ICMR Summer School on Novel Superconductors

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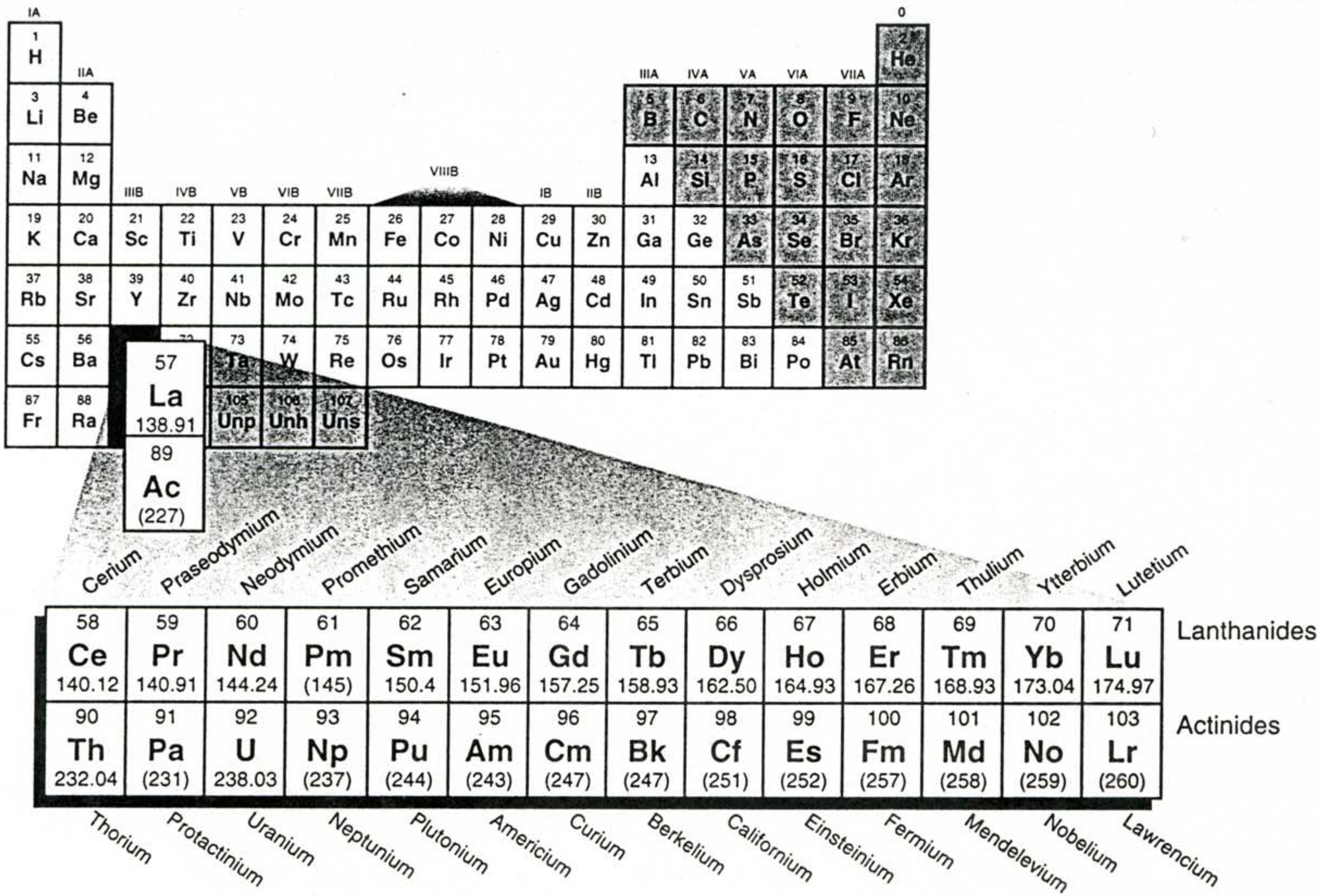
Lecture 1 (Tuesday, 8/11/09):
Interplay between superconductivity and magnetism in f-electron systems
(conventional superconductivity)

Lecture 2 (Wednesday, 8/12/09):
Unconventional superconductivity, magnetic and charge order, and quantum criticality in heavy fermion f-electron materials
(unconventional superconductivity)

f-electron materials

- f-electron materials – multinary compounds and alloys containing rare earth (R) and actinide (A) ions with partially-filled f-electron shells and localized magnetic moments
- Localized magnetic moments of R and A ions interact with the momenta and spins of the conduction electrons
- Certain R ions (Ce, Pr, Sm, Eu, Tm, Yb) and A ions exhibit valence instabilities; i.e., localized f-states hybridize with conduction electron states
- Competing interactions — readily "tuned" by x, P, H ("knobs")
- Wide variety of correlated electron phenomena
  - Rich and complex phase diagrams in the hyperspace of T, x, P, H
- Situations involving competing interactions
  - One phenomenon survives at the expense of another
  - Interactions conspire to produce a new phenomenon (e.g., sinusoidally-modulated magnetic state that coexists with SC in FM superconductors)
- Brief survey – point of view of experimentalist
- Materials driven physics!
  - Materials – reservoir of new electronic states and phenomena
  - Opened up new research directions in condensed matter physics

