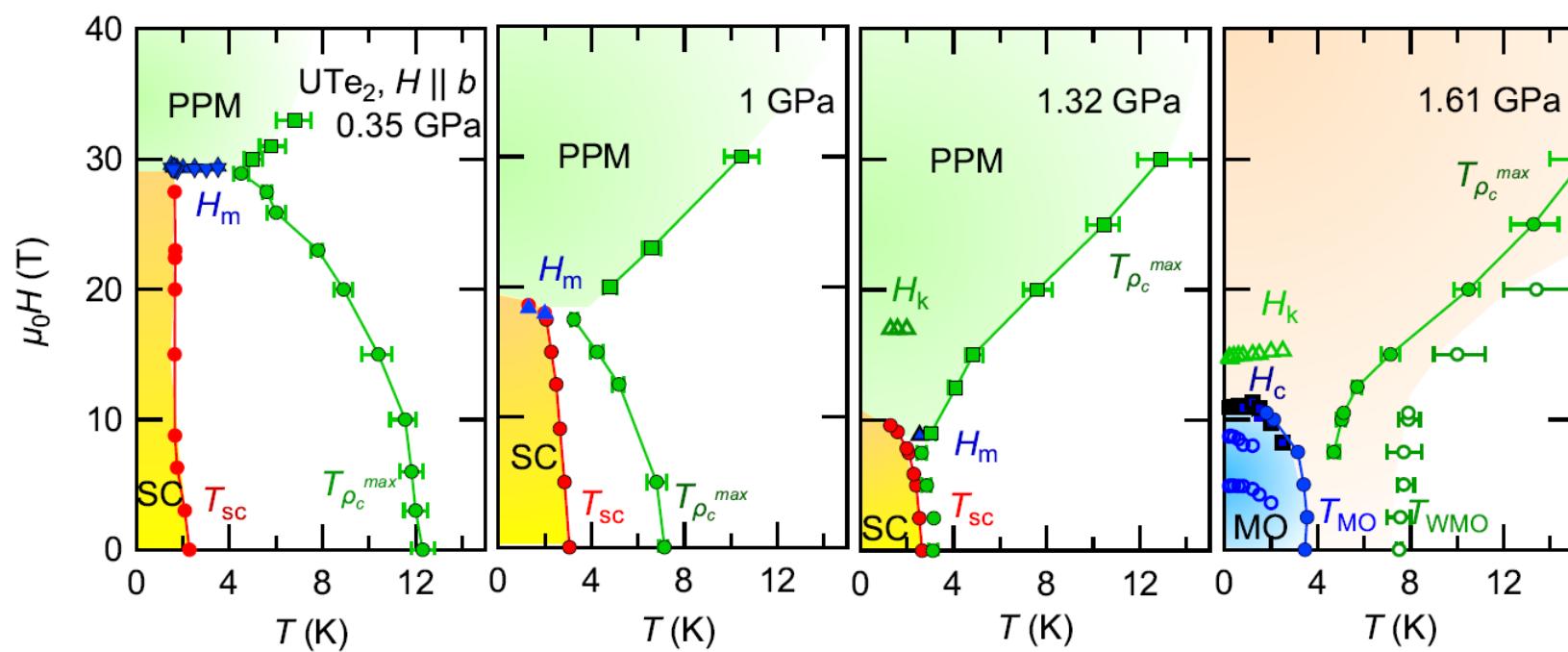
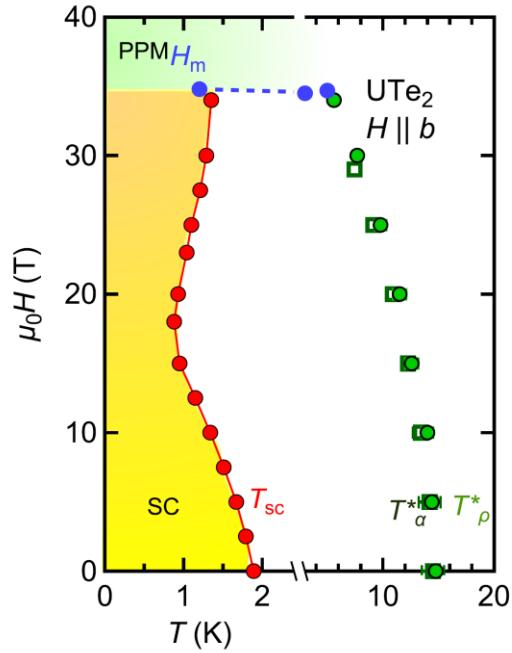


