Heavy-Fermion Superconductivity, Magnetism and Their Relationship

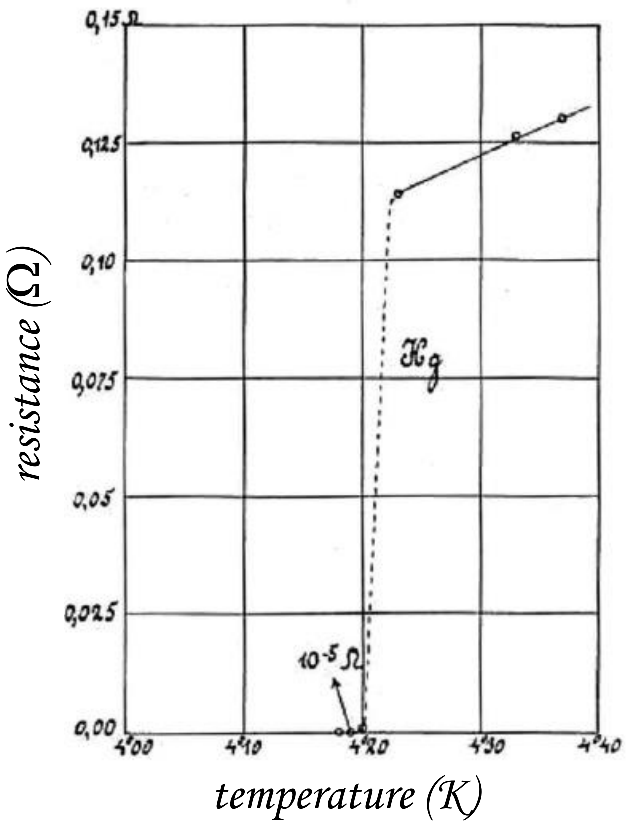
J. D. Thompson
Los Alamos National Laboratory

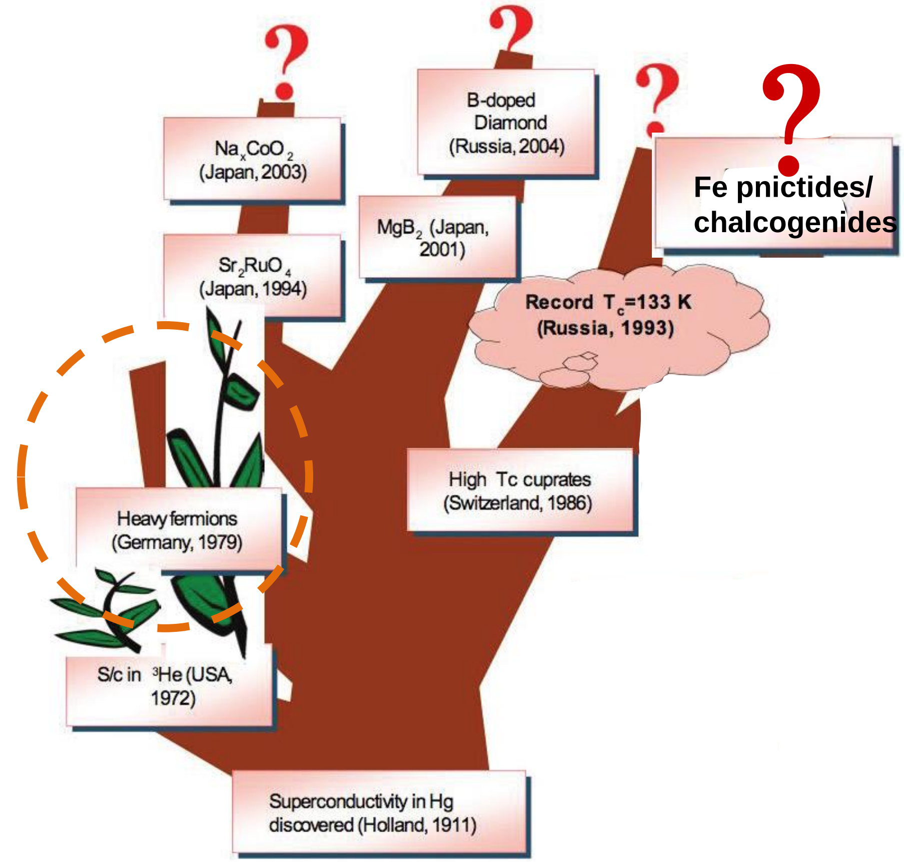
Outline:

-- introduction
-- U-based heavy-fermion superconductors
-- $CeIn_3$ and derivatives
-- summary

Spring School on the Heavy Fermions
and Quantum Phase Transitions
Zhejiang University
April 12-14, 2015

the beginning





Heike Kamerlingh Onnes (1911)

♦ 'trunk' of a tree of superconductivity research started by the discovery of Onnes in 1911

♦ subsequently many 'limbs' but focus on the 'limb' of heavy-fermion materials, which also connects to the cuprate and iron-pnictide/chalcogenide limbs

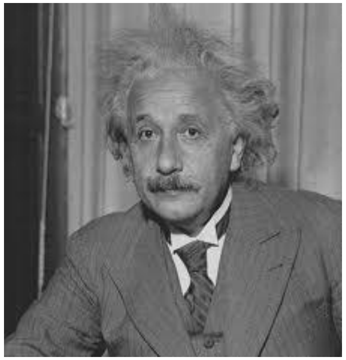
superconductivity

- superconductivity recognized as one of the most important problems in theoretical physics through the first half of the 20th century

N. Bohr
Nobel 1922

L. Landau
Nobel 1962

R. Feynman
Nobel 1965



A. Einstein
Nobel 1921

W. Heisenberg
Nobel 1932

- founding fathers of quantum mechanics and modern theory of physics – each fascinated by the problem and worked on it

superconductivity

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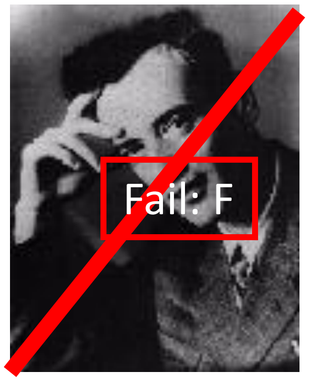
N. Bohr
Nobel 1922



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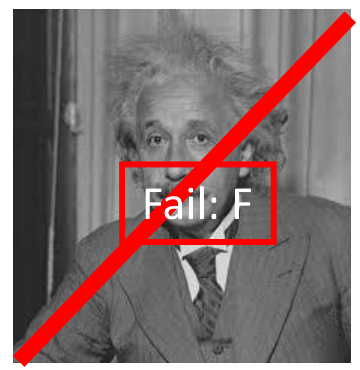


A. Einstein
Nobel 1921

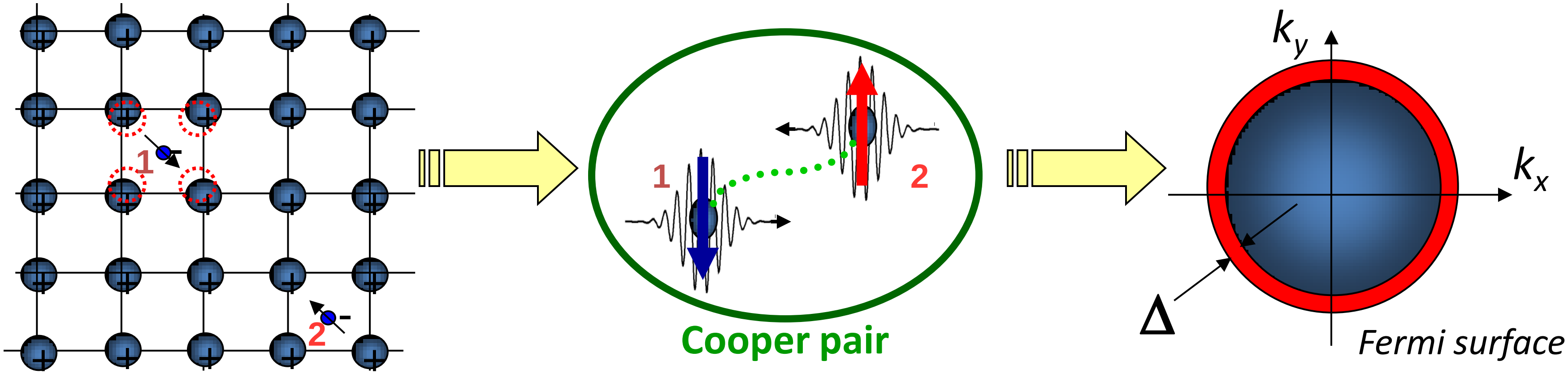
W. Heisenberg
Nobel 1932

- founding fathers of quantum mechanics and modern theory of physics—all failures

- Richard Feynman: "No one is brilliant enough to figure it out" (but BCS did in 1957)

- ***magnetism*** <u>and</u> superconductivity, a similarly challenging unsolved problem for the 21^{st} century

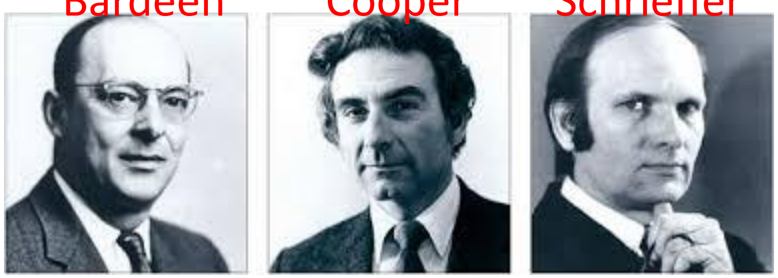
BCS: conventional superconductivity



♦ temporary lattice distortion of positively charged ions responding to electron 1 ⇒ phonon-mediated attractive interaction for electron 2 with opposite spin and momentum

♦ creates electron (Cooper) pairs with zero net momentum and spin (L=0, S=0) whose energy is lower by an amount Δ relative to unpaired electrons ⇒ finite energy gap $\Delta \propto T_c \propto \theta_D \exp(-1/\lambda)$ between paired and unpaired electrons ⇒ below T_c, thermal-activated T-dependence of any property that depends on $N(E_F)$

Bardeen Cooper Schrieffer



♦ some basic ingredients of theory:

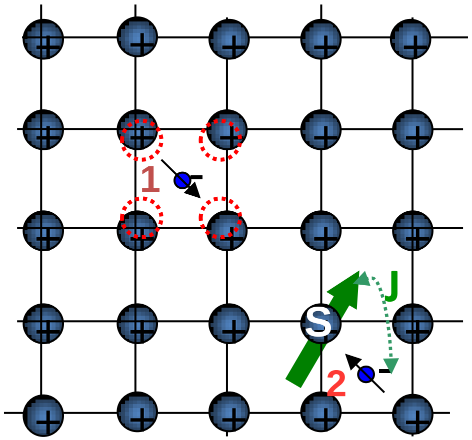
-- bosonic excitations (phonons) couple electrons within $\sim k_B\theta_D$ of E_F

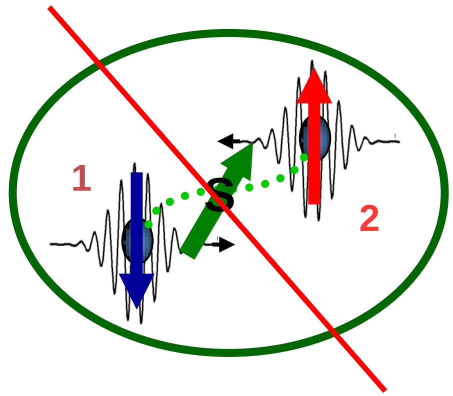
-- $k_B\,\theta_D << E_F$ (E_F/k_B=the electron degeneracy temperature)

-- coupling strength $\lambda \approx N(E_F)V$, where V is the attractive pair potential

-- for large $N(E_F)$ Coulomb repulsion important and attractive interaction weakened

magnetism is 'bad'



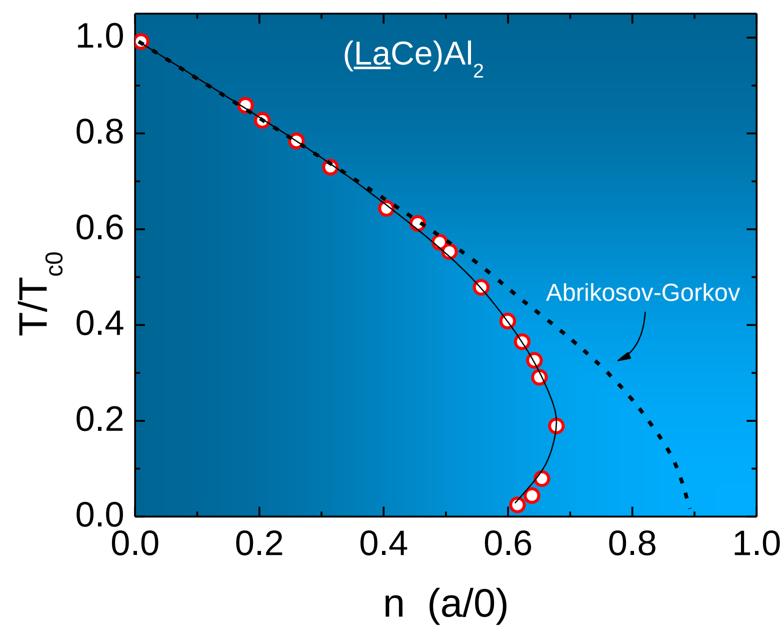


♦ introducing a tiny number of magnetic impurities with spin **S** ⇒ interaction (J**S•s**) of spin **S** with conduction electron spin **s** breaks time-reversal symmetry of Cooper pairs and $T_c \rightarrow 0$ (Abrikosov & Gorkov, JETP **12**, 1243 (1961))

$$(\mathbf{k},\downarrow),(-\mathbf{k},\uparrow) \neq (-\mathbf{k},\uparrow),(\mathbf{k},\downarrow)$$

♦ rapid suppression of BCS superconductivity in $LaAl_2$ by magnetic Ce impurities

♦ initial decrease in $T_c(n)$ consistent with theory of Abrikosov-Gor'kov: $T_c \rightarrow 0$ with less that 1% Ce

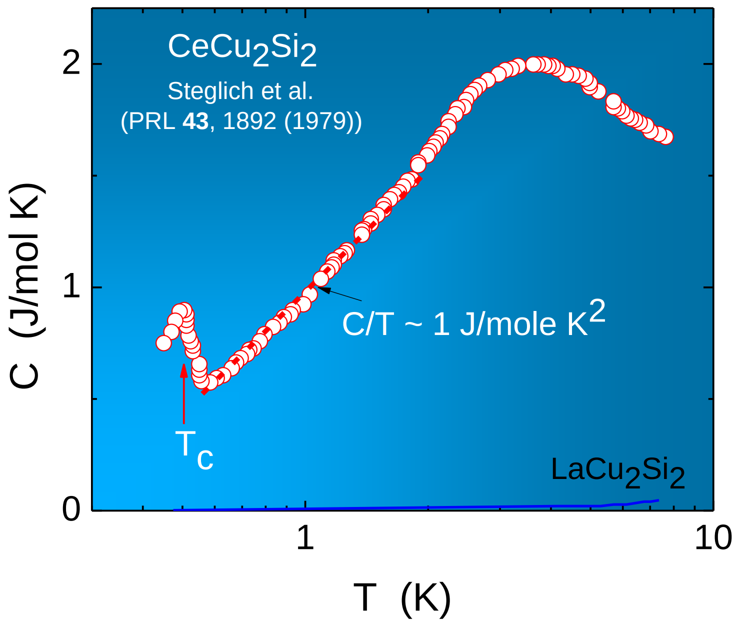


M. B. Maple et al., Solid State Commun. **11**, 829 (1972)

A. Abrikosov

L. Gor'kov

$CeCu_2Si_2$



- ♦ correspondingly huge jump in C at T_c

Frank Steglich

♦ discovery by Steglich et al. in 1979 of superconductivity near 0.5 K in $CeCu_2Si_2$ in which 1/5 of all atoms (Ce) carry a large moment; should NOT be superconducting

♦ above T_c , huge T-linear specific heat: $C = \gamma T$, with $\gamma \approx 1$ J/mol $K^2 \Rightarrow$ huge $N(E_F) \Rightarrow$ strong Coulomb repulsion, detrimental to BCS superconductivity

♦ in Sommerfeld theory of metals, $\gamma \propto m^* \propto 1/T_F$, where m^* is the effective mass of itinerant electrons and T_F is the degeneracy temperature of electrons with mass m^*

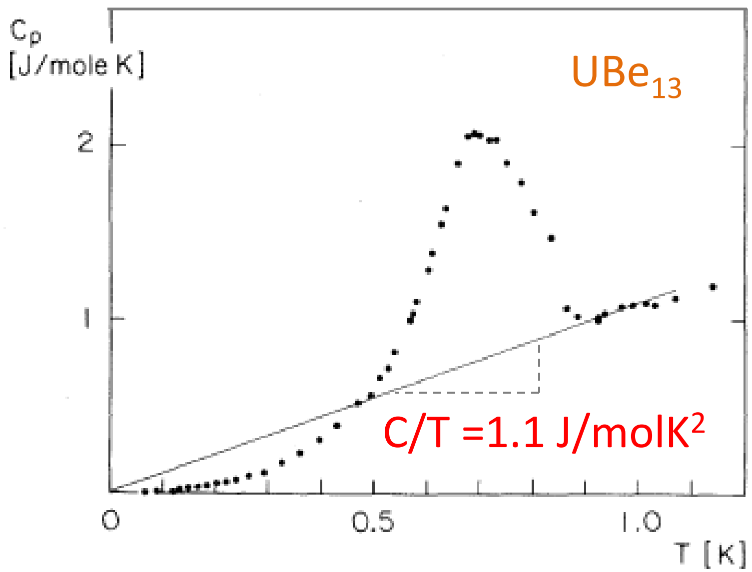
♦ compared to $LaCu_2Si_2$ where $\gamma \approx 5$**m**J/mol K^2 and $m^* \approx m_e$, electron (quasiparticle) mass in $CeCu_2Si_2$ is huge, ≈ 1/3 the mass of a proton; hence, the name 'heavy-fermion'

♦ very massive quasiparticles responsible for superconductivity

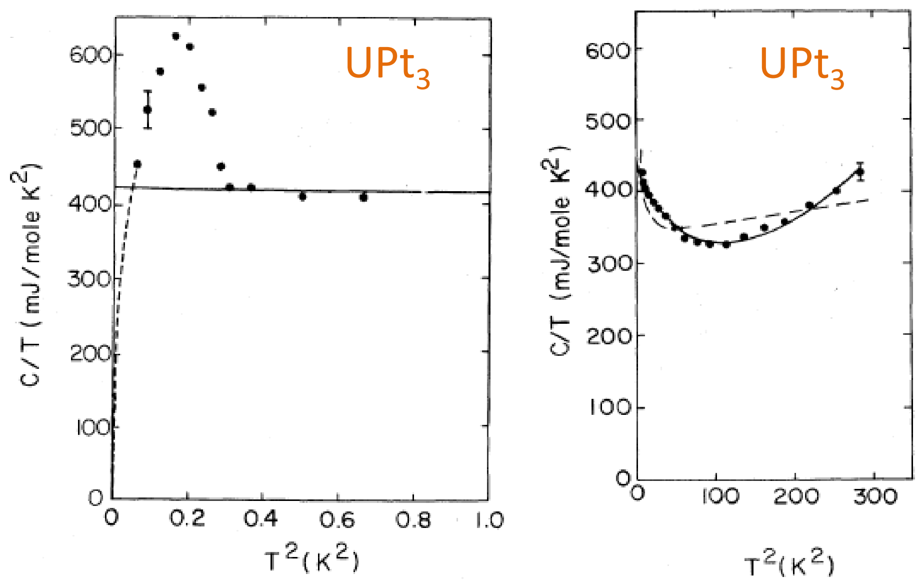
♦ key assumption in conventional theory of phonon-mediated superconductivity: $\theta_D/T_F << 1$; but, in $CeCu_2Si_2$, $\theta_D/T_F \approx 20! \Rightarrow$ *superconductivity cannot be described by conventional theory*

♦ further, $T_c \approx 0.05T_F \Rightarrow$ *high temperature superconductivity* (compare to conventional superconductors where $T_c \approx 10^{-3}T_F$) F. Steglich et al., PRL **43**, 1892(1979)

confirmation of heavy-fermion superconductivity



H. R. Ott et al. PRL **50**, 1595 (1983)



G. R. Stewart et al., PRL **52**, 679 (1984)

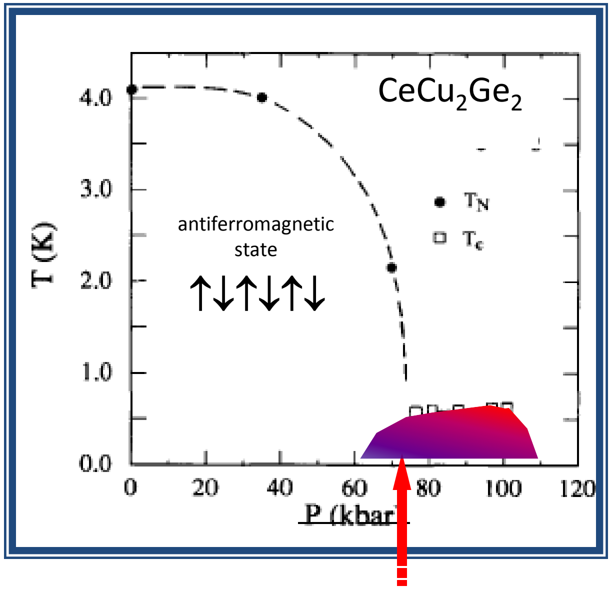
♦ discovery of heavy fermion superconductivity in UBe_{13} (1983) with γ comparable to that of $CeCu_2Si_2$ and soon thereafter in UPt_3 (1984) with large but smaller γ

♦ important confirmations of Steglich's principal conclusions $\Rightarrow$ doubts subside

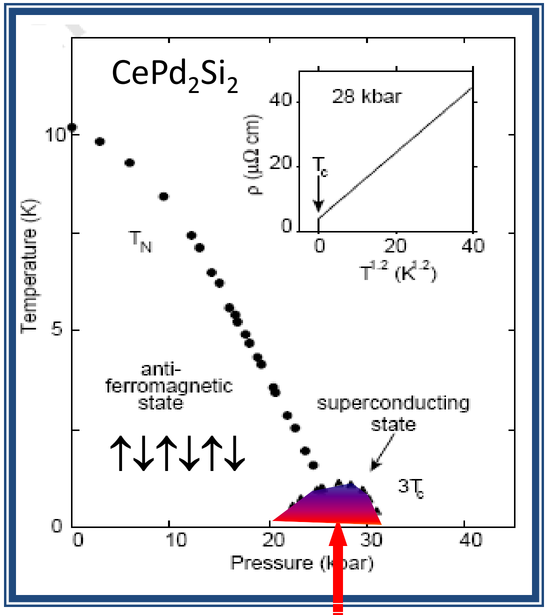
♦ again, no superconductivity in non-f-analogs $\Rightarrow$ f-derived magnetism necessary for superconductivity

♦ in UPt_3, a prominent $T^2 \ln T/T_{SF}$ contribution in addition to a large T-linear term to describe C/T; such a term, motivated by a spin fluctuation contribution in nearly ferromagnetic ^3He, and unusually large $\chi/\gamma \Rightarrow$ possibility of spin-triplet Cooper pairs in UPt_3