- antiferromagnetic order above p_c
 $\mathbf{k}_m = (0.07, 0.33, 1)$ close to the vector of AF fluctuations

Knafo et al. PRX under review



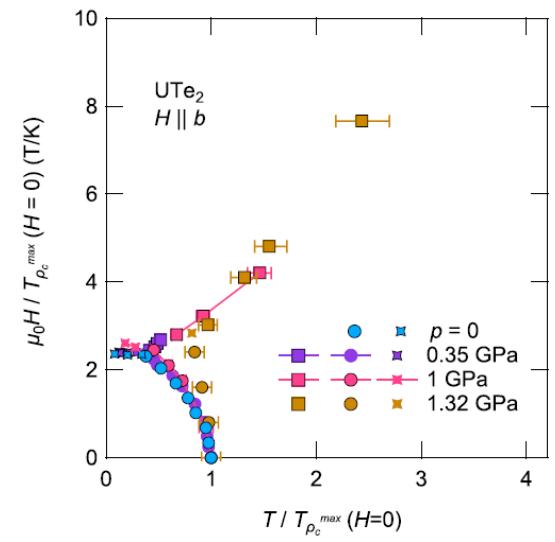
High pressure $H \parallel b$ axis



H_m scales with T^*

Scaling by T^* of phase diagram

Knebel et al., Phys. Rev. B 109, 155103 (2024)





Ac calorimetry $H \parallel b$ under pressure (end may 2024)

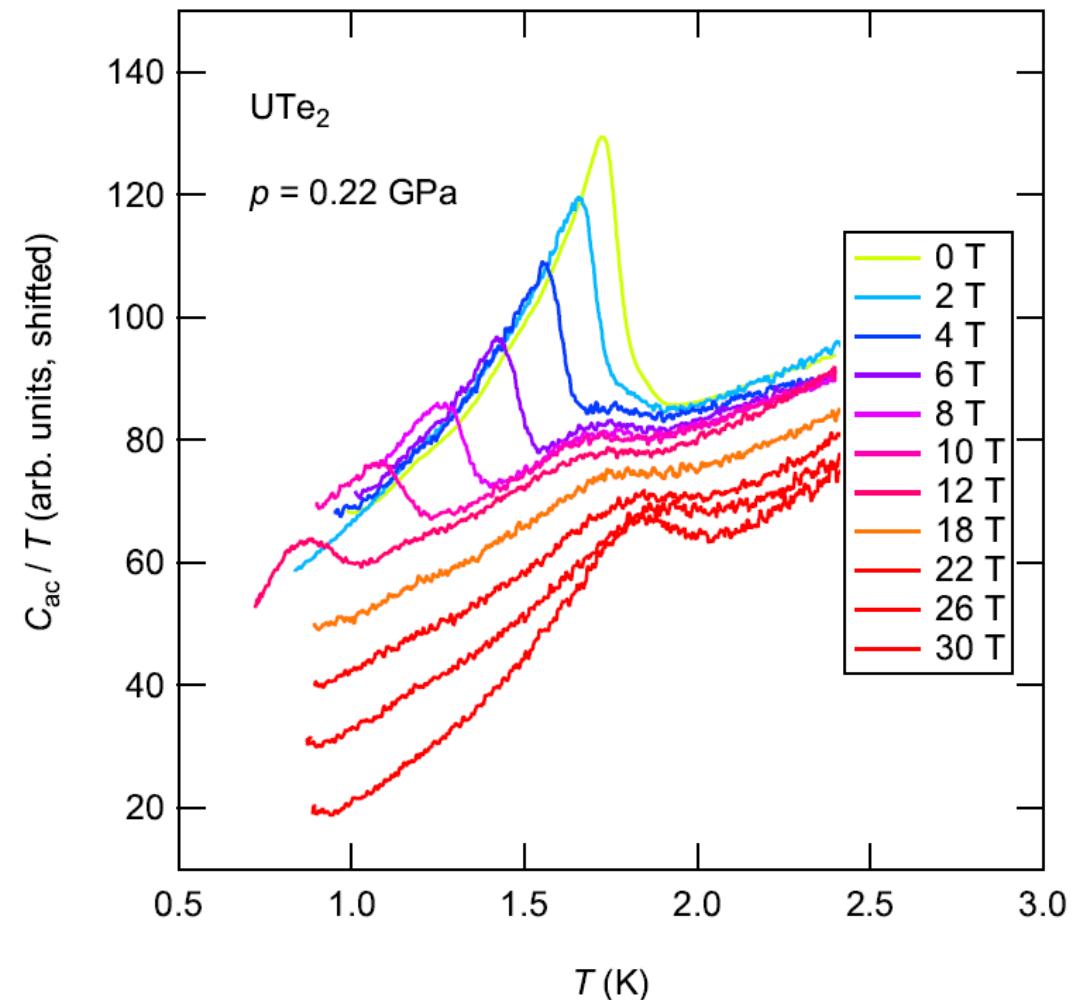
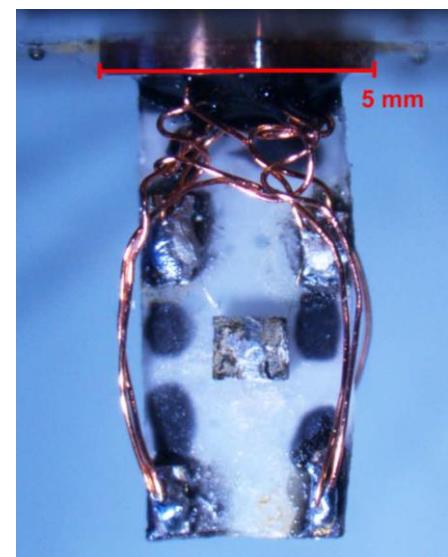
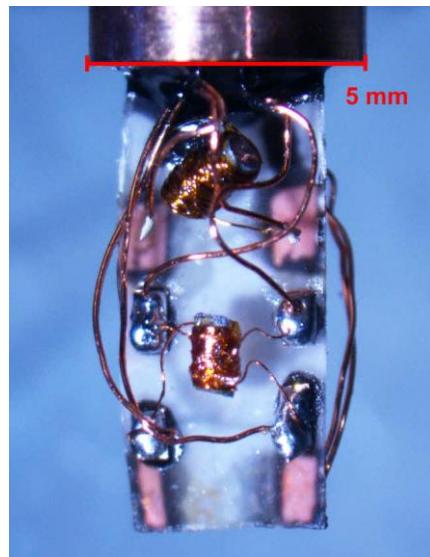
Experimental set up:

20 mm Cu/Be-NiCrAl pressure cell

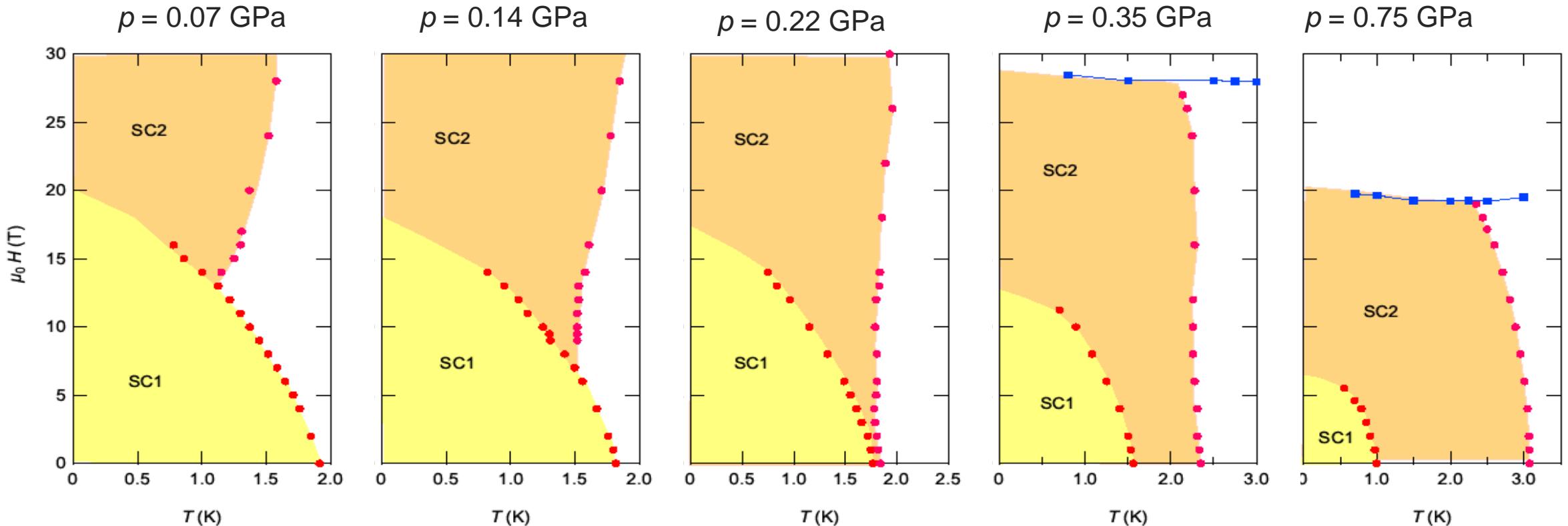
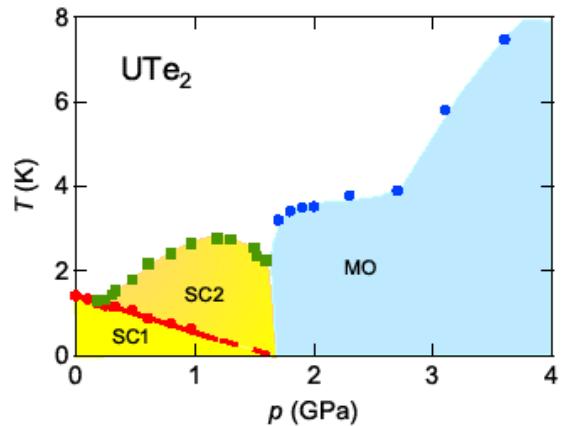
Daphne 7373 as pressure medium

^3He cryostat

Cernox thermometer, carbon past as heater



Phase diagrams from ac calorimetry



High temperature superconducting phase under pressure corresponds to high field phase at ambient pressure.

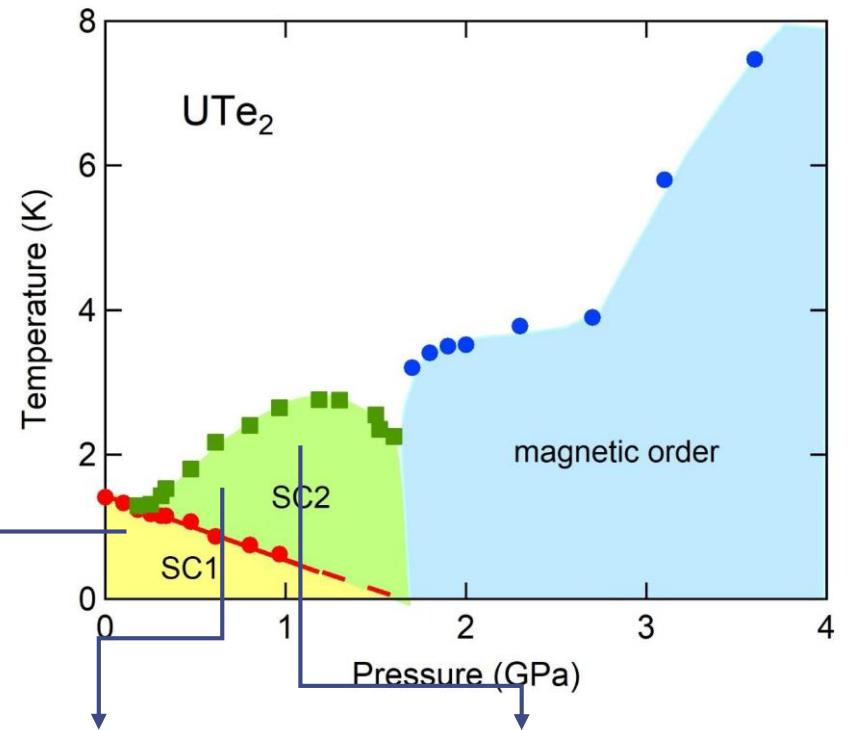
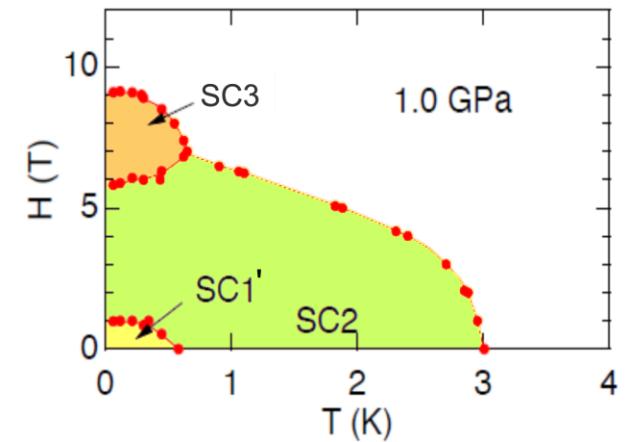
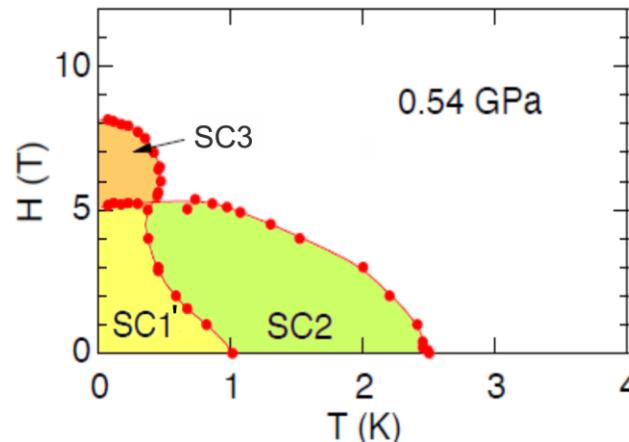
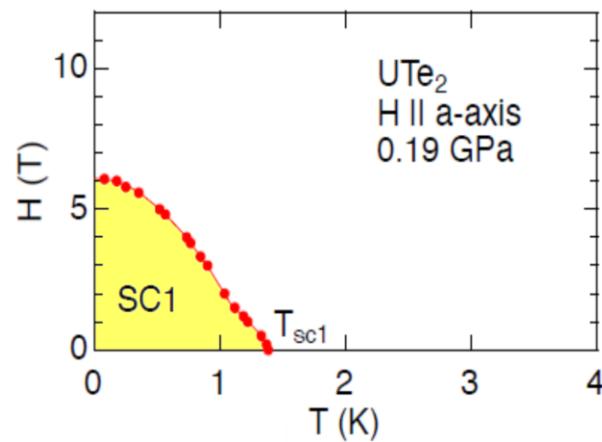