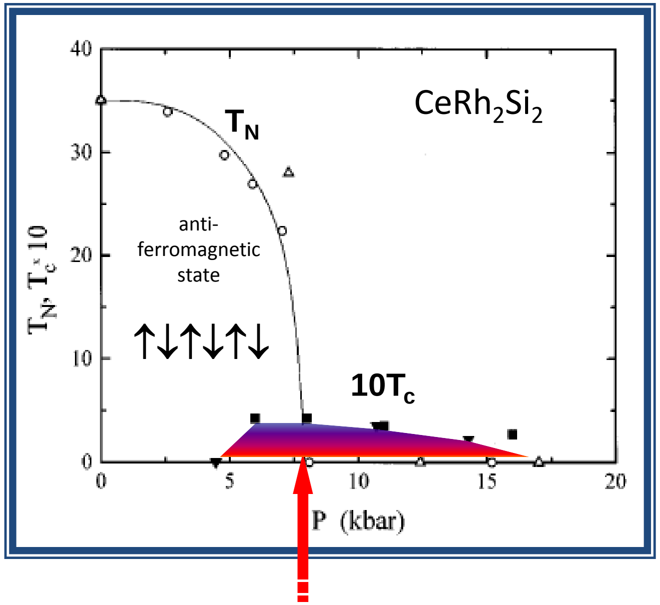
subsequent discoveries



D. Jaccard et al., Phys. Lett. A **163**, 475 (1992)



F. M. Grosche et al., Physica B **224**, 50 (1996)

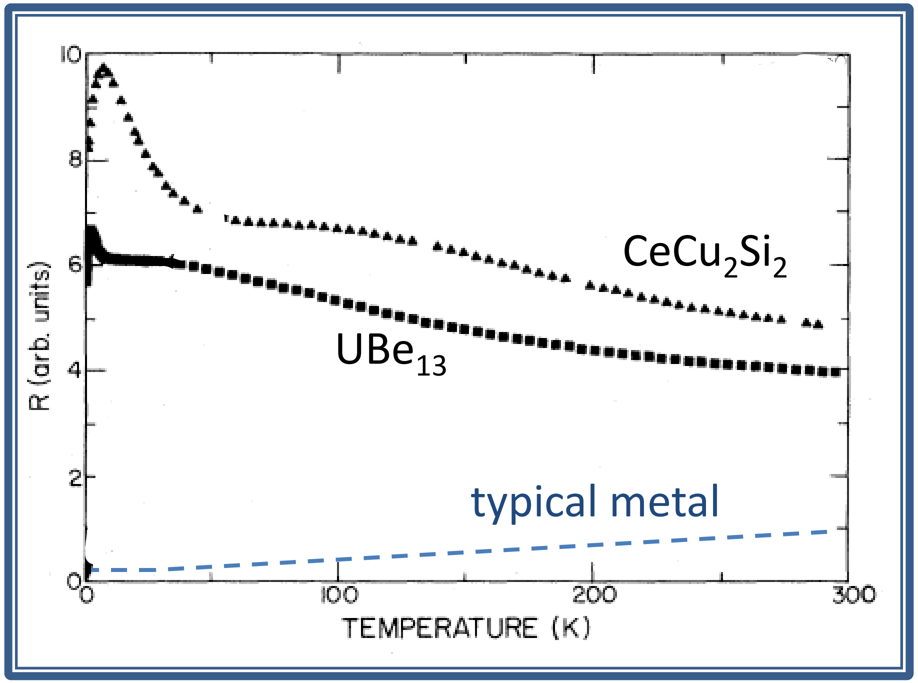


R. Movshovich et al., PRB **53**, 8241 (1996)

♦ appearance of superconductivity near a pressure-tuned T=0 antiferromagnetic-to-paramagnetic boundary in heavy-electron materials with the $CeCu_2Si_2$ structure type

♦ not specific to this structure type and absence of superconductivity in non-magnetic analogs based on La ⇒ something special about this boundary and now a specific direction for where to look for new examples

electrical transport



♦ unusual temperature dependence of electrical resistivity ⇒ unusual scattering mechanism

♦ not all but many heavy-fermion systems with similarly large ρ(300K) ~ many tens to a hundred or more $\mu\Omega$cm

♦ often one or more regions with $d\rho/dT < 0$ and a maximum at low temperatures

♦ reminiscent of a Kondo-impurity like resistivity, with $\rho(T) \propto -\ln T/T_K$ for $T \sim T_K \propto (1/E_F)\exp(-1/2JN_0)$ and exchange $J \propto |V_{kf}|^2 U/E_f(E_f+U) < 0$ (Schieffer&Wolff, Phys. Rev. **149**, 491 (1966))

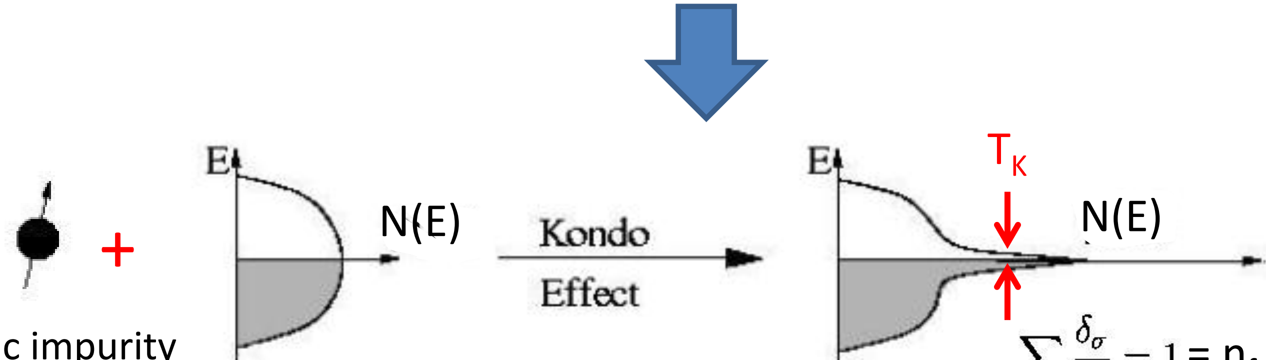
Jun Kondo

♦ unlike a Kondo impurity $\rho(T)=\rho_0(1-(T/T_K)^2)$ [$\rho(0)= \rho_0 \sin^2\delta$, where scattering phase shift $\delta=\pi/2$ and ρ_0 is a material-dependent constant/impurity], $d\rho/dT > 0$ for $T < T_{max}$

♦ the consequence of a periodic lattice of Ce or U ‘Kondo impurities’

Suhl resonance and density of states

♦ process of resonant scattering leads to an Abrikosov-(Nagaoka)-Suhl resonance in the density of states:



magnetic impurity

$$\sum_\sigma \frac{\delta_\sigma}{\pi} = 1 = n_f$$

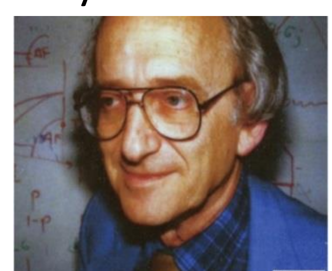
required by Friedel sum rule

♦ width of resonance: $\sim k_BT_K \Rightarrow N_0 \sim 1/k_BT_K$ and hence $\gamma(0)$/impurity $\propto 1/k_BT_K \sim 1/k_BT_F \propto m^*/m_e \Rightarrow$ electrons get ‘heavy’

♦ for $T >> T_K$, magnetic electron on impurity localized and ‘small’ Fermi volume determined by conduction electrons of host

♦ for $T << T_K$, magnetic electron part of the Fermi sea $\Rightarrow$ Fermi volume ‘large’ and spin of conduction electrons collectively and exactly compensate spin of impurity $\Rightarrow$ a Pauli paramagnetic ground state with large $\chi(0)$/impurity $\sim \gamma(0)$/impurity

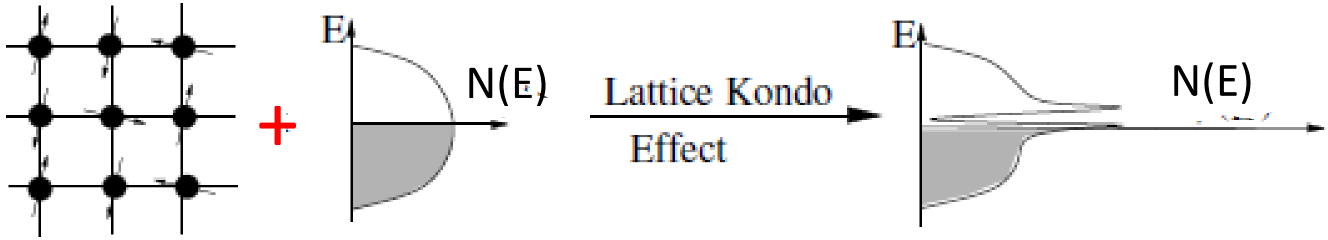
Jacques Friedel



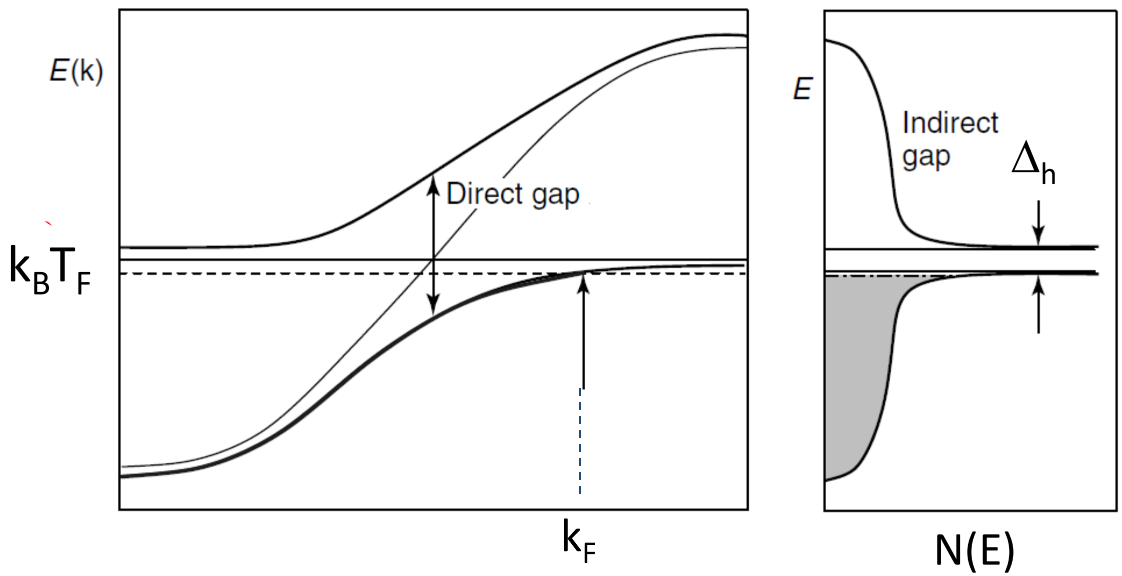
Harry Suhl

the Kondo lattice

lattice of Kondo ions



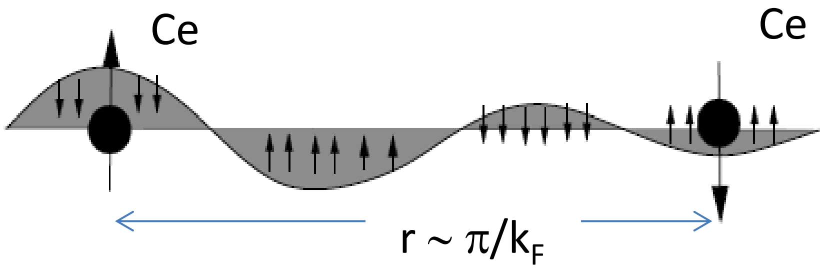
from P. Coleman 'Local moment physics in heavy electron systems'



♦ hybridization between f-electrons and conduction electrons ⇒ an (indirect) hybridization gap with peaks of width ~ k_BT_K on each side of the gap

♦ if T_K very small, band of conduction states very narrow ⇒ Coulomb repulsion among electrons produce a strongly correlated response, even magnetic order

competing interactions



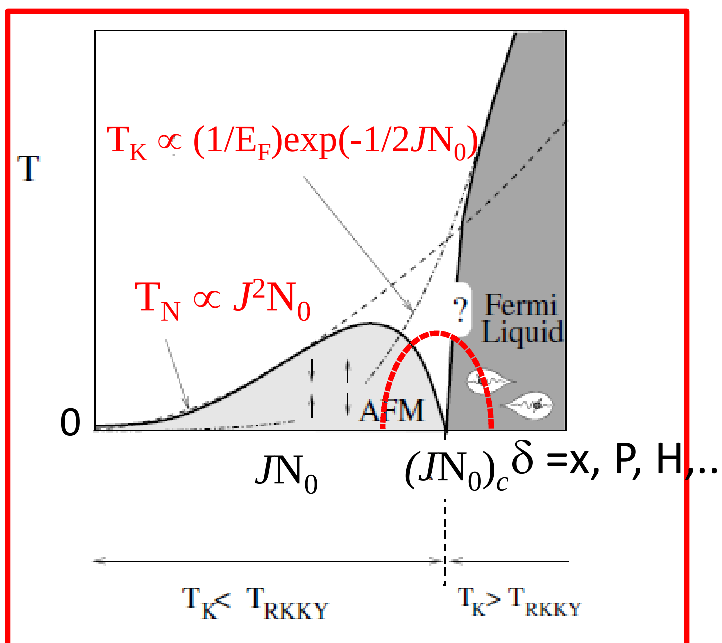
♦ in the absence of a Kondo effect, a periodic array of magnetic moments in a metal interact (indirectly) through the oscillating (Friedel) polarization of conduction electron spins: the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction $J_{RKKY} \propto -J^2 N_0[\cos 2k_F r]/|k_F r|^3$

♦ negative $J_{RKKY} \Rightarrow$ anti-parallel alignment of moments $\Rightarrow$ Néel order at $T_N \propto J^2 N_0$

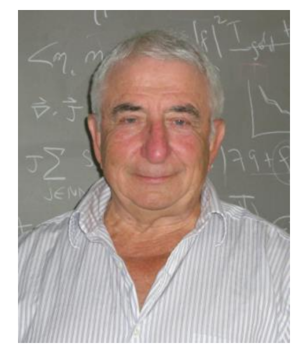
♦ allow simultaneous Kondo and RKKY interactions: Kondo 'wants' a non-magnetic state below $T_K \propto (1/E_F)\exp(-1/2JN_0)$ but RKKY 'wants' long range antiferromagnetic order $\Rightarrow$ Doniach model

♦ model: a 1-dimensional necklace of, eg., Ce atoms

♦ as a function of a non-thermal tuning parameter δ = x, P, H,.., $T_N \rightarrow$ 0 at a critical value of JN_0

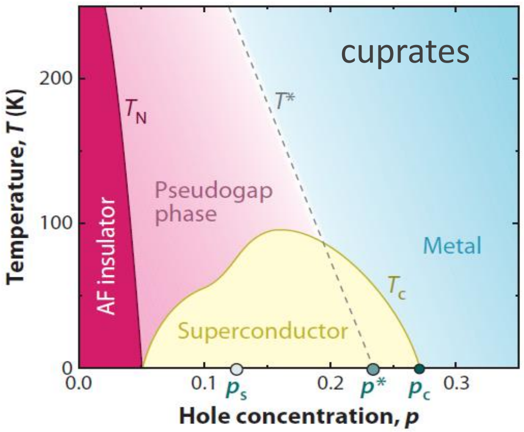


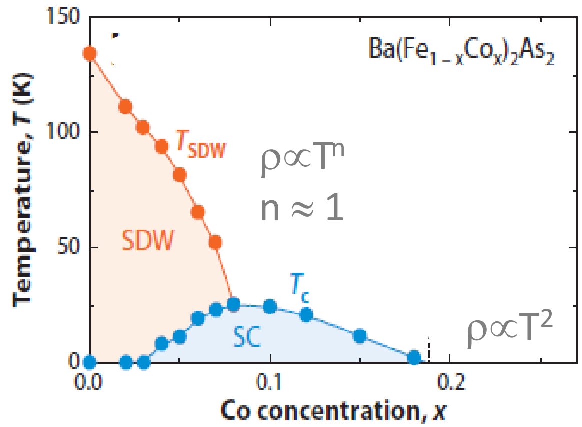
Sebastian Doniach

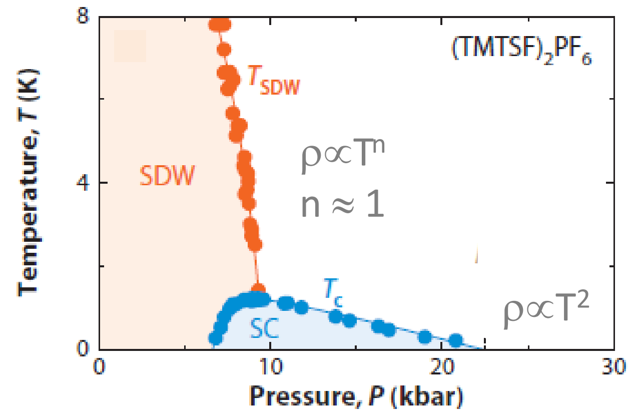


♦ *critical value of JN_0 special – a T=0 magnetic/non-magnetic boundary (a quantum critical point) where magnetic fluctuations proliferate and HF superconductivity often appears*

similar phase diagrams

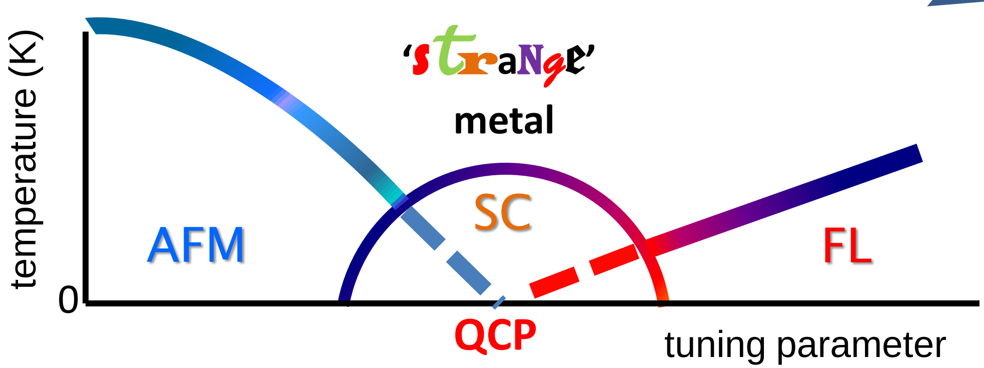






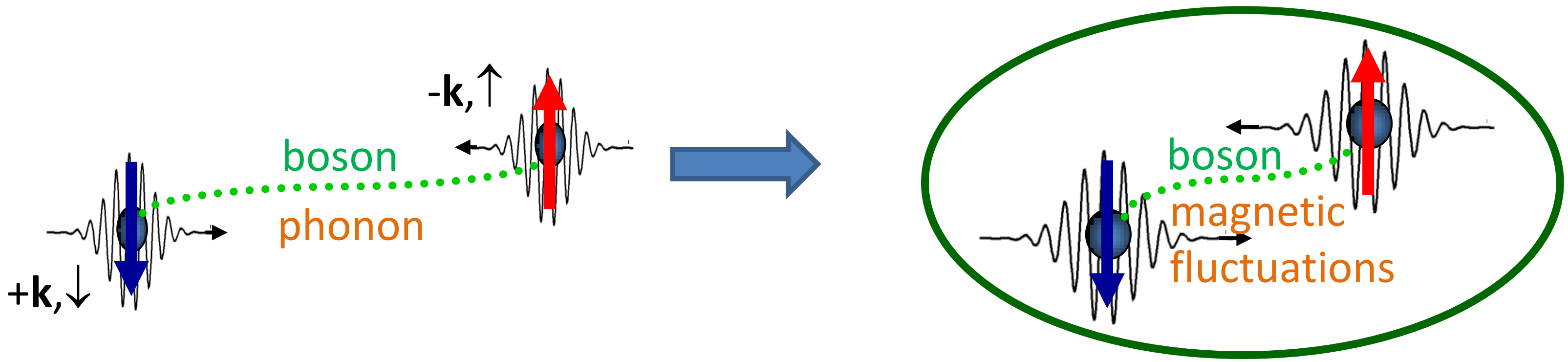
♦ underlying physics different in detail, but like the heavy-fermion materials, a dome of superconductivity that emerges in proximity to a T=0 antiferromagnetic boundary

♦ above the dome, a strange metallic state with resistivity $\rho \propto T^n$, where n = 1-1.5, that evolves into a Fermi-liquid T^2 dependence



♦ a generic relationship *suggesting* that magnetism is important for superconductivity; more specifically that T=0 (quantum) fluctuations of magnetic order, responsible for strange metallic behavior, also may produce an unconventional form of Cooper pairing

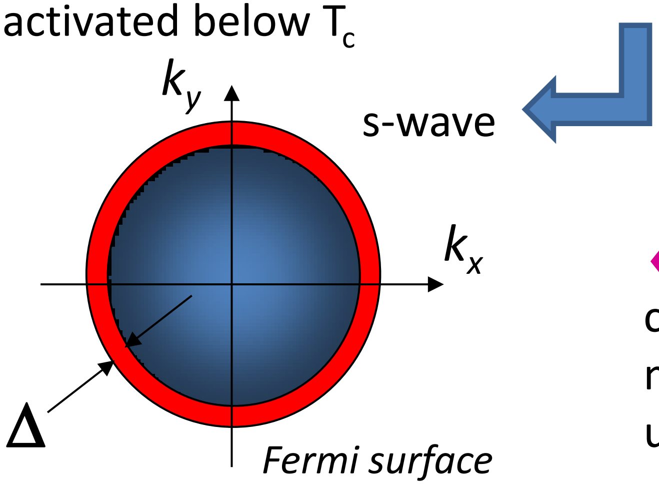
conventional vs unconventional superconductivity



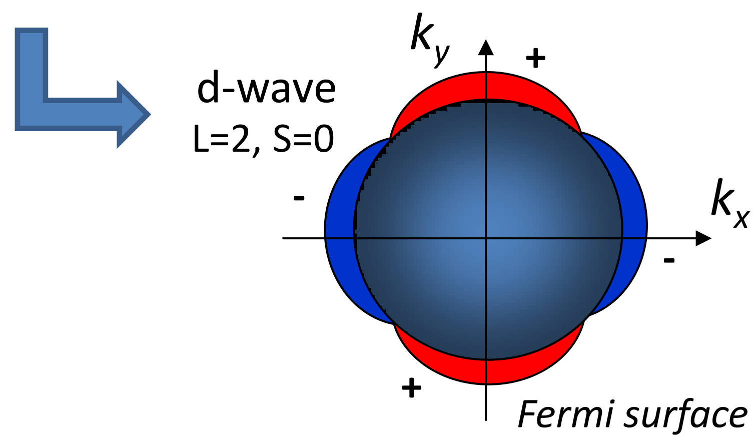
Cooper pair

◆ <u>conventional</u>: boson--lattice excitations (phonons); Cooper pairs with momentum **L** =0 and spin **S** =0**;** finite energy gap Δ that separates occupied and unoccupied electronic states $\Rightarrow$ any property that depends on these states is thermally activated below T_c

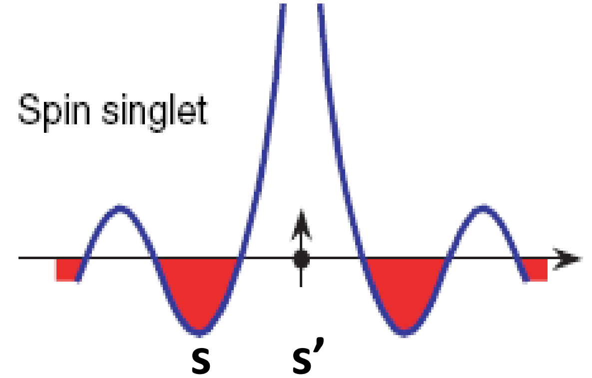
◆ <u>unconventional</u>: boson -- eg., magnetic fluctuations; Cooper pairs with **L** $\geq$ 0 and spin **S** $\geq$ 0; momentum-dependent gap Δ(**k**) that goes to zero at nodes on the Fermi surface $\Rightarrow$ any property that depends on the electronic state density has a power-law T-dependence below T_c



◆ BCS theory for conventional but no microscopic theory for unconventional



magnetically mediated unconventional superconductivity

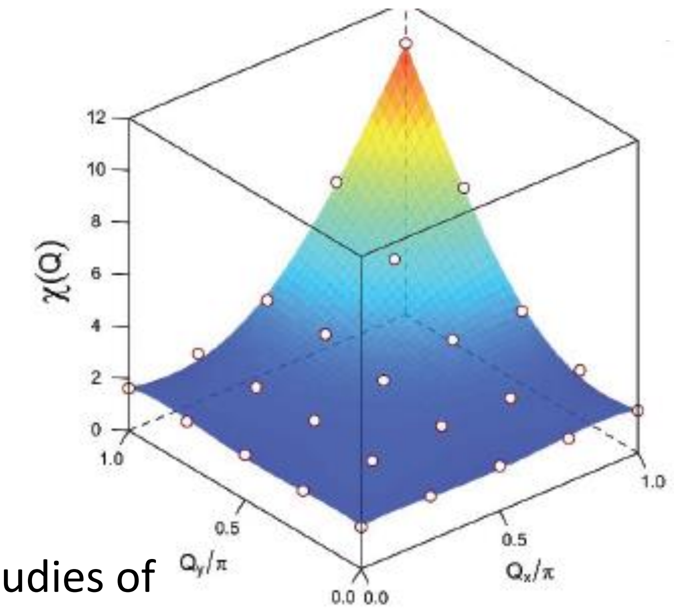


heuristic argument

♦ attractive Cooper pairing interaction V= -**s**•**s**'$g^2\chi$(**r**,t) repulsive at origin (**s'** site) but attractive at **r** > 0 ⇒ conducive to d-wave (L=2, S=0) pairing

P. Monthoux et al., Nature **450**, 1177 (2007)

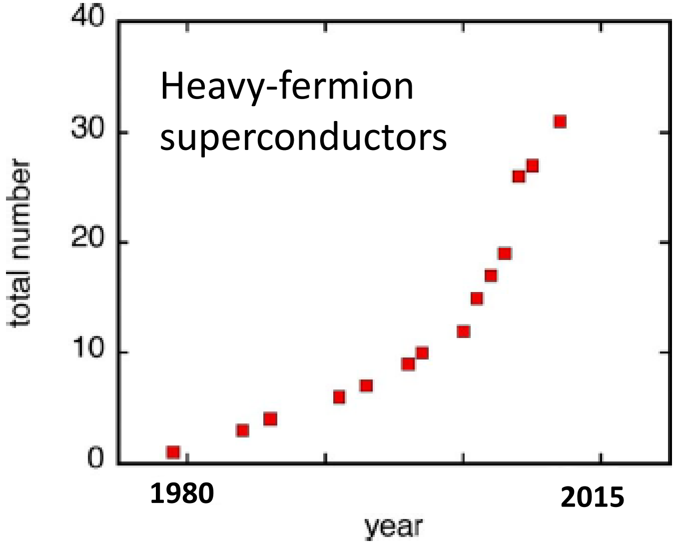
at an antiferromagnetic QCP:



♦ as $T_N \rightarrow 0$, magnetic excitations become quantum critical ⇒ magnetic susceptibility diverges at magnetic ordering wavevector **Q** ~ $1/|r_s - r_{s'}|$, favoring an enhanced attractive interaction and robust d-wave superconductivity

♦ generally consistent with what has ben learned from studies of heavy-electron, cuprate, iron-pnictide and organic superconductors, *but difficult to prove*

more heavy-fermion superconductors



after C. Pfleiderer, Rev. Mod. Phys. **81**, 1551 (2009)

1979-$CeCu_2Si_2$; 1983-UBe_{13}; *1984*-UPt_3; *1986*-URu_2Si_2; *1991*-UPd_2Al_3, UNi_2Al_3; *1993*-$CeCu_2Ge_2$; *1996*- $CePd_2Si_2$, $CeNi_2Si_2$, $CeRh_2Si_2$; *1997*-$CeIn_3$; *2000*-$CeRhIn_5$, UGe_2; *2001*-$CeCoIn_5$, $CeIrIn_5$, URhGe; *2002*-$PuCoGa_5$; *2003*-Ce_2RhIn_8, $PuRhGa_5$; *2004*-$CeNiGe_2$, $Ce_2Ni_3Ge_5$, UIr, $CePt_3Si$, $CeIrSi_3$, $CeRhSi_3$; *2007*-UCoGe, $NpPd_5Al_2$, $CeCoGe_3$, $CePd_5Al_2$; *2010*-Ce_2PdIn_8, $CePt_2In_7$ *2012*-$PuCoIn_5$, $PuRhIn_5$; *2014*-Ce_3PdIn_{11}

♦ superconductivity in proximity to or coexisting with magnetism – antiferromagnetic or ferromagnetic – a hallmark of each example

♦ all built from f-elements that exhibit a Kondo-impurity effect; all with large paramagnetic susceptibilities above T_c

♦ a recent review: B. D. White, J. D. Thompson and M. B. Maple, Physica C (in press)

lessons from uranium-based heavy-fermion superconductors

Outline:

-- UBe_{13} and Th-doping

-- UPt_3: two superconducting transitions

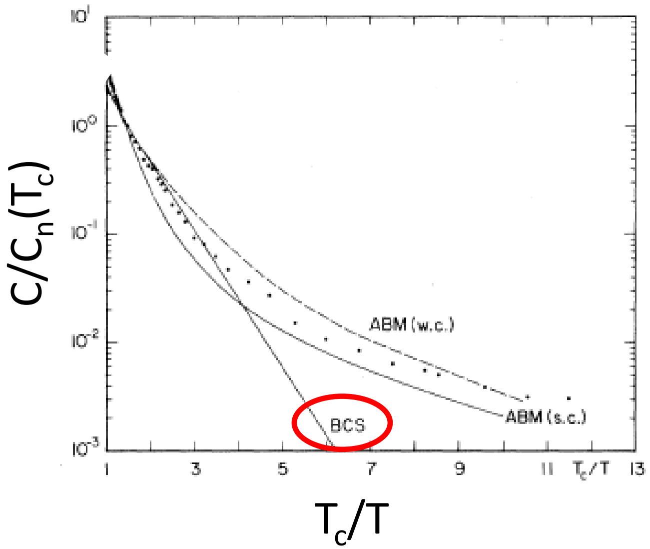
-- URu_2Si_2: coexisting hidden order

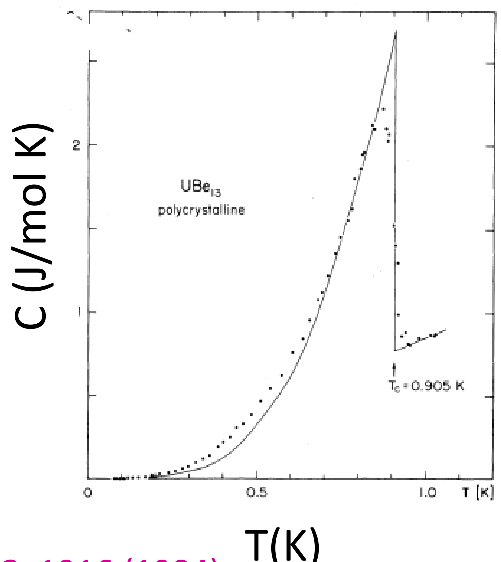
-- UPd_2Al_3: a spin gap and dual nature

-- UGe_2 and ferromagnetic relatives

-- summary

UBe_{13}



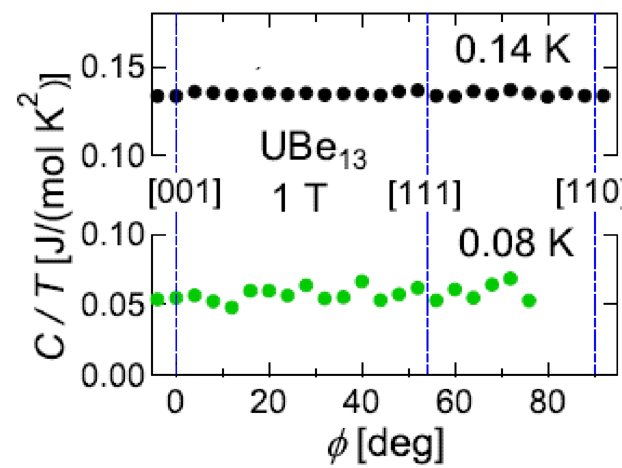


H. R. Ott et al., PRL **52**, 1916 (1984)

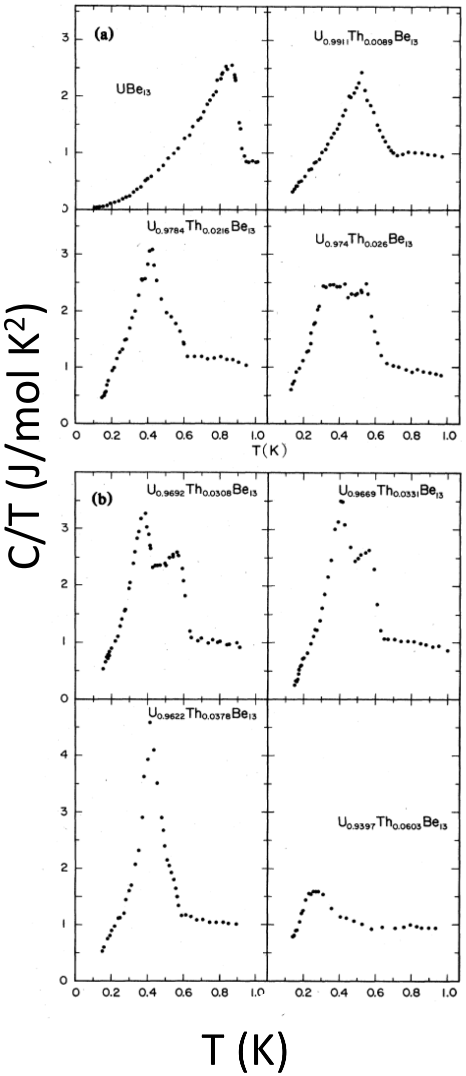
Hans R. Ott

Zachary Fisk

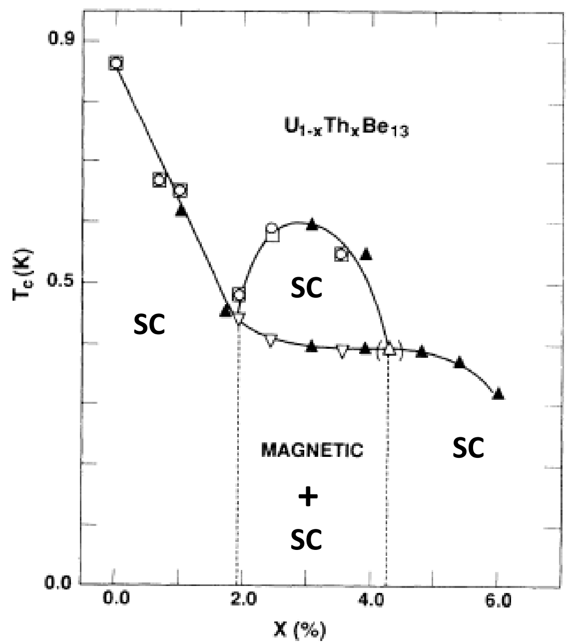
- solid curve—BCS; clearly inconsistent with experiment
- data more consistent with Anderson-Brinkman-Morel model of spin-fluctuation induced p-wave pairing in the superfluid state of ^{3}He $\Rightarrow$ point nodes in the gap function and $C \propto T^3$
- if p-wave, spin triplet, expect spin susceptibility (Knight shift) to be unchanged on cooling below T_c; Be-NMR shift consistent with expectation but μSR shift suggests a decrease below $T_c \Rightarrow$ inconclusive
- if nodal, expect a signature in field-angle dependent specific heat but not detected (Y. Shimizu et al., arXiv: 1411.1504) possibly because of band structure effects
- gap structure an open question though clear presence of spin fluctuations



Th-doped UBe_{13}

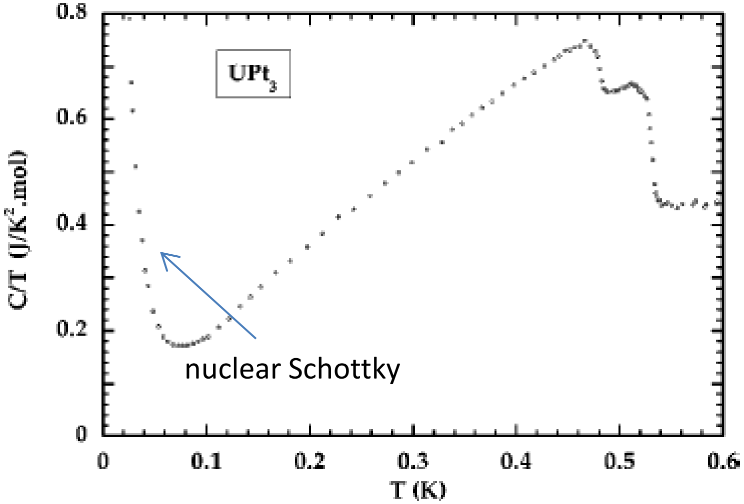


H. R. Ott et al., PRB **31**, 1651 (1985)



- non-monotonic variation in T_c and a coexisting second phase below T_c induced by slightly doping with nominally non-magnetic Th
- μSR (R. H. Heffner et al, PRL **65**, 2816 (1990)) $\Rightarrow$ second phase transition magnetic with moment 10^{-2} to 10^{-3} μ_B/U but hard to reconcile with huge specific heat anomaly $\Rightarrow$ possibility of a complex order parameter that has a magnetic component and breaks time-reversal-symmetry
- still an open question

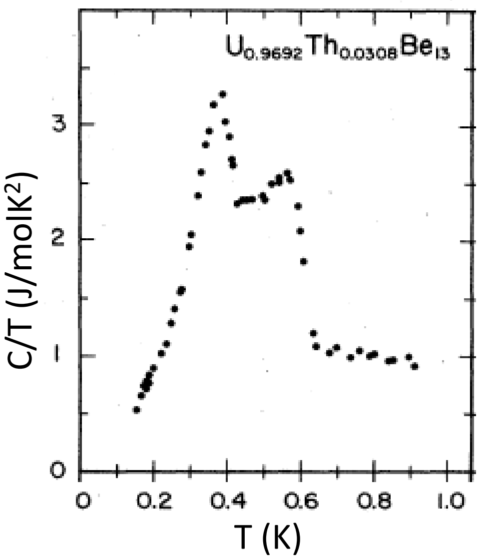
UPt_3



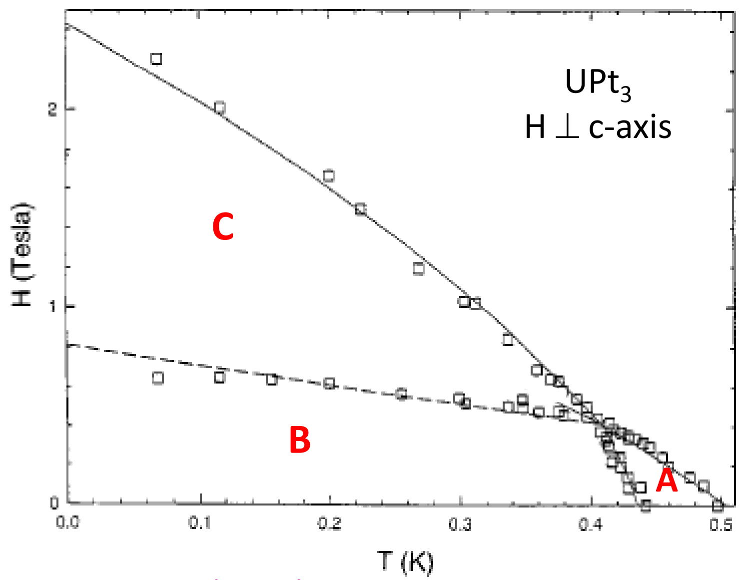
♦ two superconducting transitions in high quality single crystals (R. A. Fisher et al., PRL **62**, 1411 (1989)) ⇒ strong evidence for unconventional superconductivity and similar to Th-doped UBe_{13}

♦ below T_c, C/T approximately T-linear ⇒ gap nodes

♦ for T< 5-6 K, intrinsic (static or nearly static) commensurate antiferromagnetism with moment ~0.01-0.03μ_B/U, also similar to that in the coexisting phase of Th-doped UBe_{13}

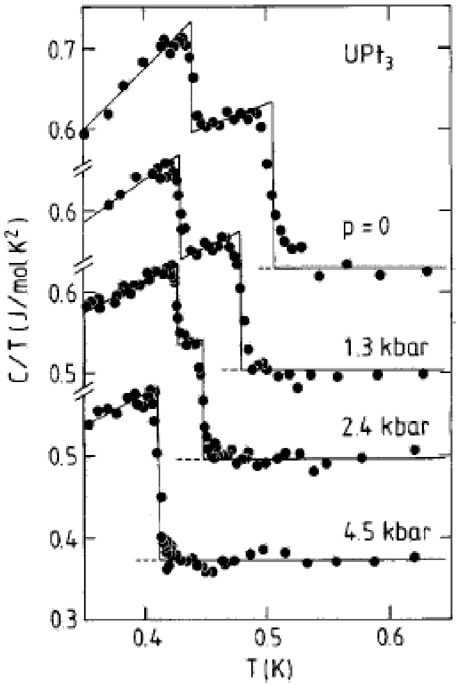


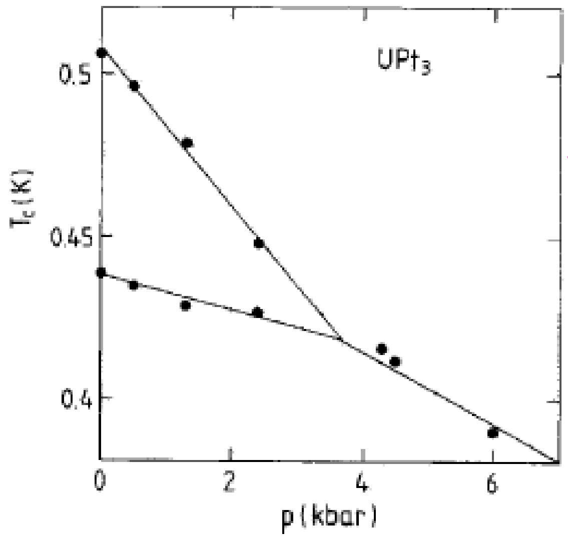
♦ in a magnetic field, three superconducting phases ⇒ further evidence for an unconventional superconducting gap structure



S. Adenwalla et al., PRL **65**, 2298 (1990)

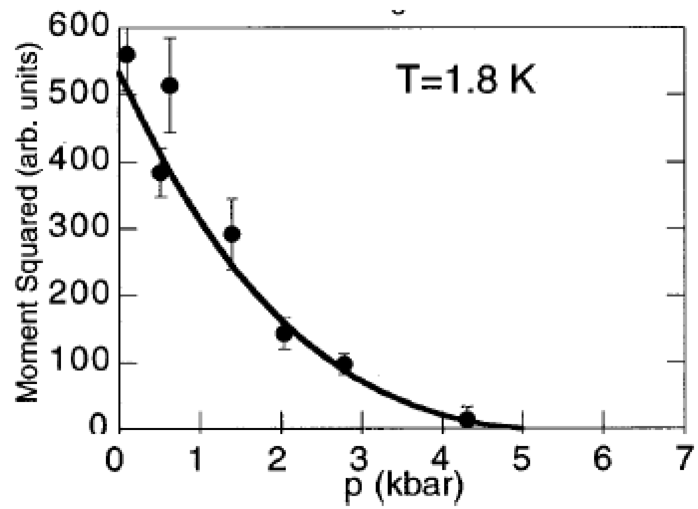
connecting superconductivity and magnetism





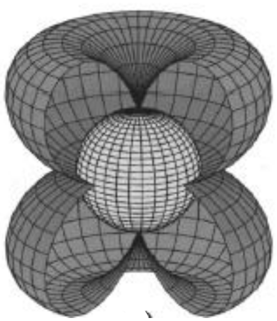
T. H. Trappmann et al., PRB **43**, 13714 (1991)

♦ pressure-induced collapse of two superconducting transitions near 4 kbar



♦ ordered moment $\rightarrow$ 0 at pressure where two superconducting transitions collapse (S. Hayden et al., PRB **46**, 8675 (1992))

point and line nodes



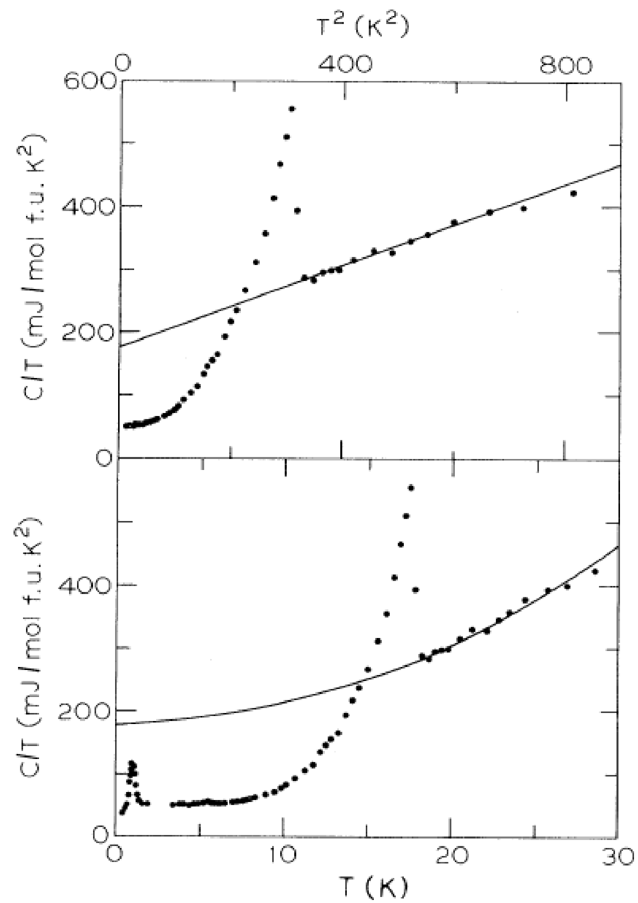
R. Joynt et al., RMP **74**, 235 (2002)

♦ two superconducting transitions, power laws in physical properties, H-T and T-P phase diagrams $\Rightarrow$ strong constraints on gap symmetry; probably a two-component order parameter with spin-triplet character; supported by TRS breaking seen in polar Kerr effect in phase B (E. R. Schemm et al., Science **345**, 190 (2014))