UTe₂ : Superconductivity tuned by pressure, $H \parallel a$

Under pressure,
“high temperature phase” close
to paramagnetic limit ($H//a$)

Braithwaite et al. Comm. Phys. 2019

Aoki et al. JPSJ 2020
Knebel et al. JPSJ 2020





UTe₂: progress in crystal growth

Chemical Vapor Transport optimization

P. F. S. Rosa et al. Comm. Materials 3, 33 (2022)

Molten Salt Flux method (2022)

H. Sakai et al. Phys. Rev. Materials 6, 073401 (2022)

$$T_c \rightarrow 2.1 \text{ K}$$

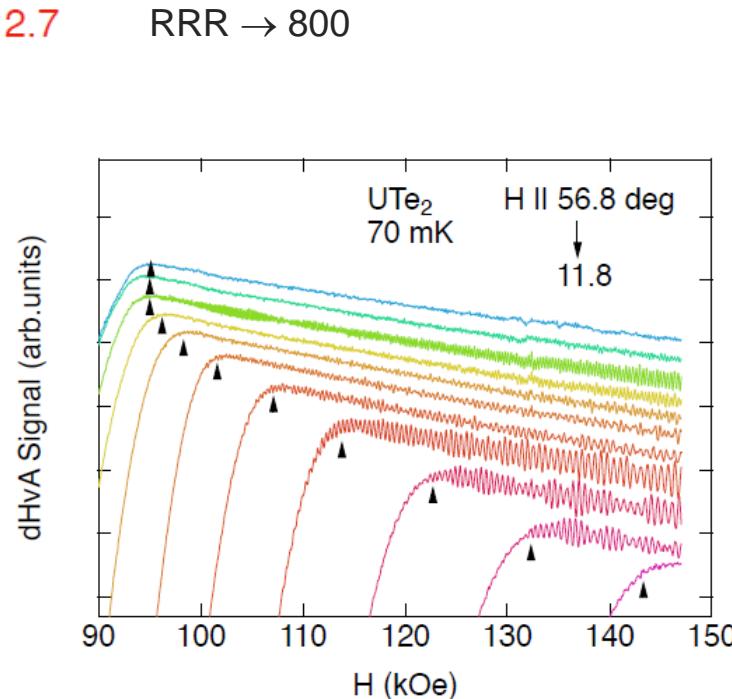
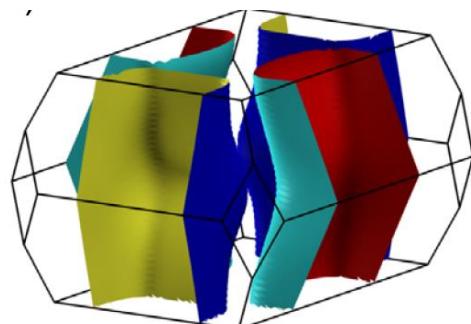
$$\gamma_0/\gamma_N \rightarrow 0$$