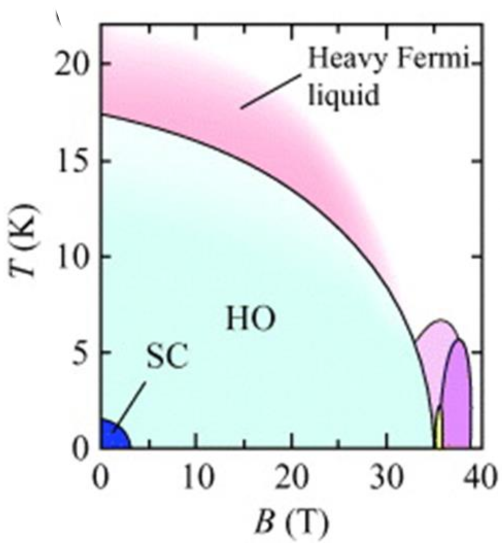
URu_2Si_2

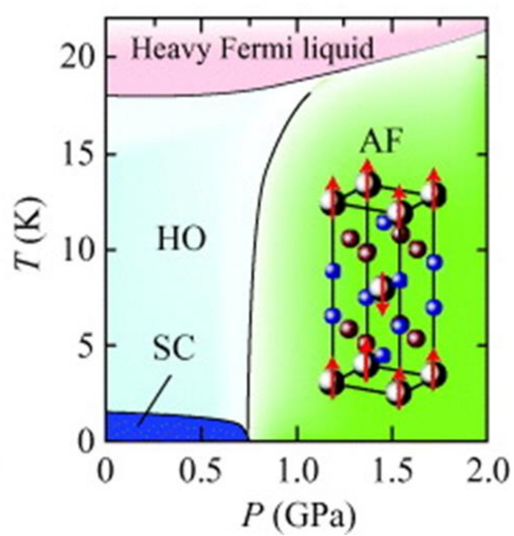
♦ huge second order specific heat anomaly at 17.5 K followed by superconducting transition near 1.5 K (T. Palstra et al. PRL **55**, 2727 (1985))

♦ transition at 17.5 K, thought initially to be local moment or spin density wave order but now known to be undetermined, hence 'hidden order' (J. A. Mydosh and P. Oppeneer, RMP **83**, 1301 (2011))



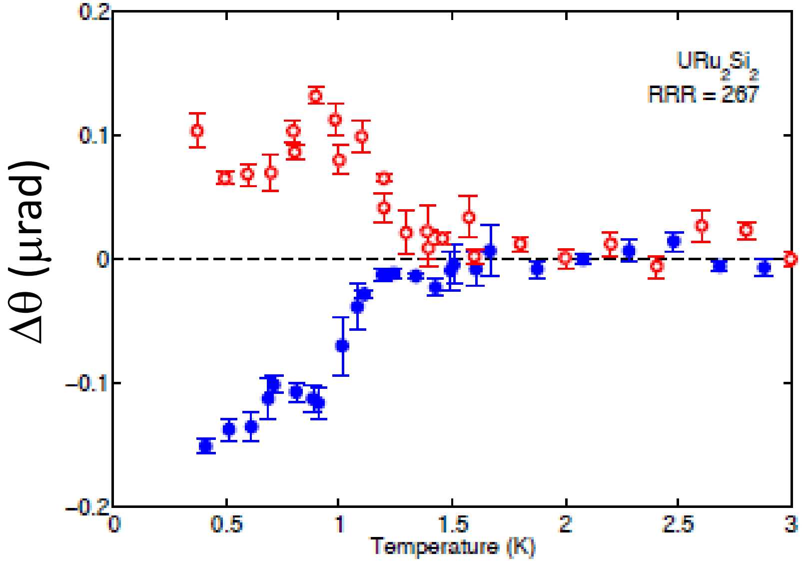
♦ by far, most attention given to trying to identify nature of the hidden order – at least 14 different theoretical models proposed, none fully consistent with experiment



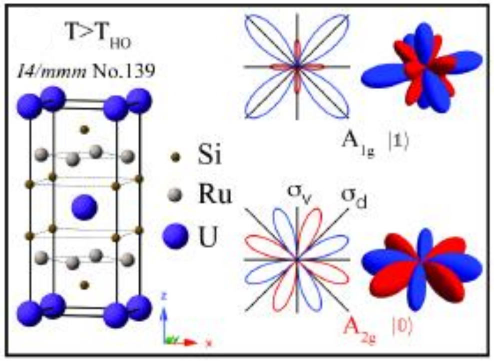


♦ superconductivity completely enclosed by hidden order vs field and suppressed when hidden order changes to large moment antiferromagnetism vs pressure (eg, Y. Kasahara et al., New J. Phys. **11**, 055061 (2009))

symmetry breaking

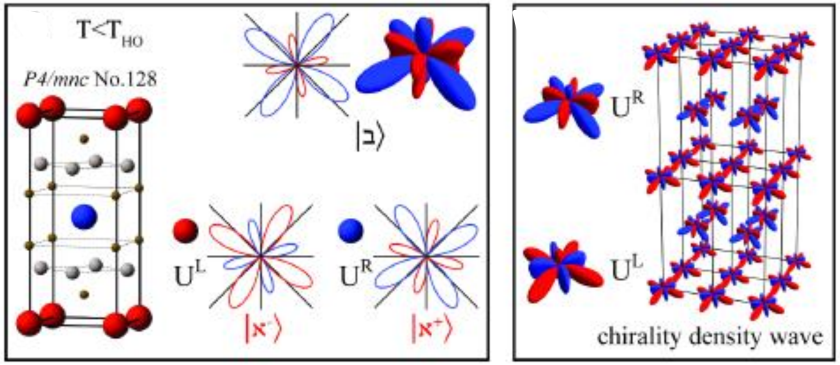


♦ dependence of the sign of polar Kerr effect on cooling below T_c in +100 and -100 Oe fields $\Rightarrow$ time reversal symmetry breaking (E. R. Schemm et al. arXiv: 1410:1479)) similar to UPt_3 $\Rightarrow$ complex order parameter with possible chiral symmetry



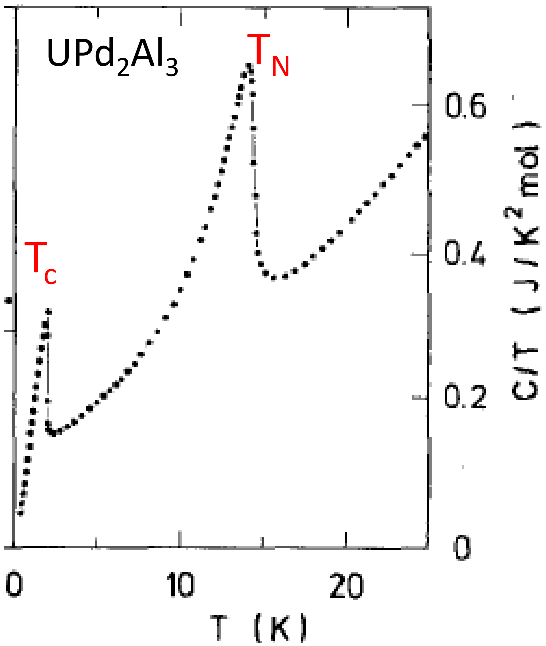
♦ local vertical and diagonal reflection symmetry breaking and chirality of U orbitals in hidden order from polarized Raman scattering (H.-H. Kung et al., Science, Feb 12, (2015))

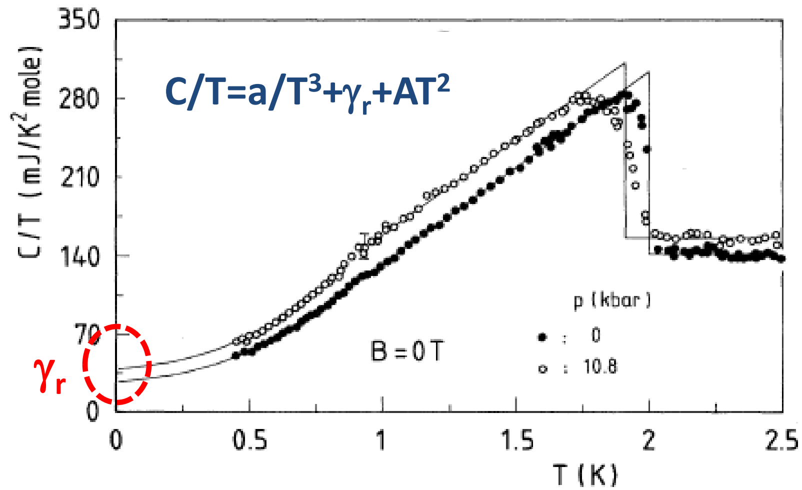
♦ with superconductivity only when HO is present, possible that SC order parameter symmetry inherited from HO



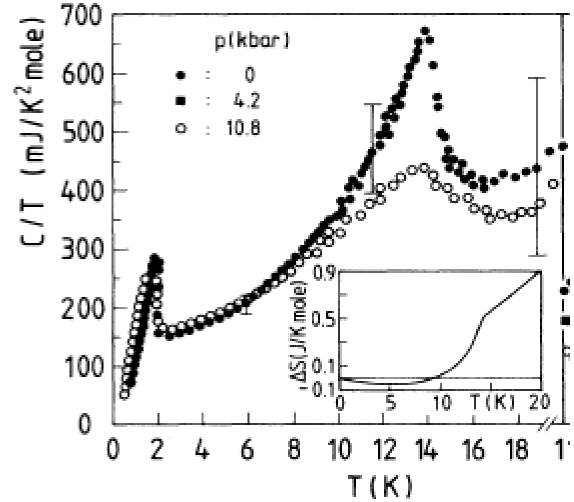
UPd_2Al_3: superconductivity + antiferromagnetism

C. Geibel et al., Z. Phys. B **84**, 1 (1991)





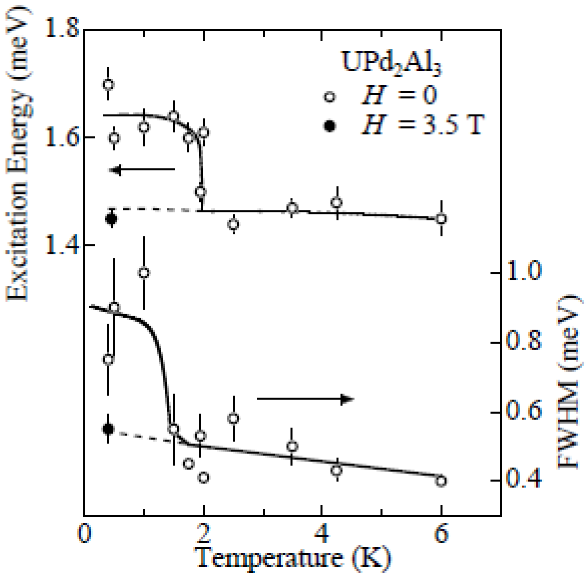
R. Caspary et al., PRL **71**, 2146 (1993)



♦ commensurate (0,0,1/2) large moment ($0.8\mu_B$) AFM coexisting with nodal SC (A. Krimmel et al., Z. Phys. B **86**, 161 (1992))

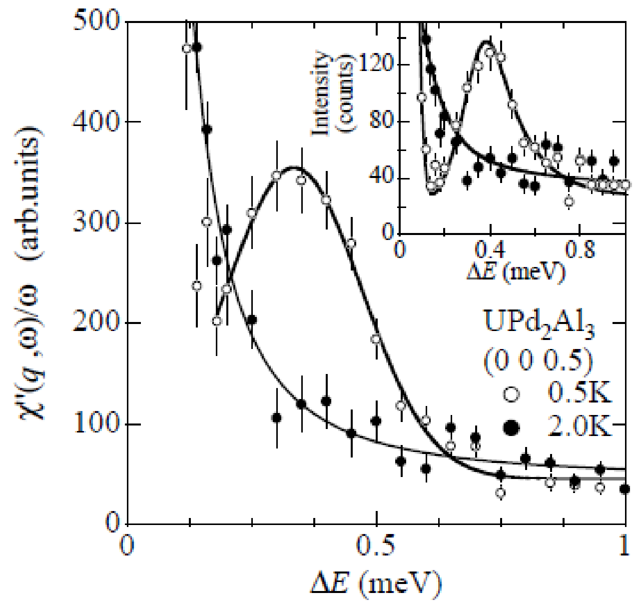
♦ analysis of pressure-dependent specific heat $\Rightarrow$ two component f-subsystems reflected in enhanced γ_r and reduced entropy due to AFM; an f-itinerant component responsible for superconductivity and a f-localized component giving AFM (so-called dual nature)

magnetism coupled to superconductivity



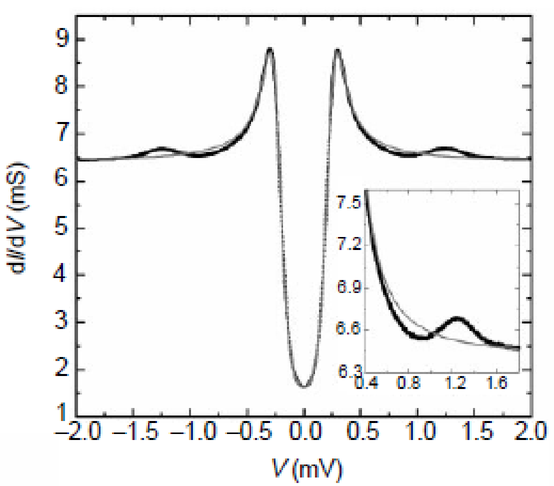
♦ shift of spin wave excitation near 1.5 meV to higher energy and broadening below T_c (and their recovery to normal state values for $H>H_{c2}$) $\Rightarrow$ strong coupling of nodal superconductivity to spin excitations

N. Metoki et al., PRL **80** 5417 (1998)



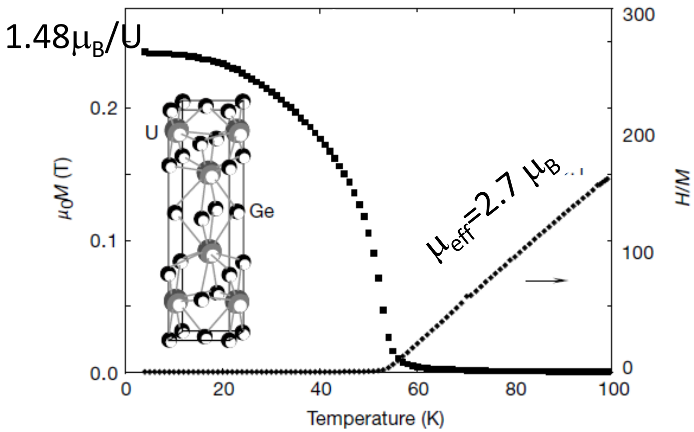
♦ quasi-elastic scattering above T_c to inelastic scattering below T_c $\Rightarrow$ a SC-induced spin gap ≈0.4meV

♦ together, strong evidence for magnetically mediated superconductivity



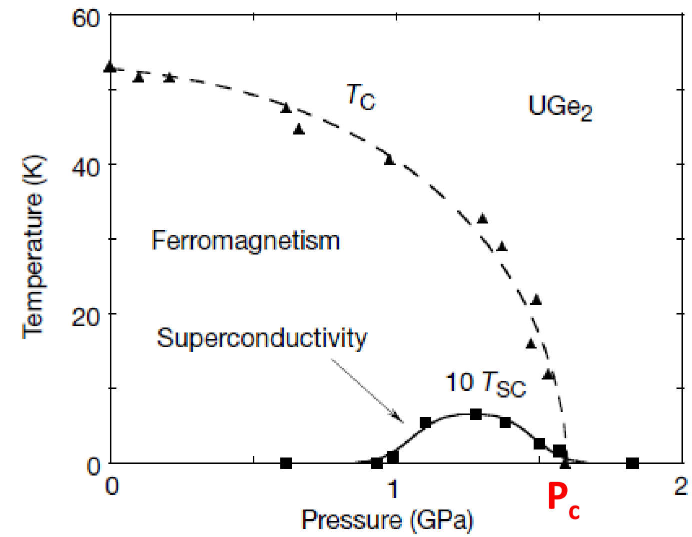
♦ conclusion supported by tunneling spectroscopy (M. Jourdan et al., Nature **398**, 47 (1999)) showing that conductance can be fit assuming spin excitations act as the equivalent of phonons in BCS superconductors; until recently, the most compelling evidence for magnetically mediated SC

UGe_2



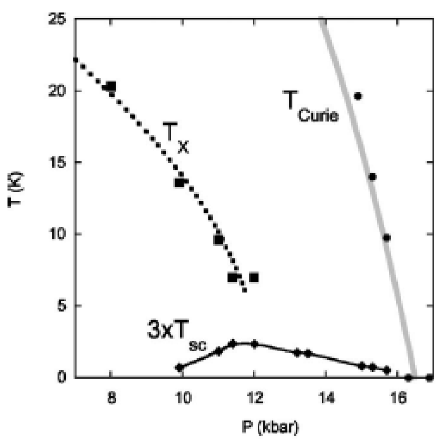
♦ itinerant ferromagnetism below 53K that is suppressed toward T=0 near P_c=1.6 GPa , but first order magnetic transition above ~1.2 GPa where ordered moment is reduced to ~$1\mu_B$/U (S.S. Saxena et al., Nature **406**, 587 (2000); A. Huxley et al., PRB **63**, 144519 (2001))

♦ superconductivity only inside the ferromagnetic phase



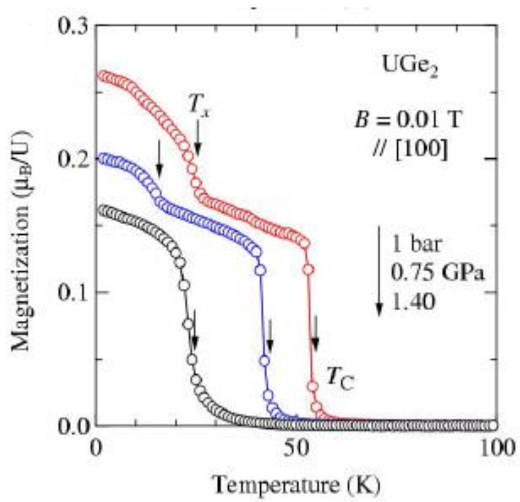
♦ if ferromagnetism and superconductivity from the same electrons, spin aligned (ferromagnetic) quasiparticles ⇒ Cooper pairs with odd-parity orbitals ⇒ spin-triplet, p-wave

♦ maximum T_c =0.8K at $P<P_c$, in contrast to other examples of P-induced superconductivity



♦ instead, max T_c where boundary T_x(P) of some other phase extrapolates to T=0 (A. Huxley et al., PRB **63**, 144519 (2001)) and Sommerfeld coefficient also a maximum ≈ 110mJ/molK^2 (N. Tateiwa et al., JPCM **13**, L17 (2001))

UGe_2 perspective

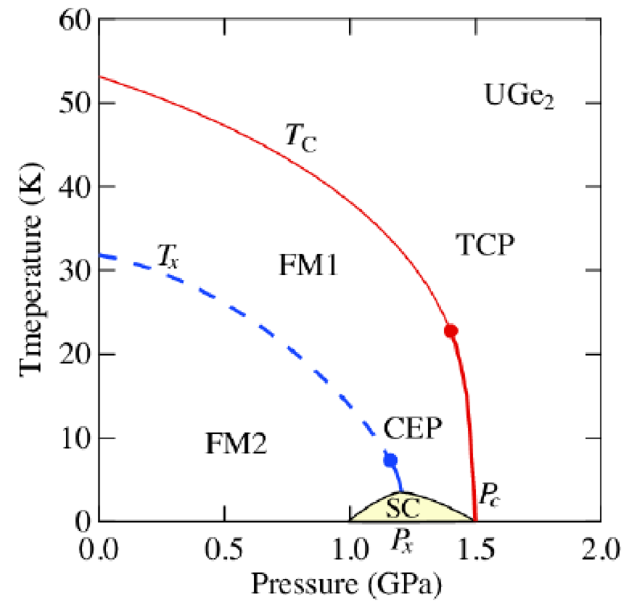


♦ for P<1 GPa, $T_x(P)$ a crossover from one ferromagnetic state (FM1) to another (FM2)

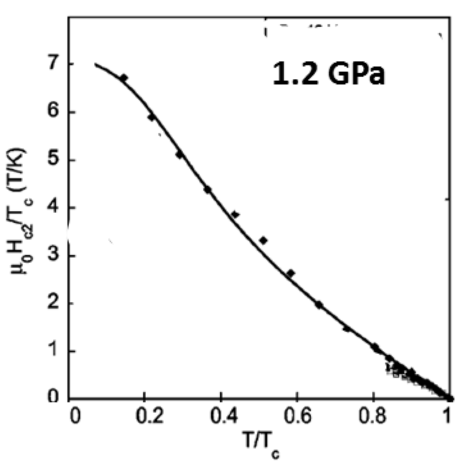
♦ at low T and P≈ 1.2 GPa, first order transition at $T_x(P)$, terminating at a finite temperature critical end point

♦ $T_C(P)$ second order up to ≈ 1.4 GPa and first order up to P_c ⇒ presence of a tricritical point

N. Tateiwa et al., J. Korean Phys. Soc. **63**, 627 (2013)



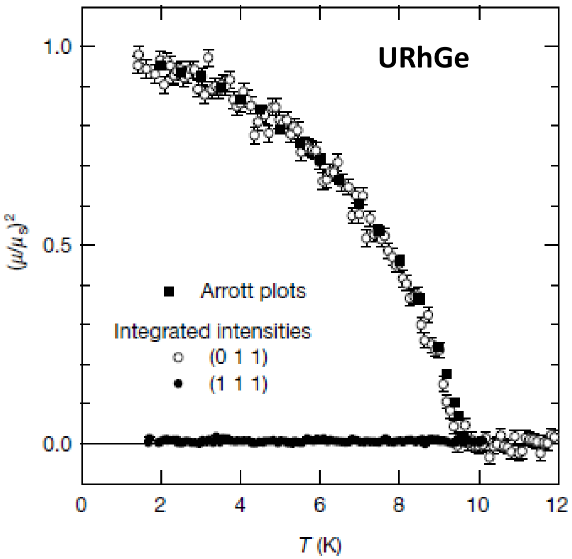
♦ with $T_x(P)$ and $T_C(P)$ first order in the limit T→ 0, absence of a quantum critical phase transition ⇒ origin of 'pairing glue'? SC driven by changes in the Fermi surface topology, giving a large density of states in majority spin surface in FM2 (K. G. Sandeman et al. PRL **90** 167005 (2003))



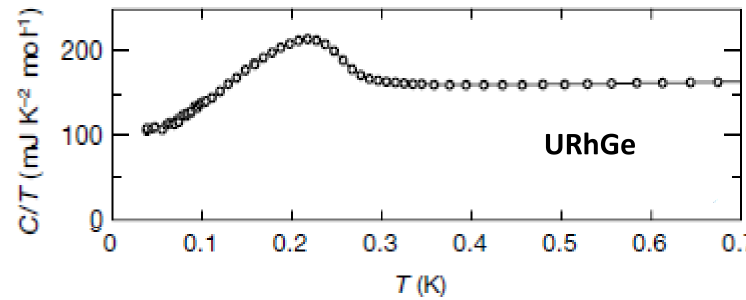
I. Seikin et al., PRB **64**, 22503 (2001)

♦ very large upper critical field exceeding orbital and Pauli limits ⇒ consistent with spin triplet superconductivity but gap symmetry and origin of and positive curvature in $H_{c2}(T)$ still open questions

two relatives



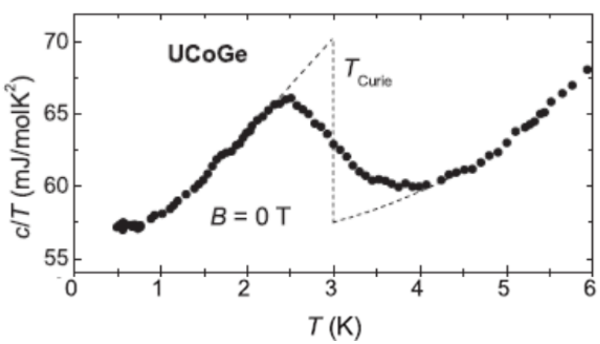
URhGe

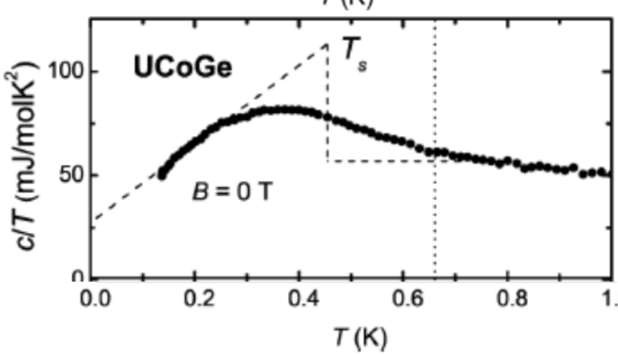


- uniaxial, itinerant ferromagnetism below 9.8 K with $0.42\mu_B$/U ordered moment

D. Aoki et al. Nature **413** , 613 (2001)

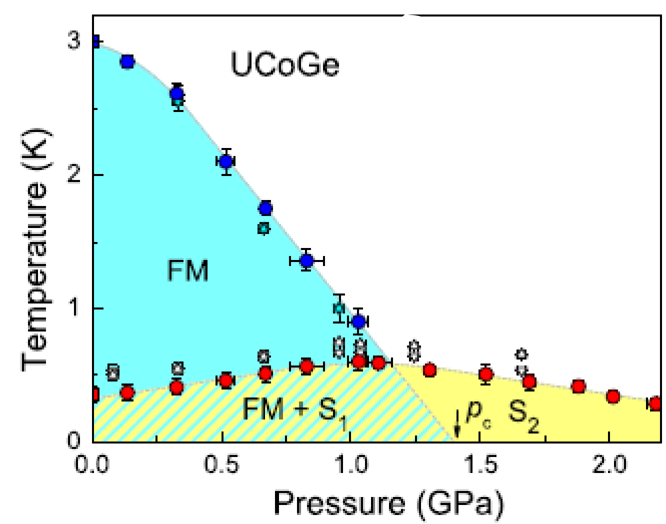
- coexisting with bulk (spin-triplet) superconductivity below T_c=0.25K; structurally and magnetically similar to UGe_2 under pressure





UCoGe

E. Slooten et al., PRL **103**, 097003 (2009)



- very weak ferromagnetism ($0.03\mu_B$/U) below 3K coexisting with (likely) spin-triplet superconductivity near 0.4K (N. T. Huy et al., PRL **99**, 067006 (2007))

- a dome of superconductivity possible related to a ferromagnetic quantum critical point

summary

- frequently, evidence for a complex superconducting order parameter (UPt_3 a prototype) and with a spin-triplet component – UPt_3, Th-doped UBe_{13}, URu_2Si_2, UGe_2, URhGe and UCoGe (and UNi_2Al_3, UIr, U_2PtC_2 not discussed)

- UPd_2Al_3 a notable counter example with large moment antiferromagnetism coexisting with magnetically mediated, spin-singlet superconductivity; concept of a ‘dual’ nature of U 5f electrons

- mechanism of superconductivity in URu_2Si_2 apparently tied to mechanism of hidden order

- still many open questions – origin of Th-doped magnetism in UBe_{13} and of hidden order phase in URu_2Si_2, why U-based heavy fermion superconductors often have complex order parameters, …

$CeIn_3$ and derivatives

Outline:

-- $CeIn_3$ as a building block

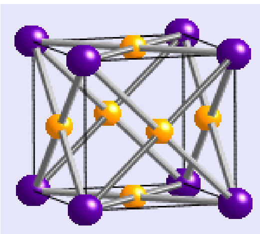
-- $CeIn_3$ superconductivity

-- magnetism and superconductivity in Ce115s and related

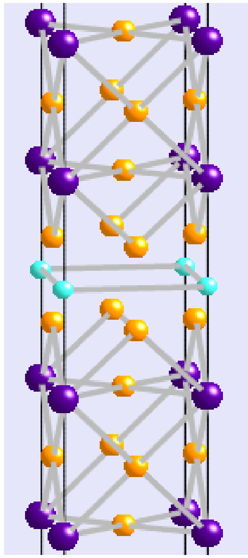
-- spin-offs from the Ce115s

-- summary

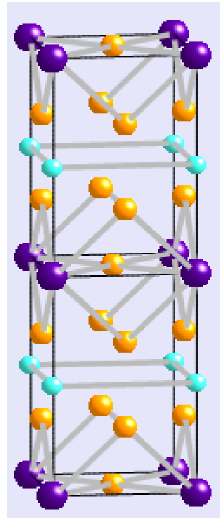
$CeIn_3$ and structural derivatives



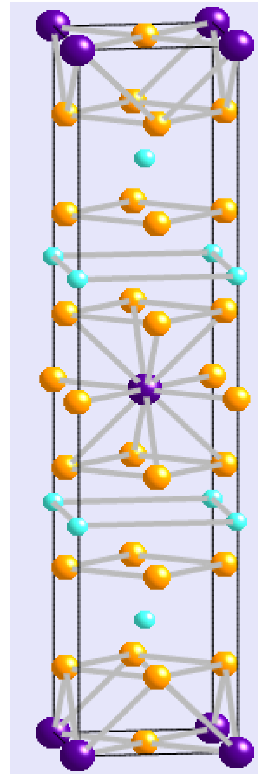
∞ layer
$CeIn_3$, T_c= 0.2K



2-1-8
(2)$CeIn_3$+(1)MIn_2
$T_c \approx$ 1K



1-1-5
(1)$CeIn_3$+(1)MIn_2
$T_c \approx$ 2.3K



2-1-7
(1)$CeIn_3$+(2)MIn_2
$T_c \approx$ 2.2K

- several layered variants of $Ce_mM_nIn_{3m+2n}$ with ground states highly tunable by pressure and M elements: Co, Rh, Ir, Pd, Pt
- basic building block of *m*-adjacent units of $CeIn_3$ separated along the tetragonal c-axis by *n*-units of MIn_2
- m=1,2 n=1 compounds discovered by Y. Grin et al. in 1982, 2-1-7 by Zh. M. Kurenbaeva in 2008 and most recently 3-1-11 and 5-2-19 by A. Tursina et al. in 2013

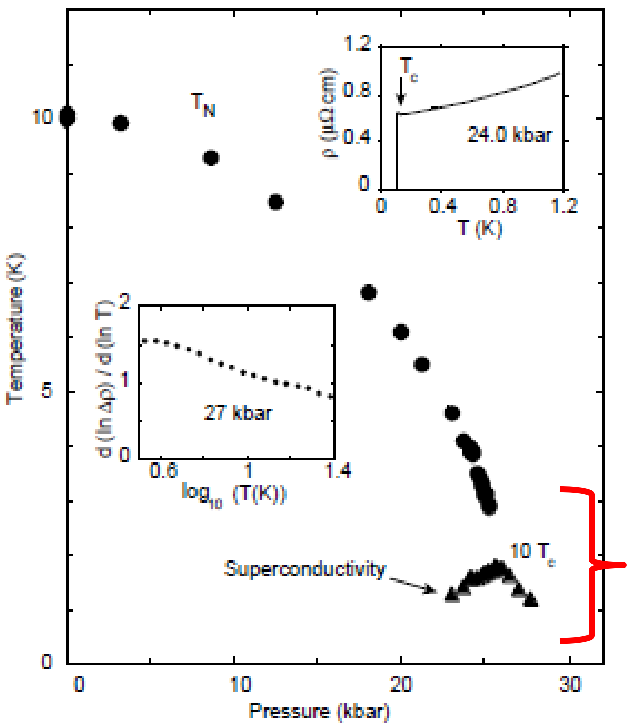
- besides $CeIn_3$, Ce115 and Ce218 most studied, with very little work on higher order derivatives that also are more difficult to prepare
- of these, 7 known to be Ce-based heavy-fermion superconductors plus 4 more based on Pu – nearly 1/3 of all heavy-fermion superconductors

magnetism and superconductivity in $CeIn_3$

♦ commensurate (1/2,1/2,1/2) magnetic structure with ordered moment ~$0.6\mu_B$ below T_N=10.2 K (J. M. Lawrence et al., PRB **22**, 4379 (1980))

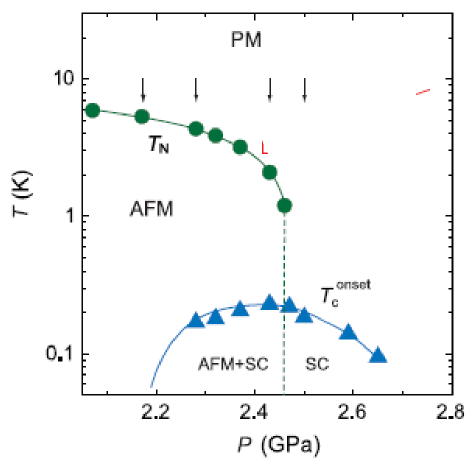
♦ with applied pressure, $T_N \rightarrow 0$ near 2.6 GPa, where superconductivity with max. $T_c \approx 0.25$ K emerges

N. D. Mathur et al., Nature **394**, 39 (1998)



♦ above dome of SC, $\rho \propto T^{1.6}$, consistent with expectations ($T^{1.5}$) of a conventional Hertz-Millis type of SDW quantum criticality ⇒ suggests unconventional superconductivity mediated by antiferromagnetic quantum fluctuations

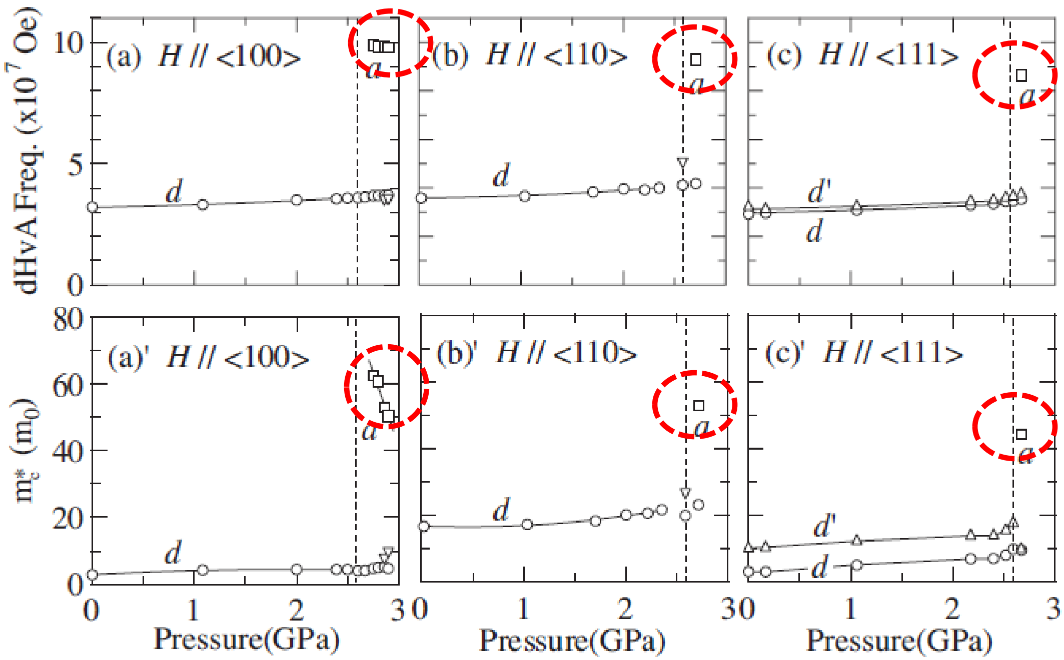
♦ internal field produced by magnetic order decreases toward 0 near 2.4 GPa but magnetic transition becomes first order (Y. Kohori et al., Physica B **281&282**, 12 (2000))



S. Kawasaki et al., arXiv:0802.2150

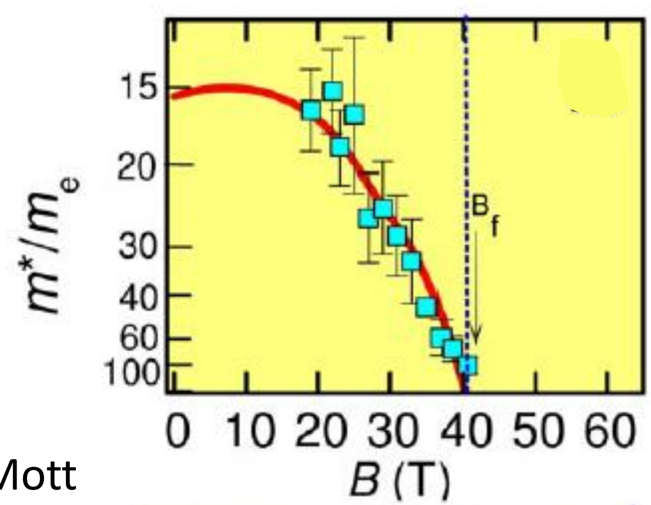
♦ superconductivity mediated by antiferromagnetic fluctuations arising from a quantum-critical end point near 2.46 GPa?

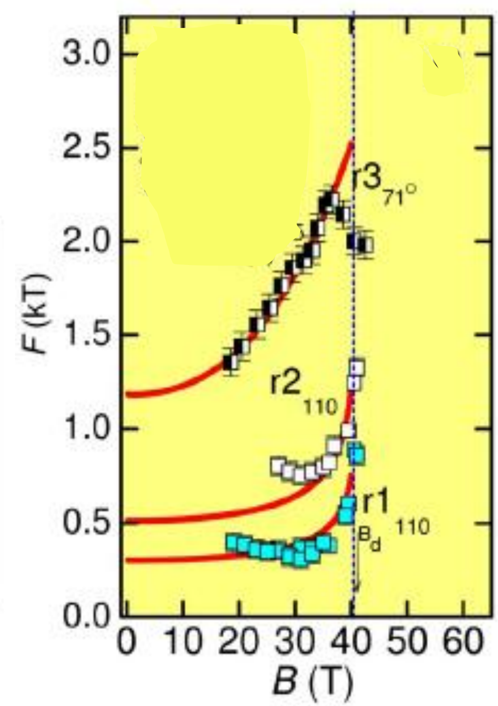
$CeIn_3$ Fermi surface and m* versus P,H



♦ near/just above P_c, deHaas-vanAlphen frequencies increase and m* increases from ~$20m_0$ to ~ $60m_0$ ⇒ small (f-localized) to large (f-delocalized) Fermi surface accompanied a sharp increase in m*, the former not expected at a SDW QCP (R. Settai et al., JPSJ **74**, 3016 (2005))

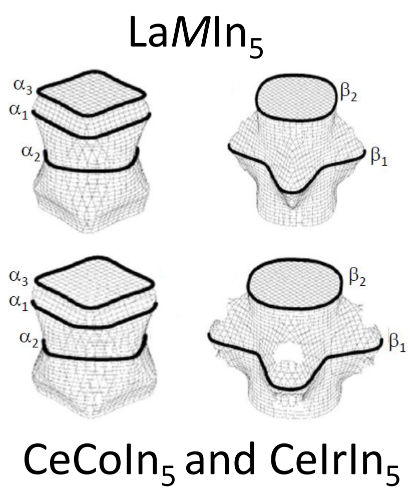
♦ as a function of field, m* diverges and dHvA frequencies grow upon approaching a critical field near 40T (S. Sebastian et al., PNAS **106**, 7741 (2009)) ⇒ a field-tuned QCP possibly related to that under pressure



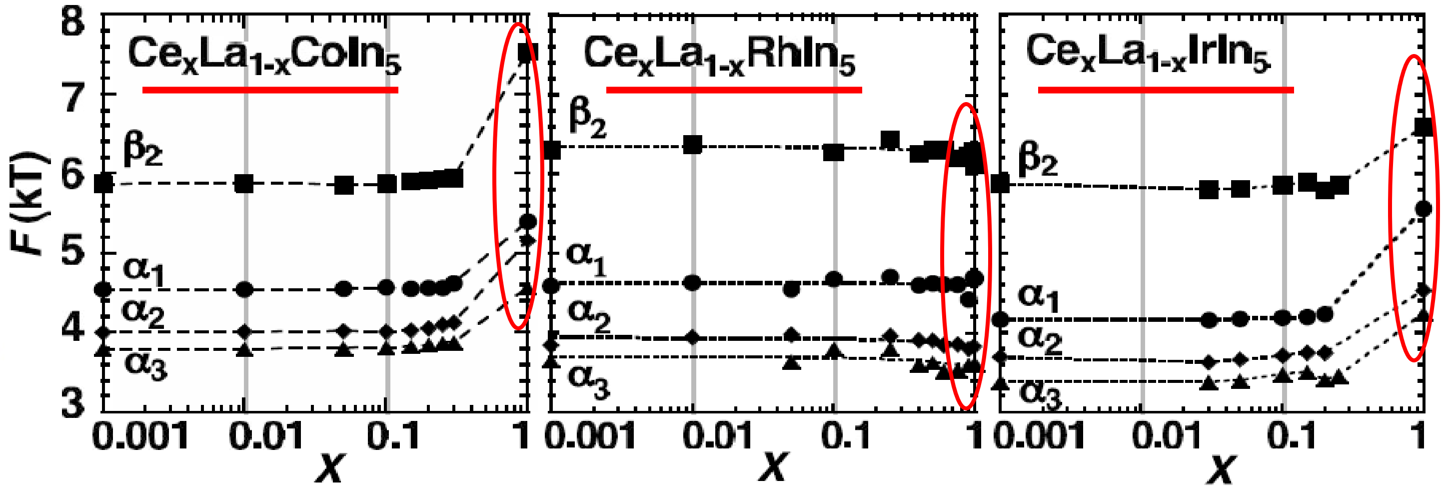


♦ Kondo-breakdown or selective Mott types of quantum criticality also produces ‘pairing glue’?

Fermi surface and properties of Ce115's



H. Shishido et al., JPSJ **71**, 162 (2002)



N. Harrison et al. PRL **93,** 186405 (2004)

♦ for T>100 K, Ce 4f electron localized, but in the limit T$\rightarrow$ 0

♦ $CeCoIn_5$: 4f contributes to Fermi surface; T_c=2.3 K; $\gamma \approx 250$ mJ/molK^2

♦ $CeRhIn_5$: $LaRhIn_5$-like $\Rightarrow$ 4f localized; T_N=3.8 K; $\gamma \approx 400$ mJ/molK^2

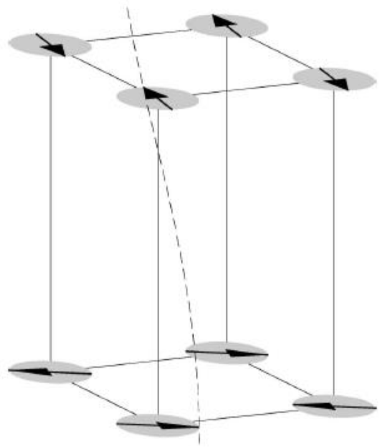
♦ $CeIrIn_5$: 4f contributes to Fermi surface; T_c=0.4 K; $\gamma \approx 720$ mJ/molK^2

♦ electronic structure of each dominated by warped cylindrical sheet $\Rightarrow$ quasi-2D, which favors higher T_c when SC is mediated by antiferromagnetic fluctuations (P. Monthoux & G.G. Lonzarich, PRB **66**, 224504 (2002))

♦ each a nodal (d-wave) heavy-fermion superconductor either at ambient or applied pressure with T_c's 4-10 times higher than in $CeIn_3$

♦ like $CeCu_2Si_2$, degeneracy temperature of heavy quasiparticles $T_F \approx (R\ln 2)/\gamma \approx 10$ K and $T_c/T_F \approx 0.04 - 0.2 \Rightarrow$ 'high'-T_c

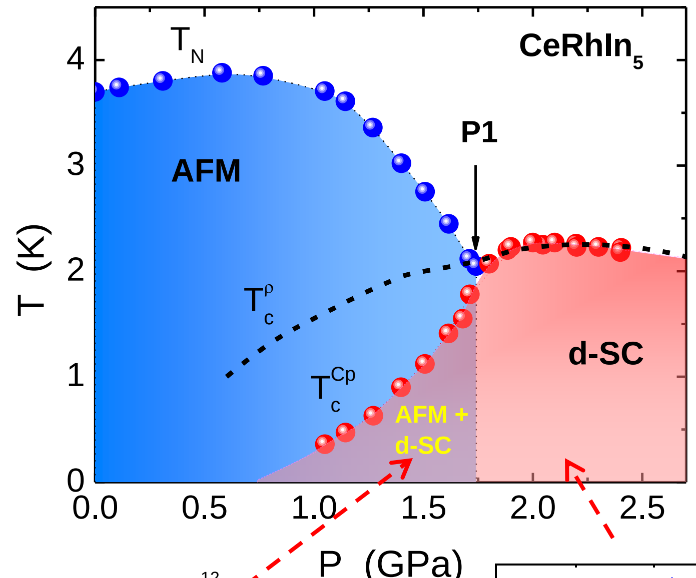
$CeRhIn_5$ magnetism and P–induced superconductivity



W. Bao et al., PRB **62**, 14621 (2000)

♦ incommensurate (1/2, 1/2, 0.297) magnetic structure with ordered moment ≈ $0.6\mu_B$ below T_N=3.8 K; moment similar to that in $CeIn_3$

♦ pressure-induced superconductivity (H. Hegger et al., PRL **84**, 4986 (2000)) coexisting with antiferromagnetic order below P1=1.7 GPa, above which evidence for magnetic order is absent (T. Mito et al., PRL **91**, 137001 (2003))

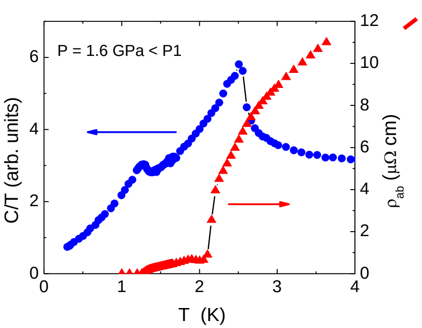


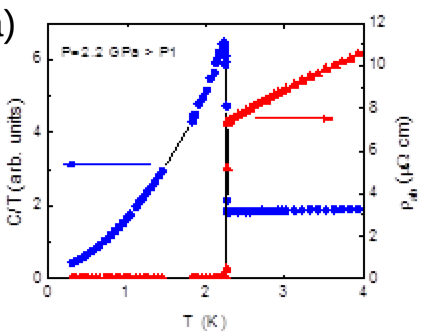
♦ in region of coexistence, anisotropic resistive T_c > bulk T_c ⇒ filamentary, 'textured' superconductivity

♦ above P1, resistive and bulk T_c's coincide

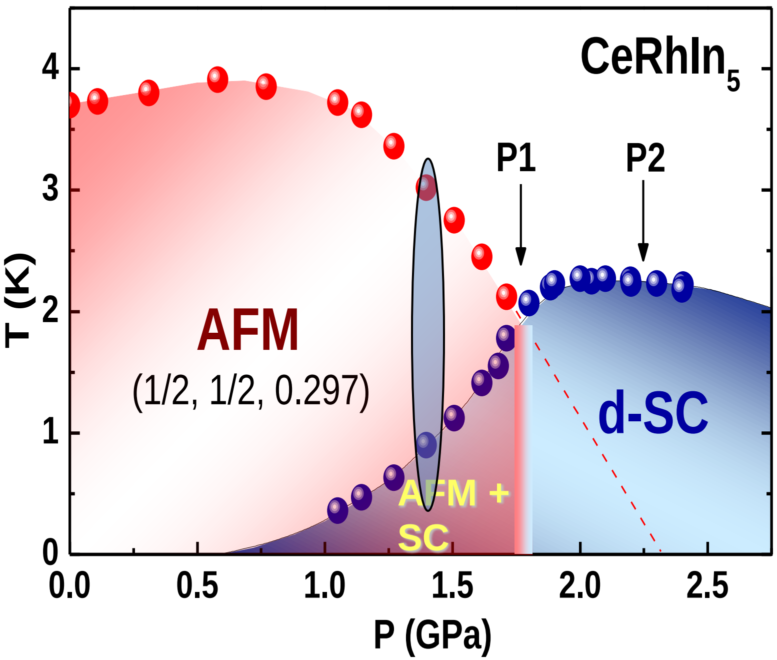
♦ similar behavior common in other heavy-fermion, cuprate and iron-pnictide materials when unconventional superconductivity coexists with some other order (T. Park et al., PRL **108**, 077003 (2012))