$$\Delta C/\gamma T_c \rightarrow 2.7$$

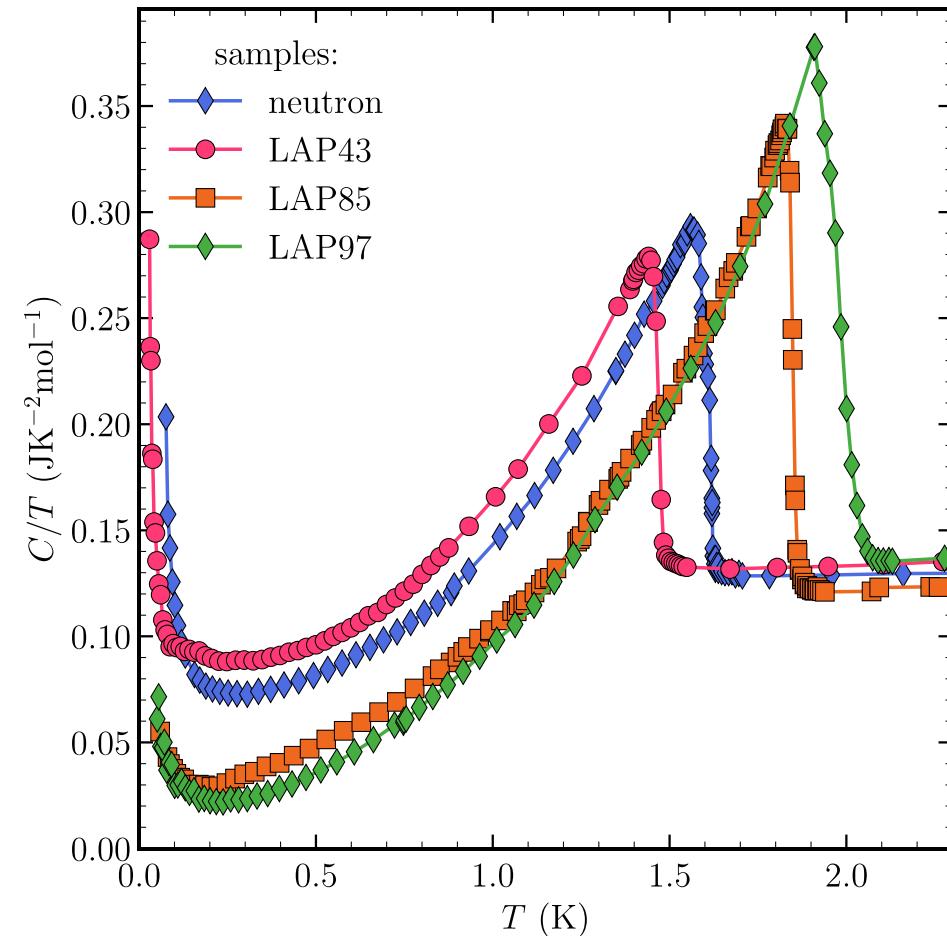
$$\gamma_0/\gamma_N \rightarrow 0$$

observation of dHvA

Quasi-2D Fermi
surfaces with strong