♦ a useful lesson

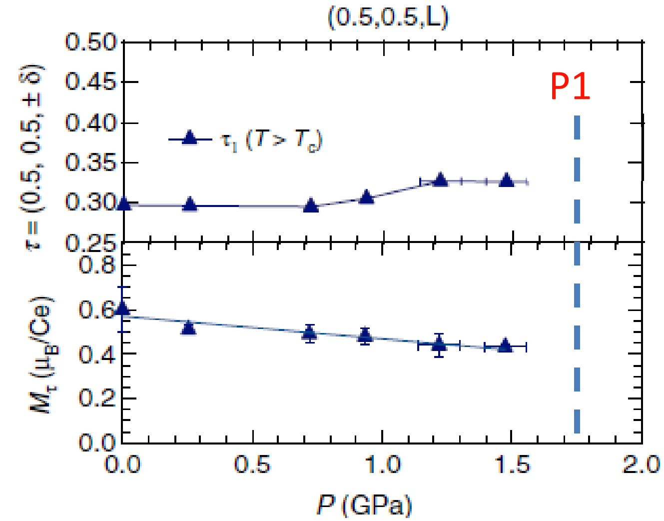




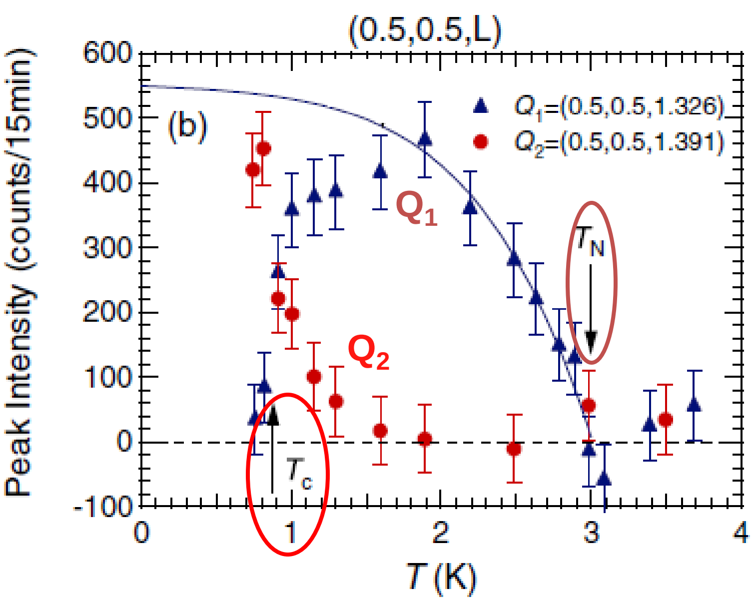
coupling of magnetism and superconductivity in $CeRhIn_5$



♦ for P < P1, slight increase in δ and ~ 25% decrease in μ_{ord}

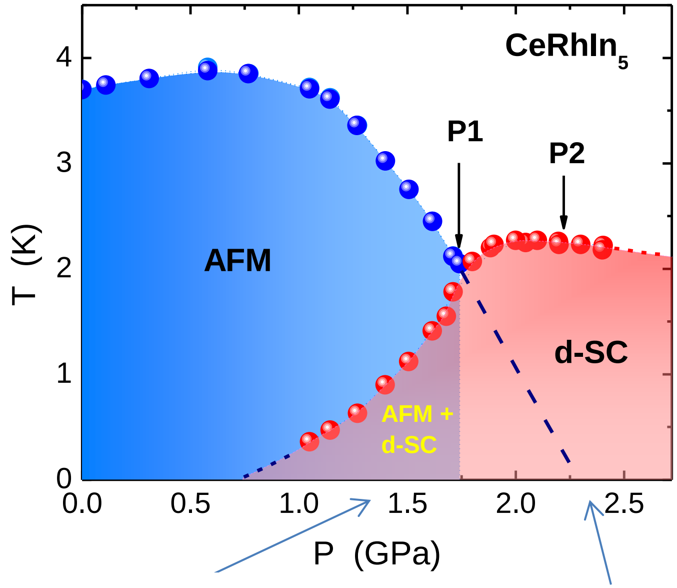


N. Aso et al., JPSJ **78**, 073703 (2009)

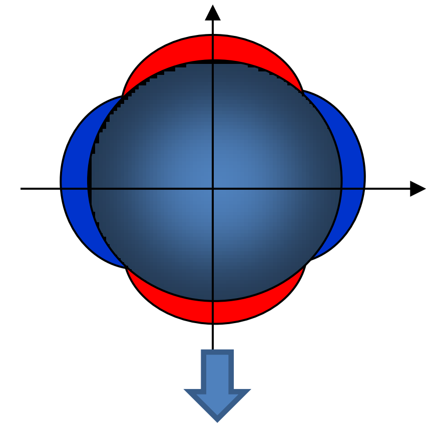


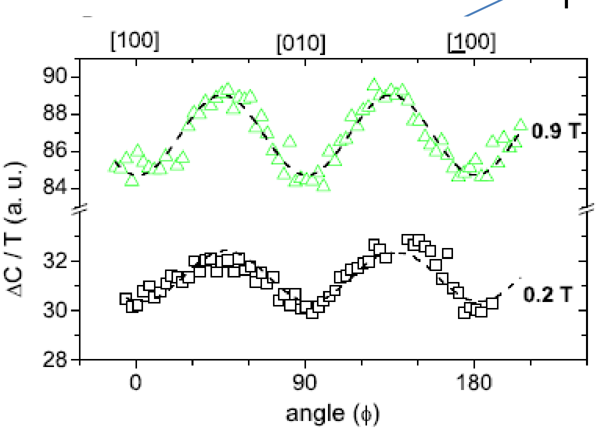
♦ in coexistence region, neutron intensity at $\mathbf{Q_1}$ increases below T_N, but decreases at lower T where a new $\mathbf{Q_2}$ (½, ½, 0.391) emerges whose intensity increases at T_c while $\mathbf{Q_1}$ intensity$\rightarrow 0 \Rightarrow$ *coupling of AFM and SC order parameters*

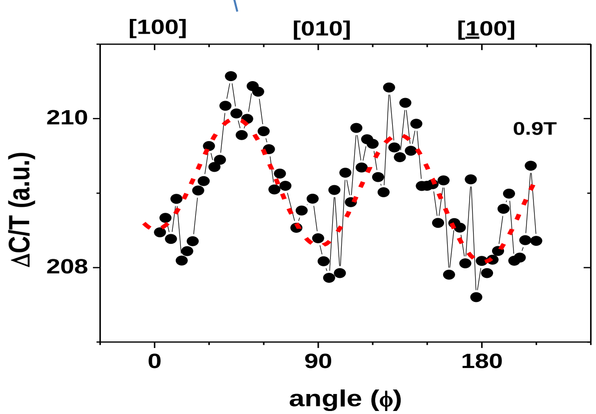
CeRhIn$_5$: P–induced nodal superconductivity



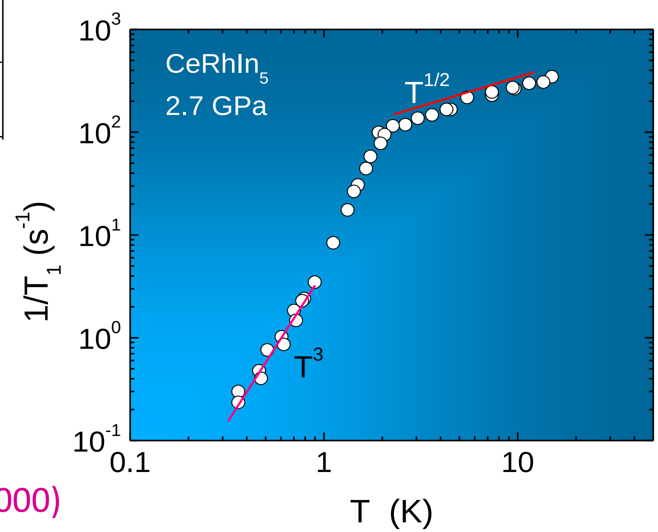
- 4-fold modulation of specific heat and T^3 power law in $1/T_1$
- strong evidence for d-wave superconductivity







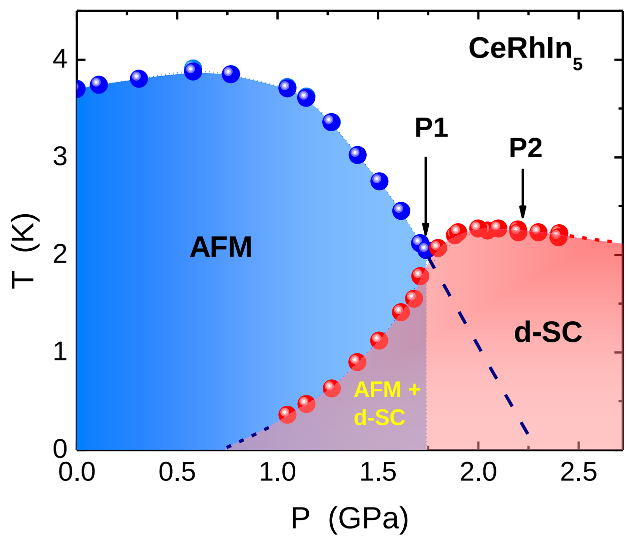
T. Park et al., PRL **101**, 177002 (2008)



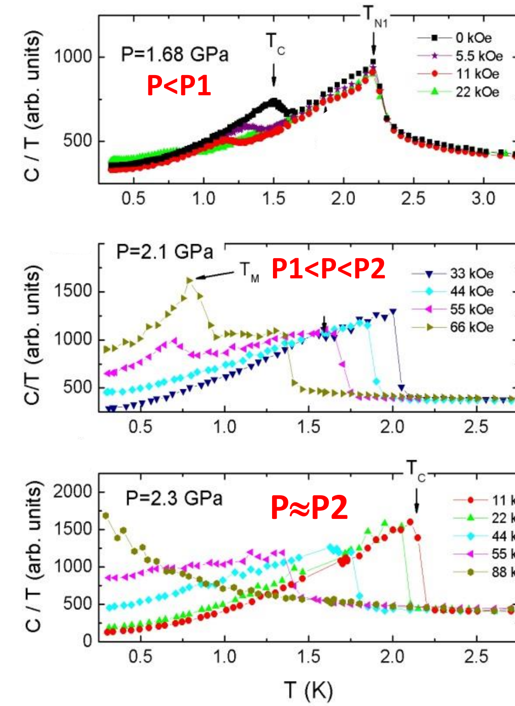
- $1/T_1 \propto T^{1/2}$ above T_c not characteristic of a normal metals but expected from strong antiferromagnetic fluctuations

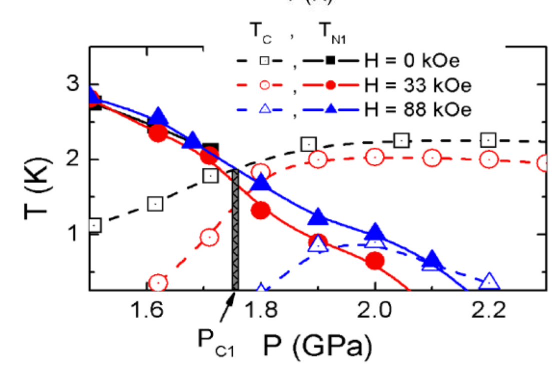
Y. Kohori et al., Eur. Phys. J. **18**, 601 (2000)

$CeRhIn_5$: field-induced magnetic order above P1

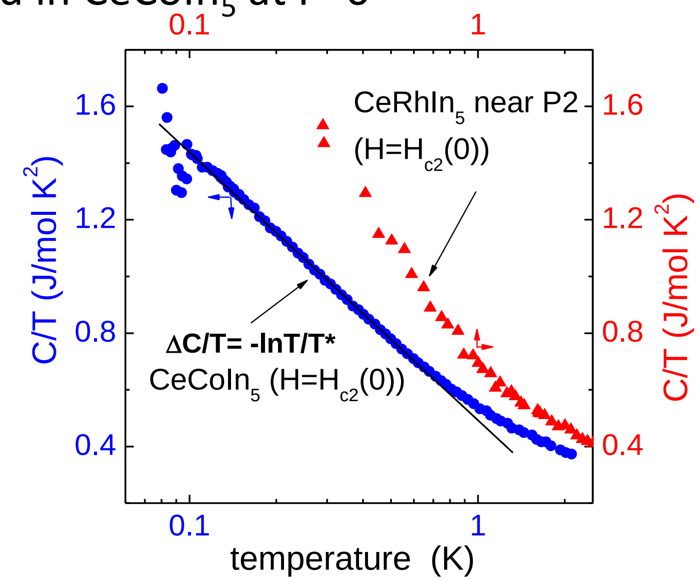


- above P1, evidence for AFM disappears in zero field
- for P1<P<P2, field-induced magnetic order in the superconducting state (T. Park et al., Nature **440**, 65 (2006); G. Knebel et al., PRB **74**, 020501 (2006))

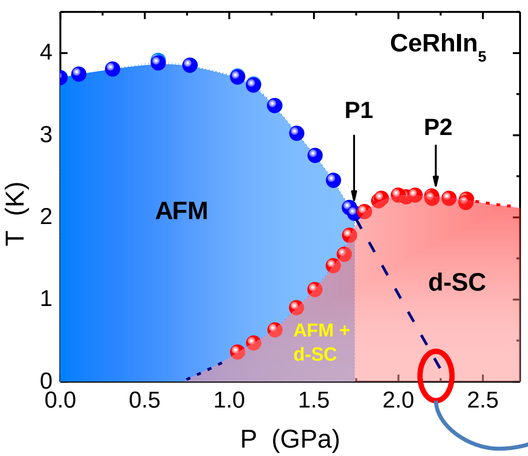


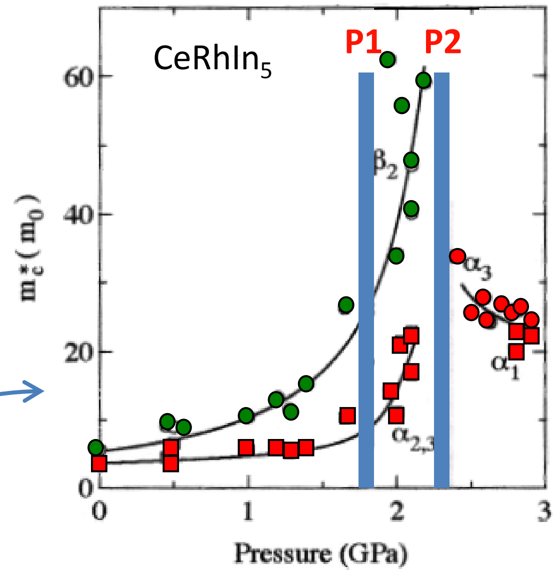


- near P2 and for $H \approx H_{c2}(0)$, divergence of $C/T \propto m^*$, as expected at an antiferromagnetic quantum-critical point and as found in $CeCoIn_5$ at P=0



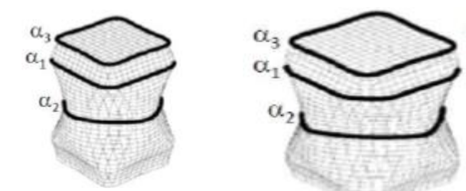
magnetic quantum + Fermi surface fluctuations





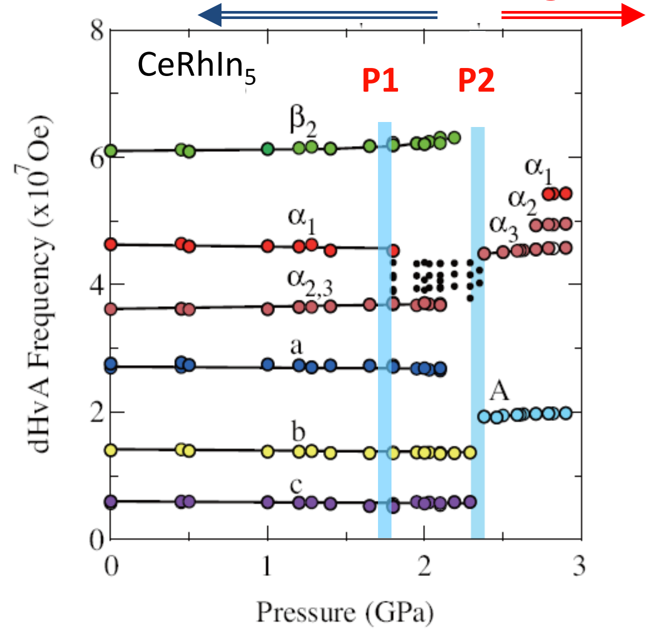
♦ at P2, divergence of m*(P) from deHaas-van Alphen measurements ⇒ confirmation of implication from C/T

H. Shishido et al., JPSJ **74**, 1103 (2005)



small FS

large FS



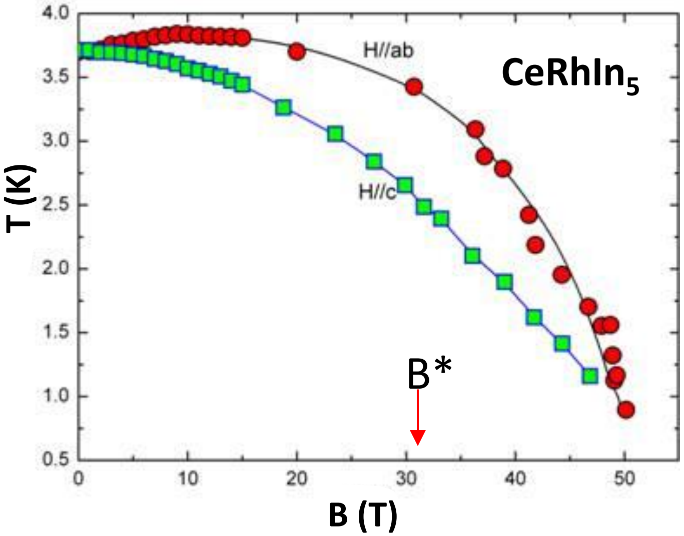
surprise: significant effect on Fermi volume at P2

♦ abrupt increase in dHvA frequencies (∝ Fermi-surface dimensions) ⇒ change from small Fermi volume below P2 (Ce 4f-electrons localized and not contributing to Fermi sea) to large Fermi volume at P>P2 (4f-electron contributes to Fermi volume) as in CeCoIn$_5$

♦ electronic degrees of freedom intimately involved; NOT expected for conventional (Hertz-type) magnetic quantum criticality ; very similar to CeIn$_3$

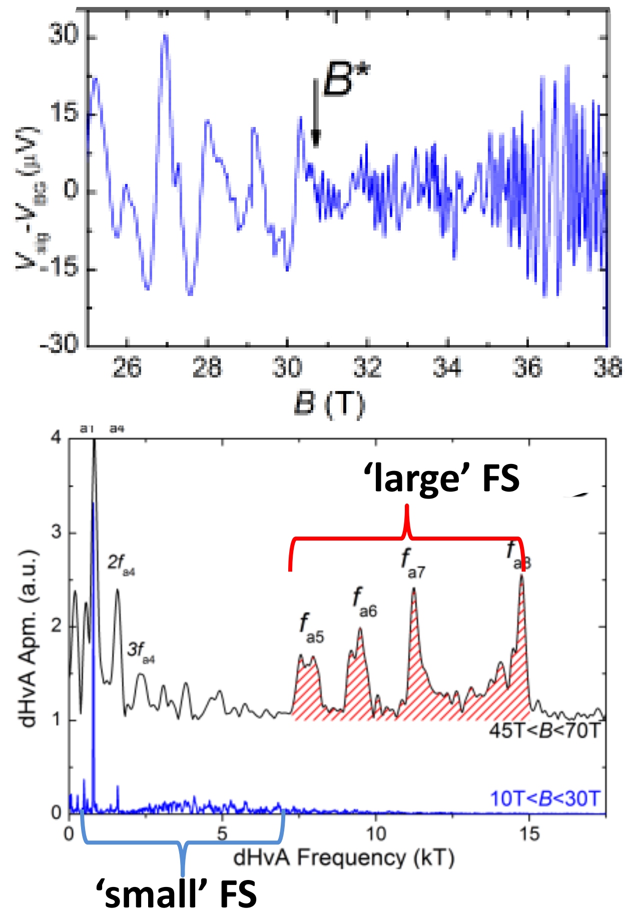
♦ superconductivity possible from fluctuations at an 'unconventional' quantum-critical point?? (T. Park et al., Nature **456**, 366 (2008))

$CeRhIn_5$ versus field at P=0

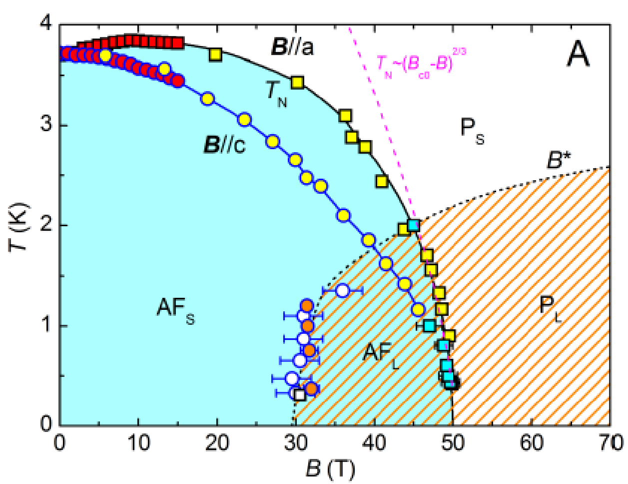


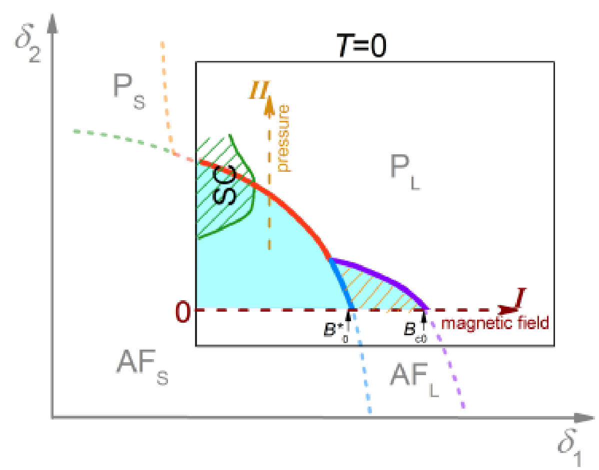
◆ P=0 deHaas-vanAlphen measurements ⇒ new frequencies above B*≈ 31T for B//*a* and *c*-axes (L. Jiao et al., PNAS **112**, 673 (2015))

◆ frequencies of 'large' FS above B* similar to those of $CeCoIn_5$



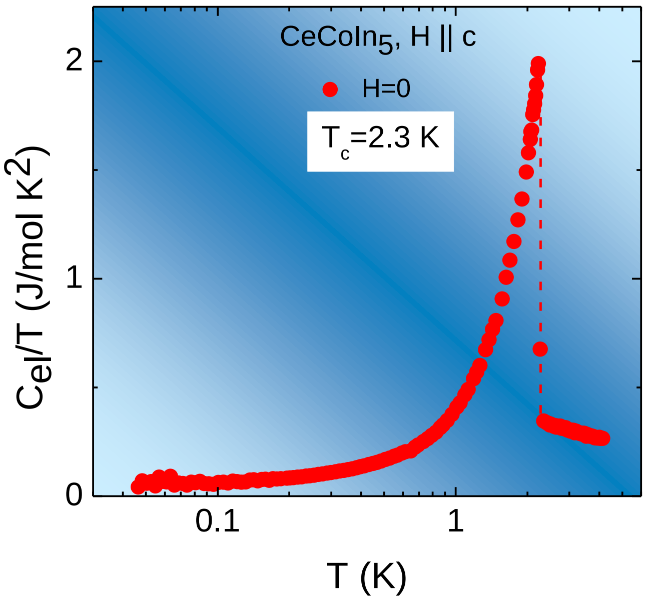
◆ QCP at 50T from a large FS ⇒ SDW-like criticality with small to large FS near 31T; unlike FS change coincident with $T_N \rightarrow 0$ at P2