corregation



D. Aoki, et al. JPSJ 91, 083704 (2022)
confirmed by Cambridge group A.G. Eaton et al

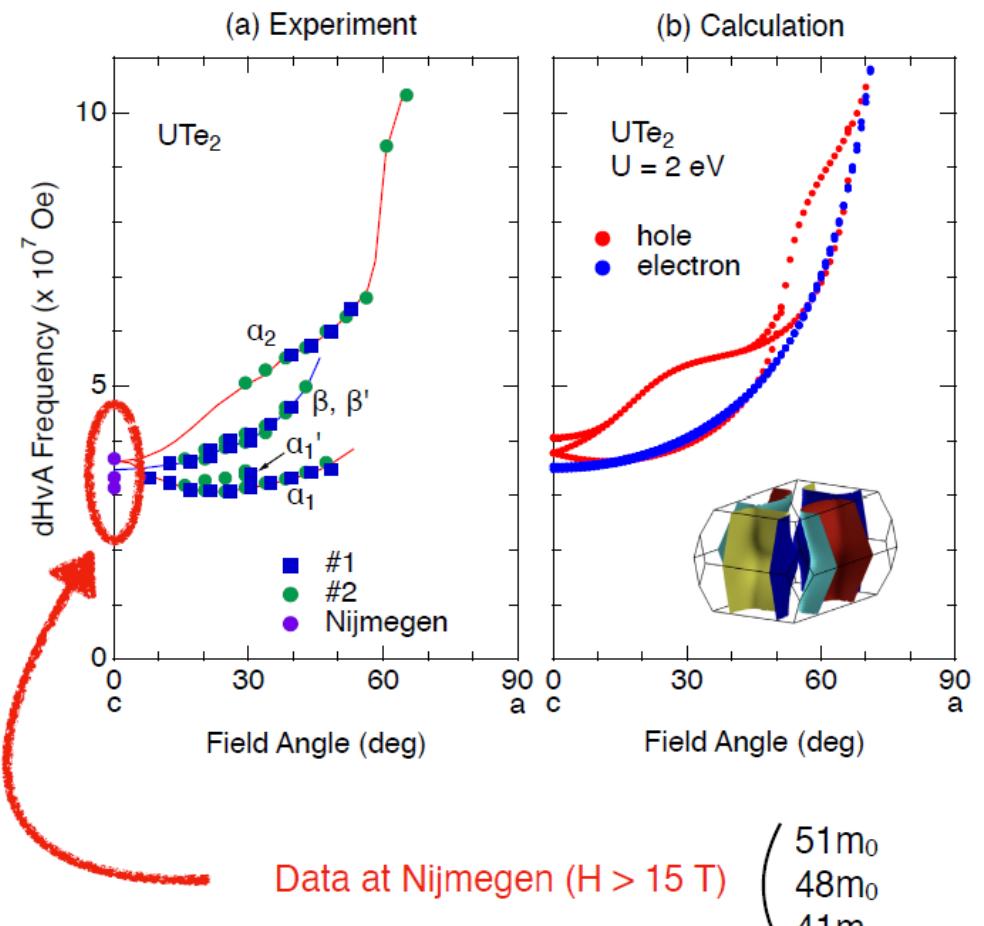
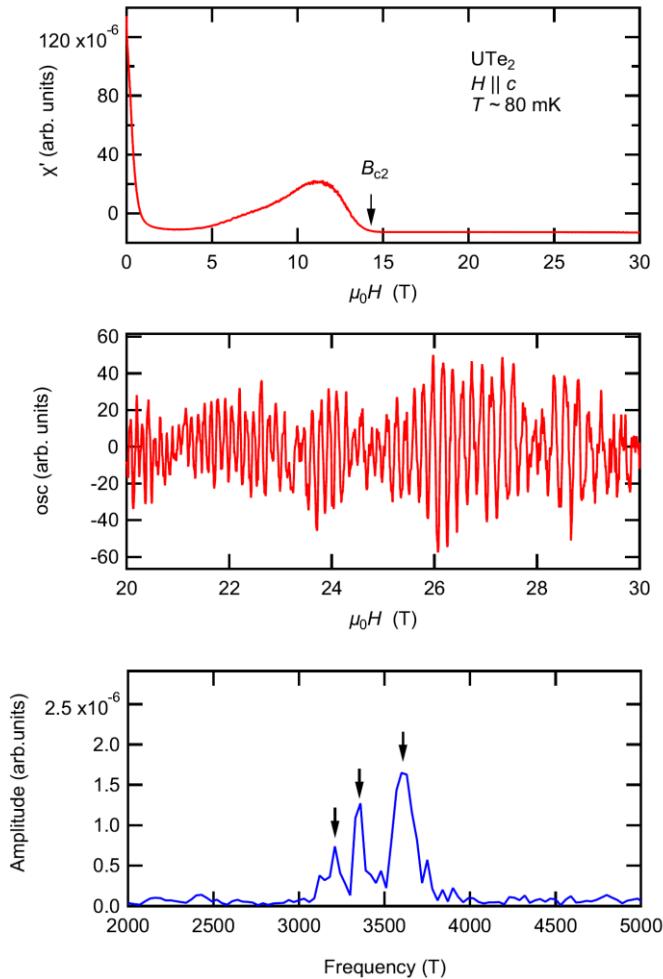