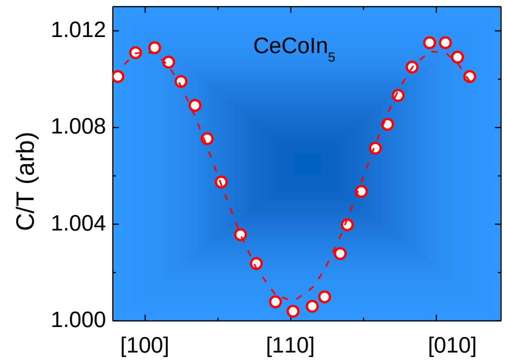


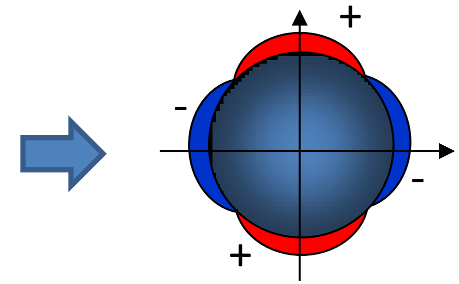
◆ appears to map onto 'global' phase diagram (Q. Si, Physica B **27**, 378 (2006); P. Coleman et al, JLTP **161**, 182 (2010))

$CeCoIn_5$: a prototype



♦ T<<T_c : power laws in C/T, κ, λ, and $1/T_1$ -- all consistent with nodal d-wave (R. Movshovich et al., PRL **86**, 5152 (2001); S. Ozcan et al. EPL **62**, 412 (2003); Y. Kohori et al., PRB **64**, 134526 (2001))

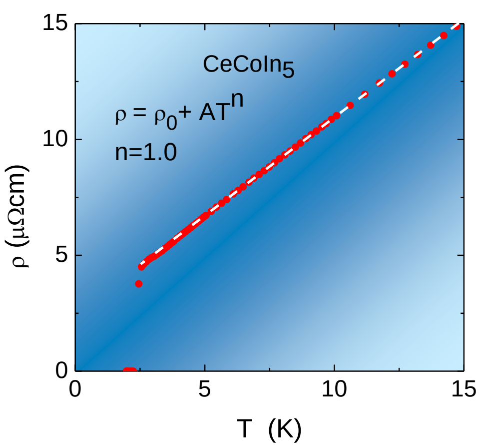


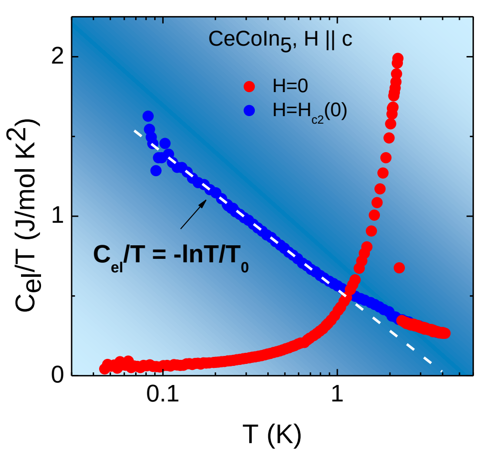


K. An et al., PRL **104**, 037002 (2010)

♦ only the 2nd Ce-based heavy-fermion superconductor at P=0 since $CeCu_2Si_2$ but with > 4 times higher T_c (C. Petrovic et al., J. Phys.:Condens. Matter **13**, L337 (2001))

♦ d-wave symmetry confirmed by STM (B. B. Zhou et al., Nature Phys. **9**, 474 (2013))





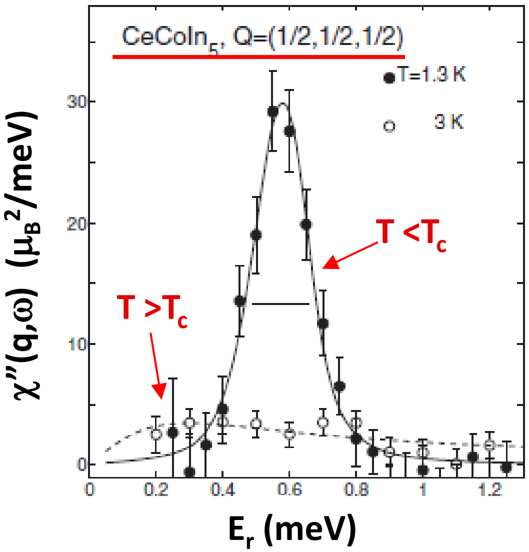
♦ T_c to ~ 20K --

- transport: $\rho \propto T$
- thermodynamics: $C/T \propto m^* \propto -\ln T/T_0$
- spin dynamics: $1/T_1 \propto T^{1/4}$

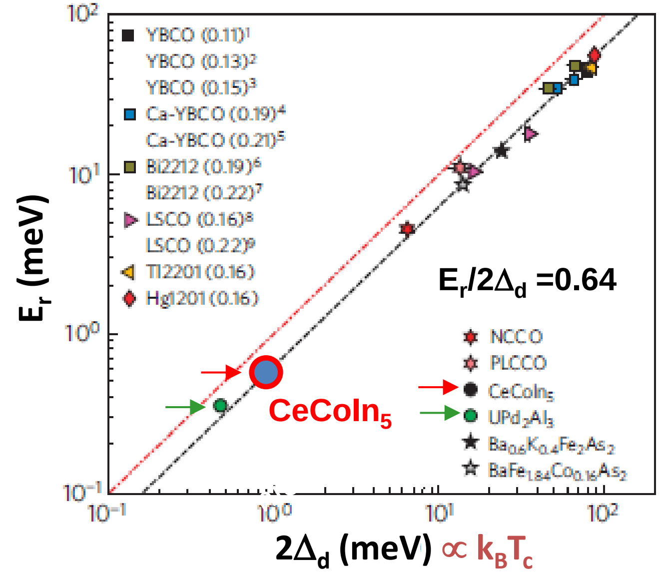
♦ *d-wave superconductivity 'born' from a NFL state above T_c consistent with expectations of proximity to an antiferromagnetic quantum-critical point*

similarities to cuprates

- a spin resonance below T_c at an energy expected from other d-wave superconductors; spectral weight $\sim 0.36\mu_B^2$ split into doublets with applied field: lower energy mode→0 near 11.2T (C. Stock et al., PRL **109**, 167207 (2012))

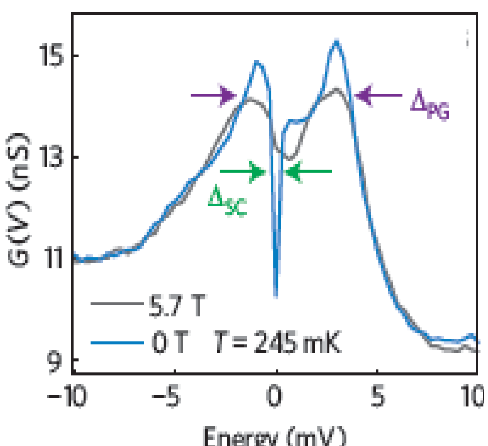


C. Stock et al., PRL **100**, 087001 (2008)

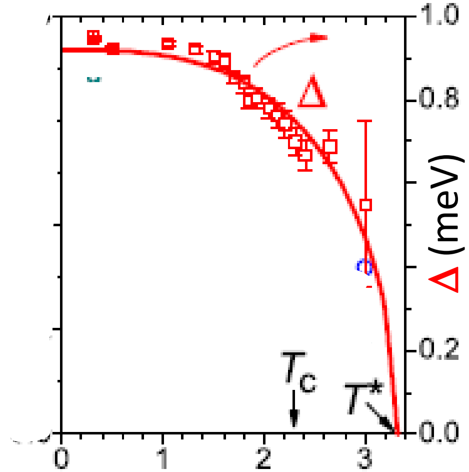


G. Yu et al., Nat. Phys. **5**, 873 (2009)

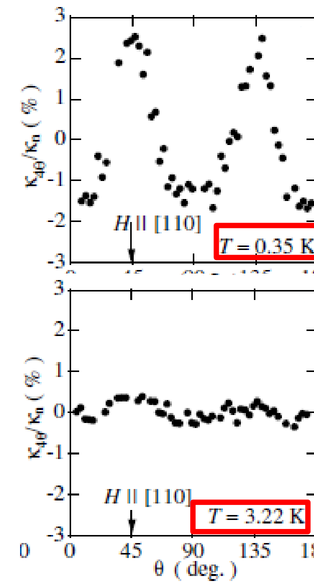
- a pseudogap above T_c that persists above $H_{c2}(0)$ and that has a d-wave like temperature and angle dependence



B. B. Zhou et al, Nature Phys., **9**, 474 (2013)



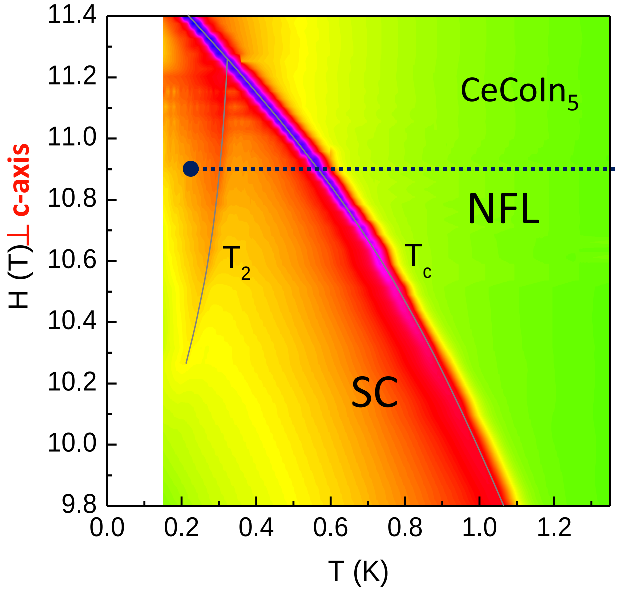
S. Wirth et al., J. Phys. Soc. Jpn. **83**, 061009 (2014)



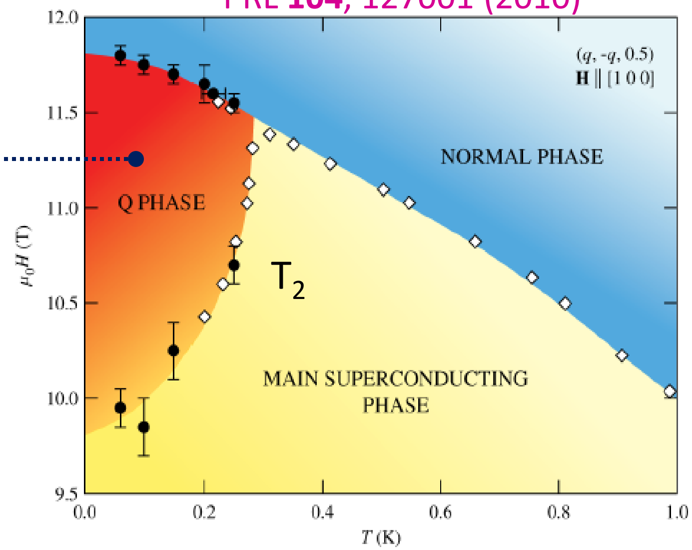
K. Izawa et al. PRL **85**, 057002 (2001)

field-induced coexistence of magnetism and superconductivity

A. Bianchi et al., PRL **91**, 187004 (2003)

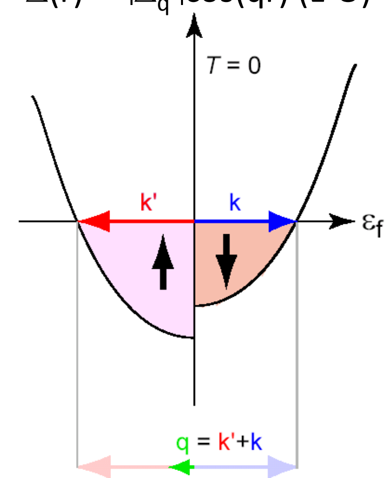


M. Kenzelmann et al., Science **321**, 1652 (2008); PRL **104**, 127001 (2010)



$\Delta(r) = |\Delta_q|\, e^{(iqr)}$ (F-F)

$\Delta(r) = |\Delta_q| \cos(qr)$ (L-O)

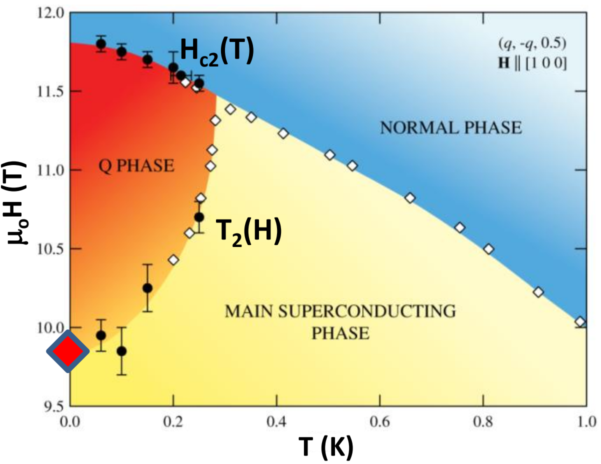


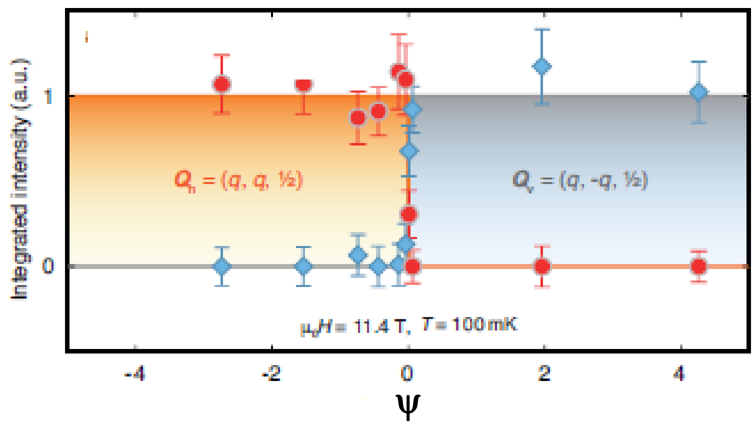
♦ $CeCoIn_5$: new phase transition at $T_2(H)$ inside the low-T, high-H superconducting state for H ⊥ c-axis, consistent with expectations of FFLO -- a spatially modulated superconducting state

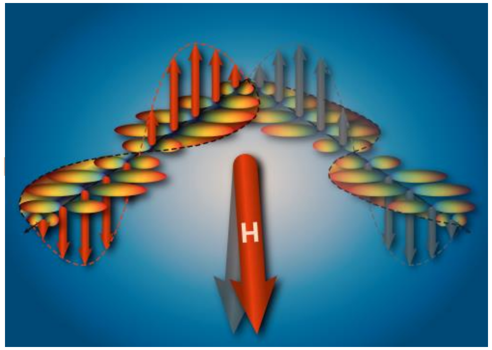
♦ subsequently, shown to be magnetic by neutrons: field-induced **Q**=(0.45, 0.45, 0.5) with $\mu_{ord} \approx 0.15\mu_B$ for H// [110] and [100] – the 'Q' phase

♦ superconductivity *necessary* for magnetic order ⇒ *coupling of AFM and SC parameters*, with magnetic correlation length (~3000 Å) >> superconducting coherence length (~80 Å) ⇒ microscopic coexistence also shown by NMR (B.-L. Young et al. PRL **98**, 036402 (2007))

field-induced QCP for H ⊥ [001]



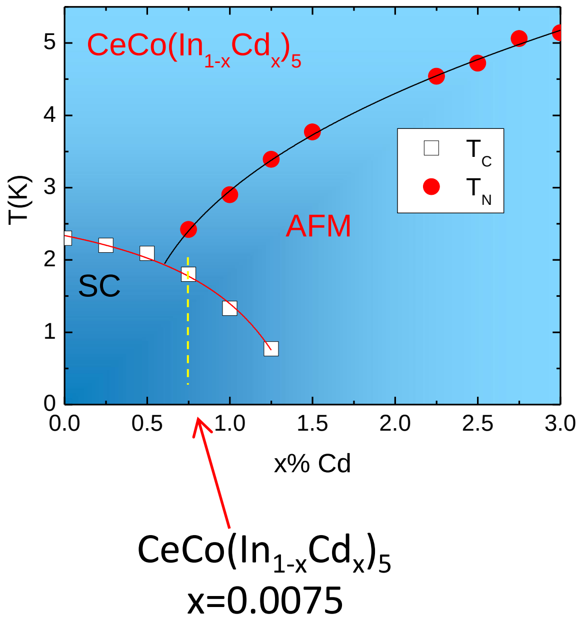


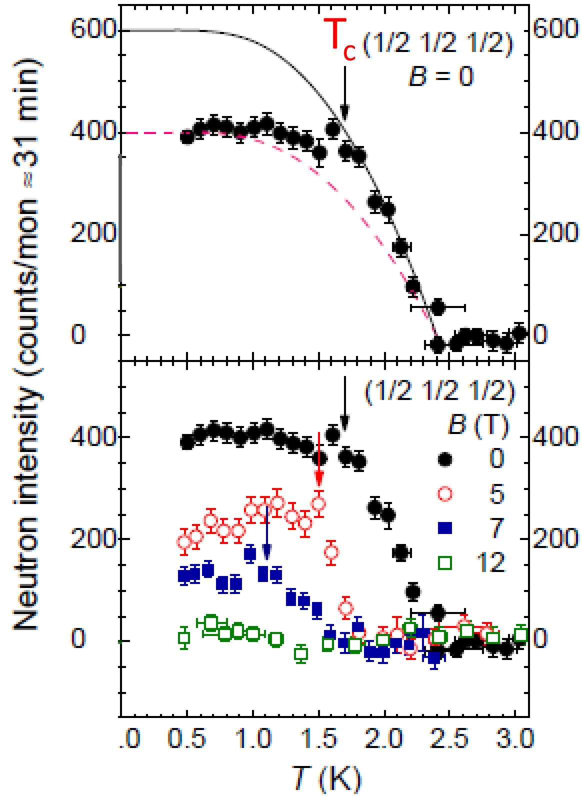


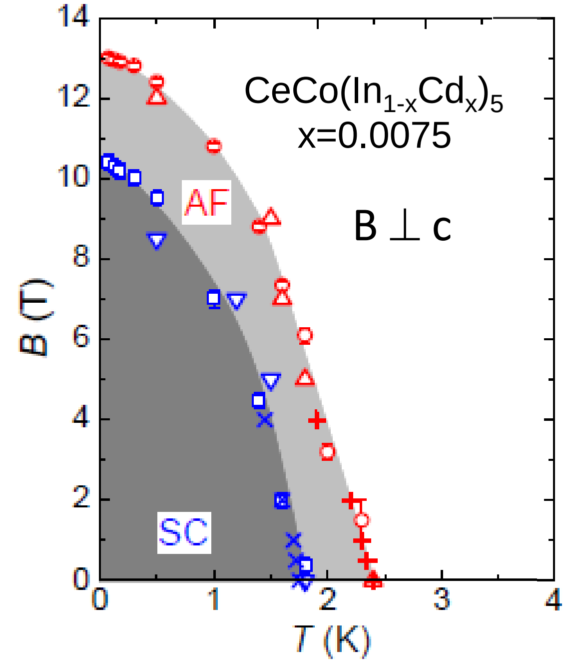
S. Gerber et al., Nature Phys. **10**, 126 (2014)

- ♦ two possible magnetic domains, $\mathbf{Q_h}$=[q, q, 0.5] and $\mathbf{Q_v}$= [q,-q, 0.5], with first order transition between domain populations as H rotated by $\Psi \leq$ **0.2°** away from [100]
- ♦ theory: *coupled* d-wave (L=2, S=0) superconductivity and antiferromagnetic order induces coexisting p-wave (L=1,S=1) superconductivity ⇒ a Cooper-pair density wave (PDW) of mixed L=2,S=0/L=1,S=1 nature (A. Aperis et al., PRL **104**, 216403 (2010))
- ♦ nodal direction of p-wave component identical to the magnetic **Q** vector ⇒ mechanism for domain switching: magnetic field controls the line node of the p-wavefunction, determining the direction of the magnetic wavevector
- ♦ a novel magneto-superconducting QCP at 9.8 T for H//[100]; divergence of m*(H) strongest near boundary, as expected for a QCP
- ♦ open question: can/does an FFLO state coexist with PDW order?

coupled orders





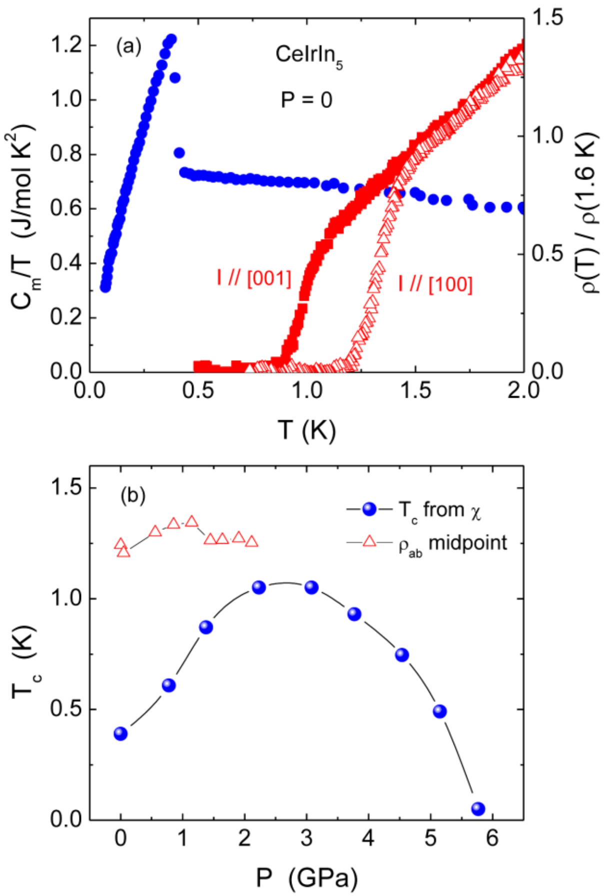


S. Nair et al., PNAS **107**, 9537 (2010)

- ♦ Cd-induced ‘large’ moment, commensurate AFM (as in $CeIn_3$); for x=0.0075, mean-field fit to I(T) for $T_c < T < T_N$ clearly deviating at T_c
- ♦ relationship between T_c and T_N unchanged in magnetic field, with $T_N(B)$ completely enclosing $T_c(B)$
- ♦ with $\xi_{AFM} \sim 3\xi_{GL}$, microscopic coexistence of f-derived pairs of heavy quasiparticles and magnetic order of f-moments $\Rightarrow$ entangled roles of f-electrons
- ♦ useful to explore nature of field-tuned T=0 antiferromagnetic transition – Hertz-Millis-like or unconventional?

$CeIrIn_5$

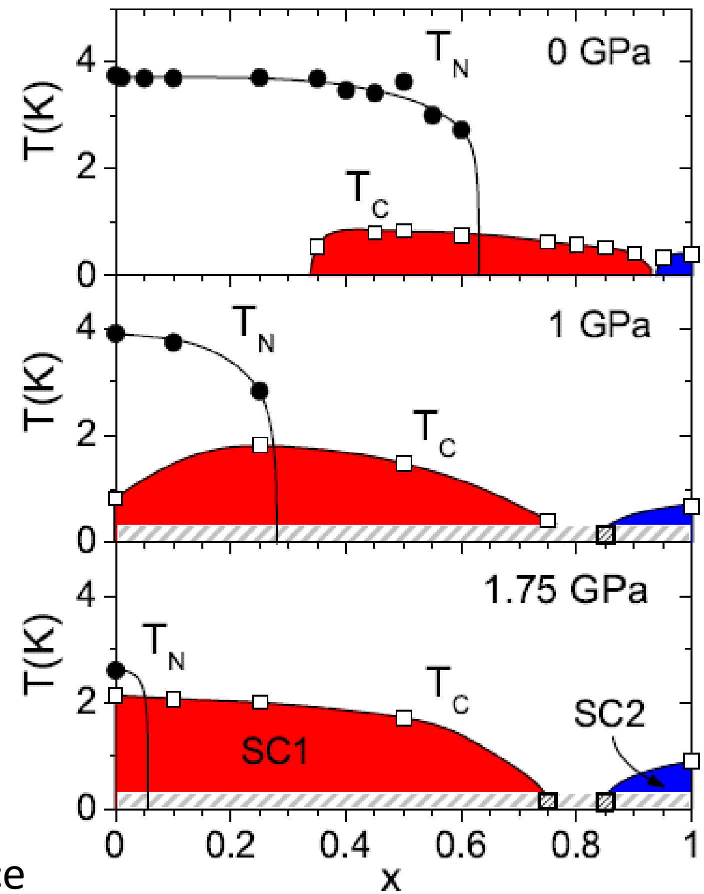
- $CeIrIn_5$ – perhaps the 'strangest' and certainly the least understood of the Ce115s



- nominally isoelectronic with and similar large Fermi surface as $CeCoIn_5$; NFL normal state; nodal superconductivity; BUT T_c~1/5 that of Co115 and Rh115 under pressure

- anisotropic resistive T_c ~ 3x bulk T_c – reproducible & robust; transitions approach near T_c max

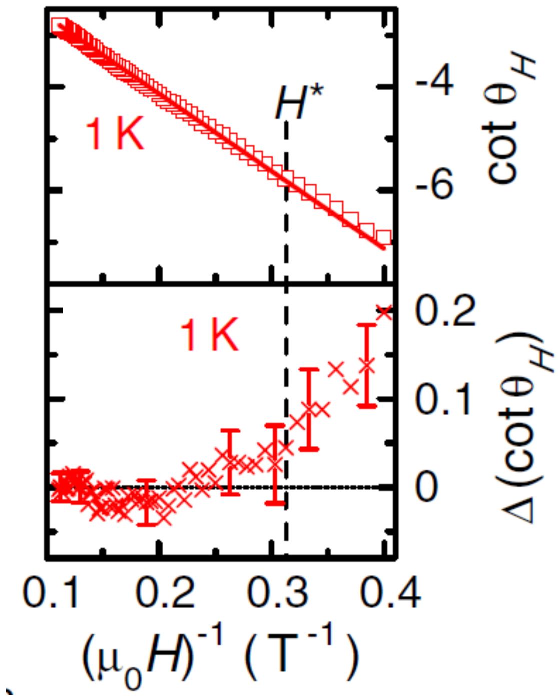
- bulk $T_c \rightarrow 0$ if substitute 10%Rh for Ir and T_c 'notch' becomes a gap under pressure (M. Nicklas et al., PRB 70, 020505(R) (2004))



$CeRh_{1-x}Ir_xIn_5$

- origin of difference in resistive and bulk transitions? different superconducting mechanism in Ir115? evidence for a magnetic mechanism (Y. Chen et al, PRL, **114**, 146403 (2015))

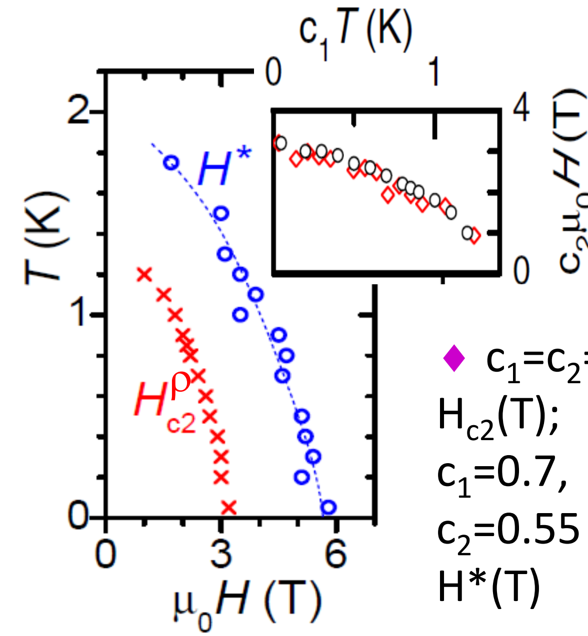
precursor state in $CeIrIn_5$



♦ deviation from $\cot\theta_H \propto 1/H \Rightarrow$ decrease in Hall mobility below H*(T)

S. Nair et al. PRL **100**, 137003 (2008)

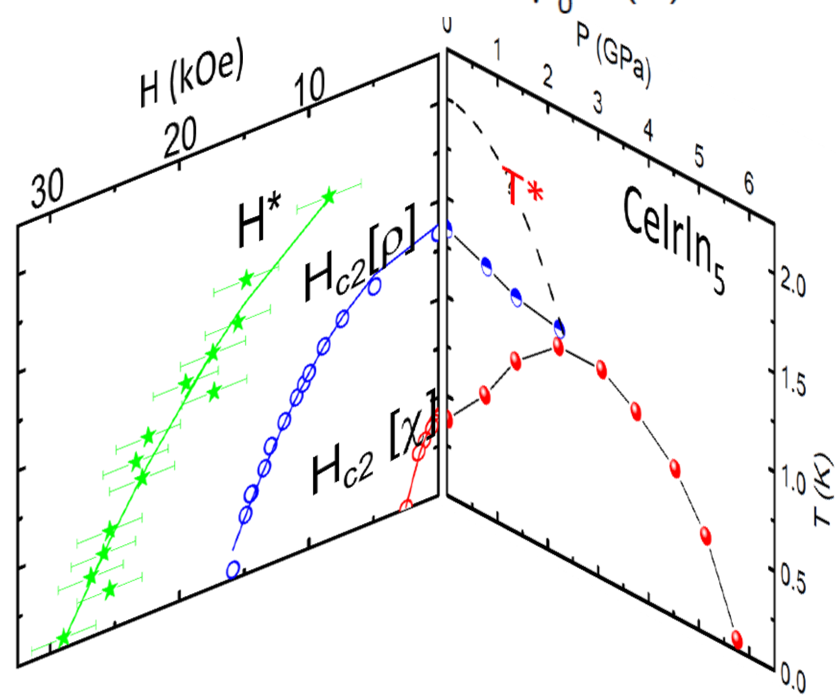
♦ scaling H*(T) onto H_{c2}(T) $\Rightarrow$ superconductivity and H* related?



♦ $c_1=c_2=1$ for H_{c2}(T); $c_1=0.7$, $c_2=0.55$ for H*(T)

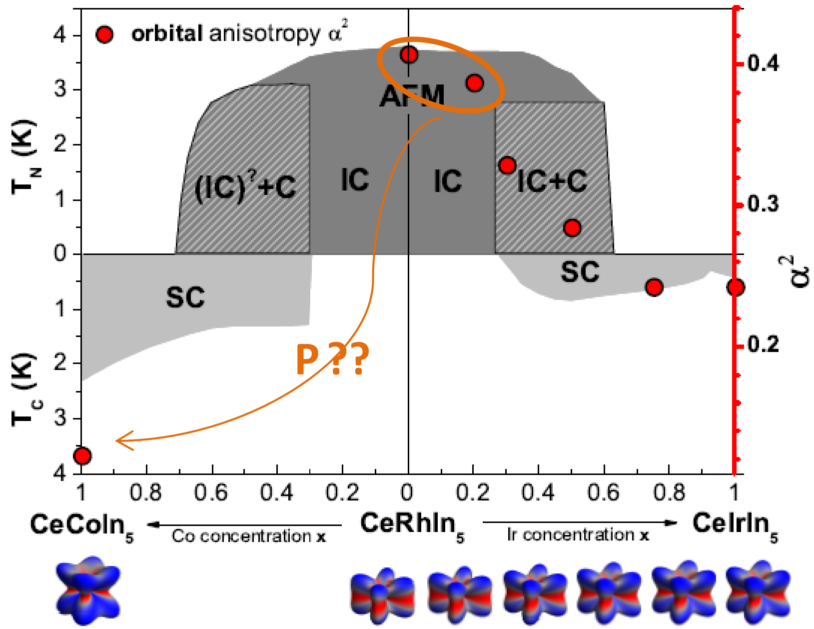
♦ not a phase transition but a pseudogap-like electronic state that extrapolates to ~2 K at zero field

♦ pure speculation: pseudogap-like phase possible origin of difference between resistive and bulk T_c's



a hint why T_c in $CeIrIn_5$ is low

♦ a Γ_7 CEF groundstate in all Ce115s: $\Gamma_7 = \alpha|\pm 5/2\rangle + \sqrt{1-\alpha^2}|\mp 3/2\rangle$, where α^2 is a measure of the out-of-plane orbital anisotropy of the 4f wavefunction

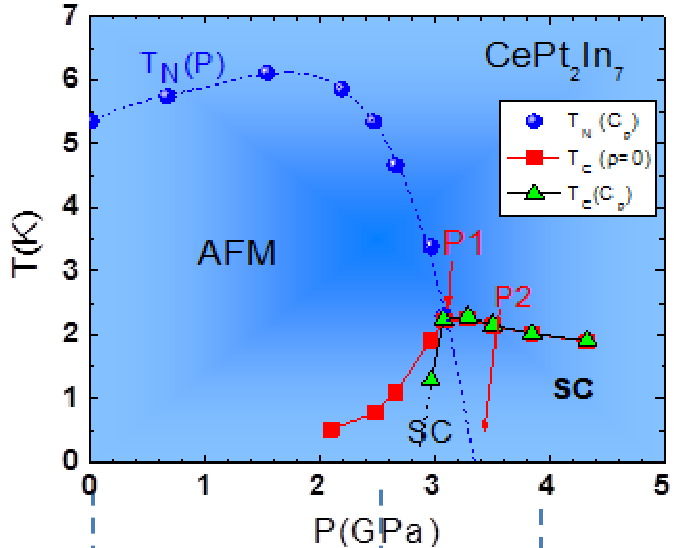


T. Willers et al., PNAS **112**, 2384 (2015)

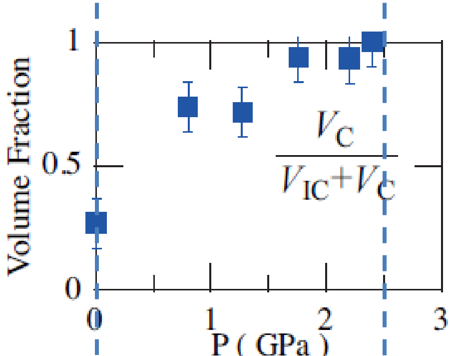
♦ momentum-dependent 4f-conduction-band hybridization influenced by α^2; reasonable that pressure promotes *f-c* hybridization sufficiently so that T_c(P) of $CeRhIn_5$ approaches that in $CeCoIn_5$ but P-induced hybridization is limited for some reason (spin-orbit coupling??) in $CeIrIn_5$?

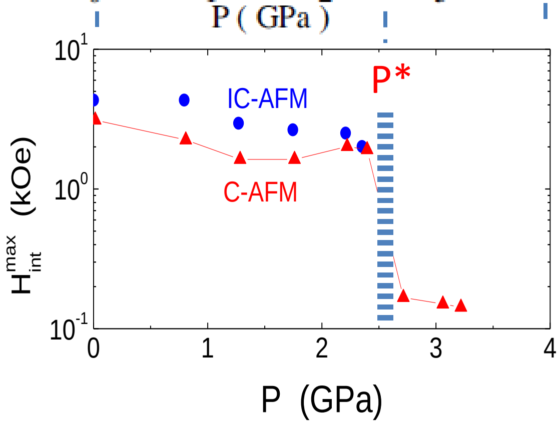
♦ still very much an open question

magnetism and superconductivity in $CePt_2In_7$

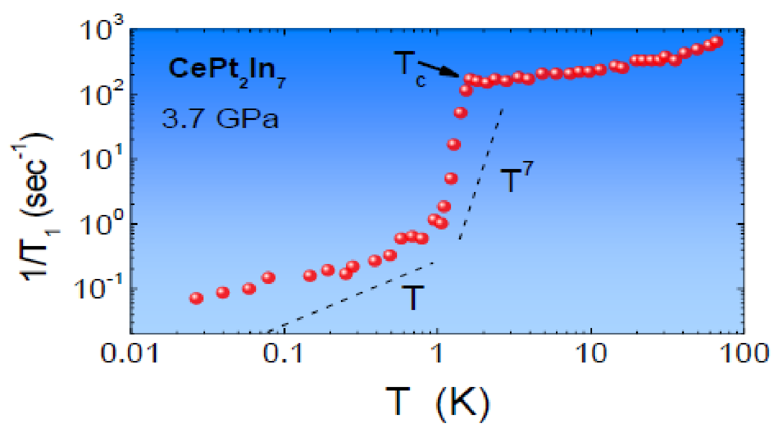


- ♦ pressure-induced SC (V. A. Sidorov et al., PRB **88**, 020503 (2013)) from complex magnetic order: commensurate AFM just below T_N followed at lower temperatures by coexisting incommensurate order; volume fraction of commensurate order ≈ 100% at P*~2.4 GPa <P1 (H. Sakai et al., PRB **83**, 140408 (2011))
- ♦ collapse of internal field produced by magnetic order by over an order of magnitude at P* (< P1< P2) where ν_Q also increases sharply ⇒ a 4f-localized/delocalized transition *in the ordered state* and approximately coincident with the emergence of bulk superconductivity (H. Sakai et al., Phys. Rev. Lett. **112**, 206401 (2014)); *expect change in Fermi volume at P**
- ♦ maximum T_c near P1 = 3.07 GPa, not at the extrapolated (SDW??) critical point P2





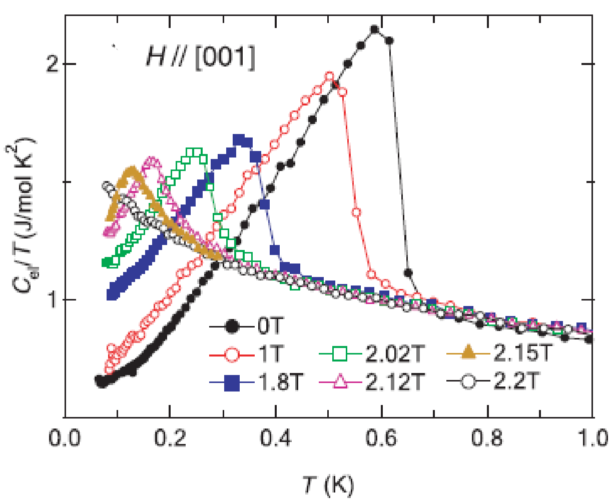
- ♦ at 3.1 GPa, $1/T_1 \propto T^3$ ⇒ nodal, possibly d-wave, superconductivity, but at 3.7 GPa > P2, apparently 1^{st} order ⇒ coupling of SC to a new nearby state?

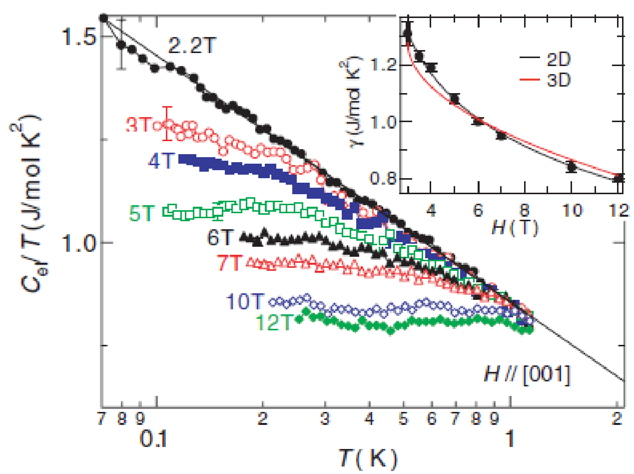


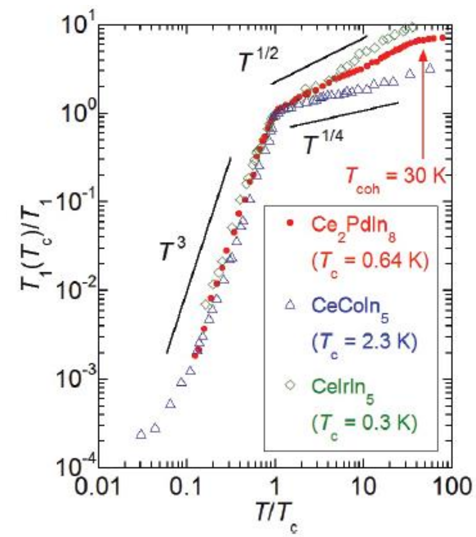
Ce_2MIn_8 superconductors (briefly)

- generally, more difficult to grow good crystals and studied relatively little
  - Ce_2RhIn_8: antiferromagnetic (T_N=2.8 K) and pressure-induced superconductivity (by resistivity) with max. T_c=2 K at 2.8 GPa (M. Nicklas et al., PRB **67**, 020506 (2003))
  - Ce_2CoIn_8: ambient pressure superconductivity with T_c = 0.4 K (G. Chen et al, JPSJ **71**, 2836 (2007))
  - Ce_2PdIn_8: ambient pressure superconductivity with T_c=0.68 K (D. Kaczorowski et al. PRL **103**, 027003 (2009)) with impurity phase antiferromagnetism

- Ce_2PdIn_8 similar to $CeCoIn_5$: T-linear $\rho(T)$ above T_c, and –lnT dependence of C/T (Y. Tokiwa et al., PRB 84, 14507 (2011)); $1/T_1 \propto T^3$ below T_c and $\propto T^{1/2}$ above T_c (H. Fukazawa et al., J.P. Conf. Series 449, 012027 (2013) $\Rightarrow$ probably nodal d-wave mediated by magnetic fluctuations

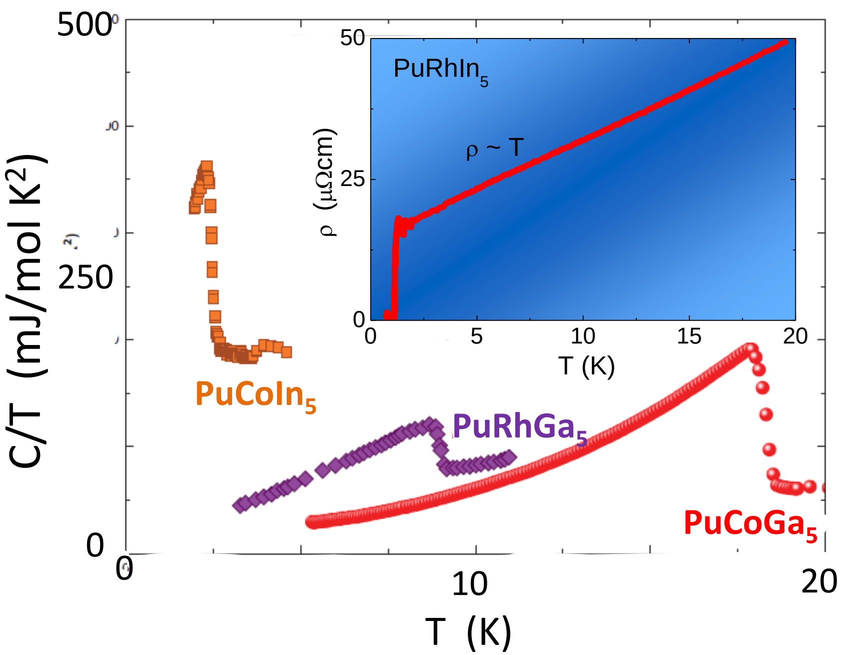






- possibility of a field-induced 'Q-phase' as in $CeCoIn_5$ or Cd-induced magnetic order?

Ce115 spin-offs: the Pu115s

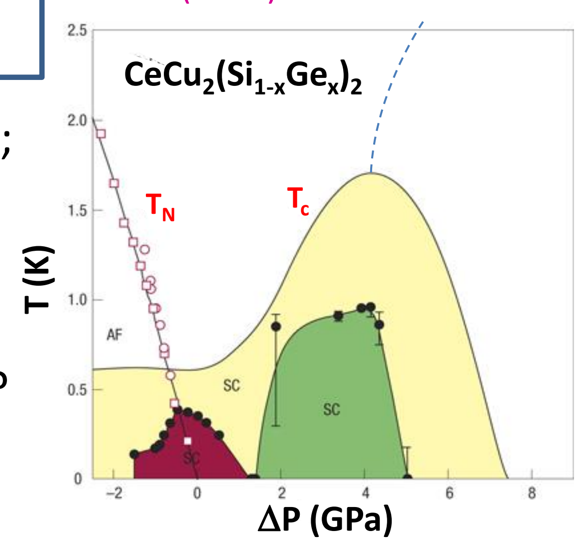


- ◆ $PuRhIn_5$ (E. D. Bauer et al., Phil. Mag. **92**, 2466 (2012)) and $PuCoIn_5$ (E. D. Bauer et al., JPCM **24**, 052206 (2012)) newer members, with T_c=1.5 K (Rh) and 2.5 K (Co) and largest Sommerfeld coefficient ($\gamma \sim 300$ mJ/molK^2) among Pu115s
- ◆ Ga analogs: $PuRhGa_5$ with T_c=9.1 K ($\gamma \sim 100$ mJ/molK^2) (F. Wastin et al., JPCM **15**, S1911(2003)) and $PuCoGa_5$ with T_c=18. 5K ($\gamma \sim 80$ mJ/molK^2) (J. L. Sarrao et al., Nature **420**, 297 (2002))
- ◆ non-Fermi-liquid behaviors above T_c in each

$V_{PuRhIn5}$ = 159 Å^3 > $V_{PuCoIn5}$ = 156 Å^3
$V_{CeRhIn5}$ = 163 Å^3 > $V_{CeCoIn5}$ = 160 Å^3
>>
$V_{PuRhGa5}$ = 127 Å^3 > $V_{PuCoGa5}$ = 122 Å^3

isostructural volume collapse (20%); possible change of Pu valence?

H. Q. Yuan et al., Science **302**, 2104 (2003)



- ◆ possibly two domes of superconductivity as in $CeCu_2(Si_{1-x}Ge_x)_2$?

summary

♦ many open questions:

- How do we really understand coexisting magnetism and unconventional superconductivity when both orders involve the single 4f electron of Ce or multiple 5f electrons in U- and Pu-systems?
- To what additional extents are $CeIn_3$ and $CeRhIn_5$ alike or different?
- Does their superconductivity arise from pairing induced by fluctuations at unconventional Kondo-breakdown quantum criticality (perhaps --J. K. Pixley et al., arXiv:1308.0839)?
- What is the origin of the pseudogap in $CeCoIn_5$ and precursor state in $CeIrIn_5$?
- Does an FFLO state reside in the field-induced Q-phase of $CeCoIn_5$?
- What is the relationship between magnetism and superconductivity in $CePt_2In_7$?
- Is there a Q-phase in Ce_2PdIn_8?
- Are the Pu115s analogs of $CeCu_2Si_2$ under pressure?

♦ much not discussed:

- superconductivity in β-$YbAlB_4$ and the lack of other Yb-based HF superconductors
- non-centrosymmetric heavy-fermion superconductors, eg. $CePt_3Si$ and related compounds
- ……….

♦ On an absolute scale heavy-fermion T_c's are low but in a real sense just as high or higher than any cuprate/iron-pnictide – an exciting and challenging field worthy of more attention