Studies on crystal LAP85:

- $T_c \sim 1.85 \text{ K}$
- $\frac{\Delta T_c}{T_c} \sim 0.01$
- $\frac{\gamma_0}{\gamma_N} \sim 0.1 - 0.03$



Quantum oscillations in high field



Branch	Experiment		Calculation	
	F ($\times 10^7$ Oe)	m_c^* (m_0)	F_b ($\times 10^7$ Oe)	m_b (m_0)
$\theta = 26$ deg				
α_1	3.08	32	3.80	3.0
β	3.99	48	3.93	2.5
$\theta = 29$ deg				
α_1	3.15	33	3.89	3.1
α'_1	3.38	40		
β	3.94	57	3.98	2.5
β'	4.09	34	4.05	2.6
α_2	5.05	39	5.36	2.4
$\theta = 44$ deg				
α_1	3.31	32	4.84	4.3
α_2	5.85	36	5.70	2.9
$\theta = 61$ deg				
α	9.41	55	8.95	3.6

cylindrical FS

$$\gamma = \frac{k_B^2 V}{6\hbar^2} \sum m^* k_z$$

Important information on the electronic structure.
Main Fermi surface branches are detected.
Existence of a small pocket still under discussion.

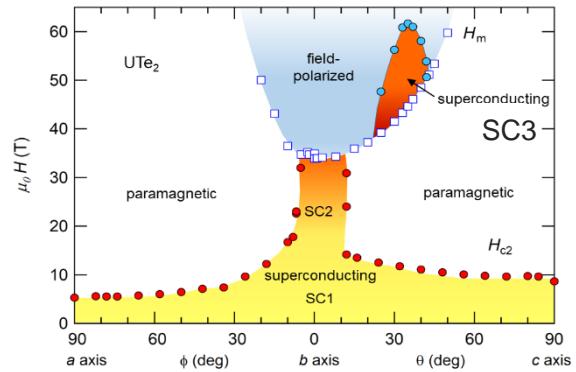
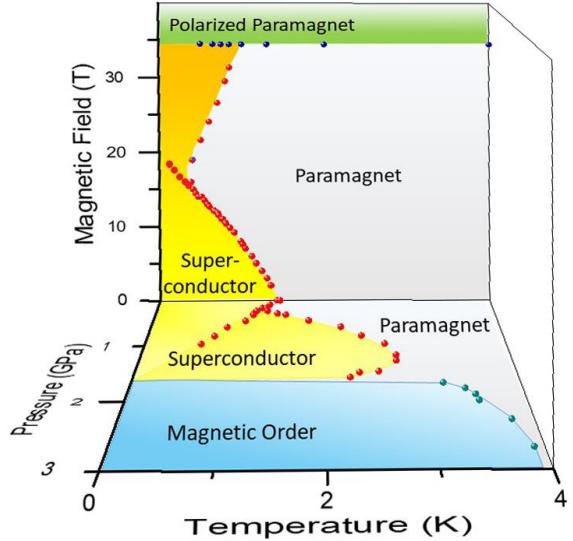
Conclusions



- Extremely rich superconducting phase diagram at ambient pressure
- Multiple superconducting phases
- Antiferromagnetic order under high pressure
- Two cylindrical Fermi surfaces determined by dHvA experiments

many open questions:

- nature/symmetry of the superconducting order parameter in different phases
- triplet \Leftrightarrow singlet \Leftrightarrow mixture of singlet triplet
- Pairing mechanism, only AF fluctuations competing with “local” FM
- Why so many superconducting phases
- Existence of PDW coupled to CDW



Wishes : “static” fields to reach SC3 superconducting phase !

Many thanks to EMFL for continuous support of experiments !