f-electron materials



Examples

- Destruction of SC at $T_{c2} < T_{c1}$ (reentrant SC) due to Kondo effect &
- Kondo effect (impurities and lattice) &
- Destruction of SC at $T_{c2} < T_{c1}$ (reentrant SC) due to FM order &
- Occurrence of sinusoidally modulated magnetic state ($\lambda \sim 100$ Å) that coexists with SC in FM superconductors &
- Coexistence of SC & AFM order &
- Coexistence of SC & itinerant FM &
- Magnetic field induced SC (MFIS) &
- Heavy fermion compounds ($m^* \sim 10^2\ m_e$) &
- Unconventional SC in heavy fermion compounds &
  - Electron pairing with $L > 0$, nodes in SCing energy gap, magnetic pairing mechanism
- Occurrence of SC near magnetic QCPs accessed by pressure &
- NFL behavior associated with QCPs &
- Heavy fermion behavior and SC in $PrOs_4Sb_{12}$, possibly due to electric quadruple, rather than magnetic dipole, fluctuations &

& Lecture 1; & Lecture 2

Lecture outlines

Lecture 1:

Interplay between superconductivity and magnetism in f-electron systems

☐Outline:

(1) f-electron materials

(2) Conventional superconductivity

(3) Magnetic interactions in superconductors

(4) Paramagnetic impurities in superconductors

(5) Magnetic field induced superconductivity (MFIS)

(6) Magnetic ordering via the RKKY interaction

(7) Magnetically ordered superconductors

Lecture 2:

Unconventional superconductivity, magnetic and charge order, and quantum criticality in heavy fermion f-electron materials

Outline:

(1) Unconventional superconductivity

(2) Heavy fermion compounds

(3) Superconductivity in heavy fermion compounds

(4) Competition between Kondo effect and RKKY interaction

(5) Non-Fermi liquid (NFL) behavior and quantum criticality

(6) SC near antiferromagnetic (AFM) quantum critical points (QCPs) under pressure

(7) Coexistence of SC and itinerant ferromagnetism

(8) Heavy fermion behavior and unconventional SC in $PrOs_4Sb_{12}$; evidence for electron pairing via electric quadrupole interactions

Some remarks about heavy fermion f-electron materials

f-electron materials & hybridization

- *Hybridization of localized f- & conduction-electron states*
  ⇒ interesting correlated electron physics!
- *Hybridization strength:*
  Weak ⇒ ionic behavior (f occupation number <n> integral)
  Moderate ⇒ Kondo behavior (<n> ~integral)
  Appreciable ⇒ Valence fluctuations (<n> nonintegral)
  Strong ⇒ f-bands
- *Competing interactions & coupling between spin, charge, lattice degrees of freedom ⇒ Novel electronic ground states & phenomena*
  – Magnetic & quadrupolar order
  – Valence fluctuations
  – Heavy fermion behavior
  – Hybridization gap semiconductivity (Kondo insulator behavior)
  – Non-Fermi liquid behavior
  – Unconventional (p- or d-wave) superconductivity

f-electron materials & hybridization

- *Complex phase diagrams in the hyperspace of T, P, H, composition*
- *Systems with quantum critical point (QCP):*
  - QCP: value of control (tuning) parameter δ (e.g., composition x, P, H) where 2nd order phase transition suppressed to 0 K (quantum phase transition)
  - Fluctuations or order parameter (magnetic, quadrupolar) lead to breakdown of Landau Fermi liquid theory & to formation of other phases such as glassy types of magnetic order & unconventional superconductivity
- *Materials:*

  $CeIn_3$, $Ce(Rh_{1-x}Co_x)In_5$, $PrOs_4Sb_{12}$,
  UGe_2, $Sc_{1-x}U_xPd_3$, $URu_{2-x}Re_xSi_2$

Heavy fermion f-electron materials

Underlying physics — significant hybridization between localized f- & conduction-electron states

- AFM exchange interaction

  $H_{ex} = - 2J\mathbf{S\bullet s}$ where $J \sim -\langle V_{kf}^2\rangle/(E_F - F_f) < 0$

- Kondo effect (lattice of Ln, U ions)

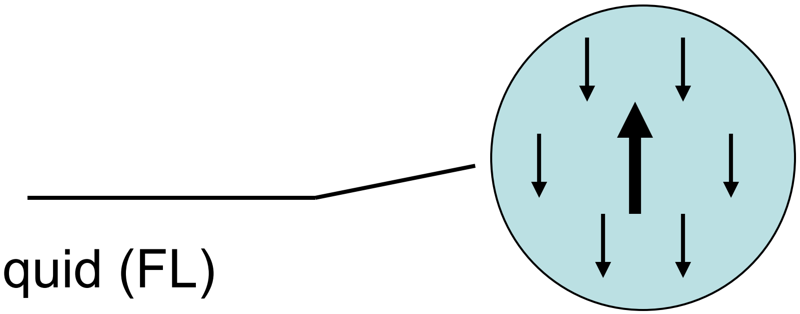
  $T_K \sim T_F \exp(-1/N(E_F)|J|) \sim T^*$ (effective T_F)

- T >> T*: local moment behavior

  $\chi(T) \sim N\mu_{eff}^2/3k_B(T+T^*)$

  $\rho(T) \sim -\ln T$

- T << T*: many body singlet

  Nonmagnetic heavy Fermi liquid (FL)

  $\chi(T) \rightarrow \chi_o \propto m^* \propto 1/T^*$

  $\gamma(T) = C_e(T)/T \rightarrow \gamma_o \propto m^* \propto 1/T^*$

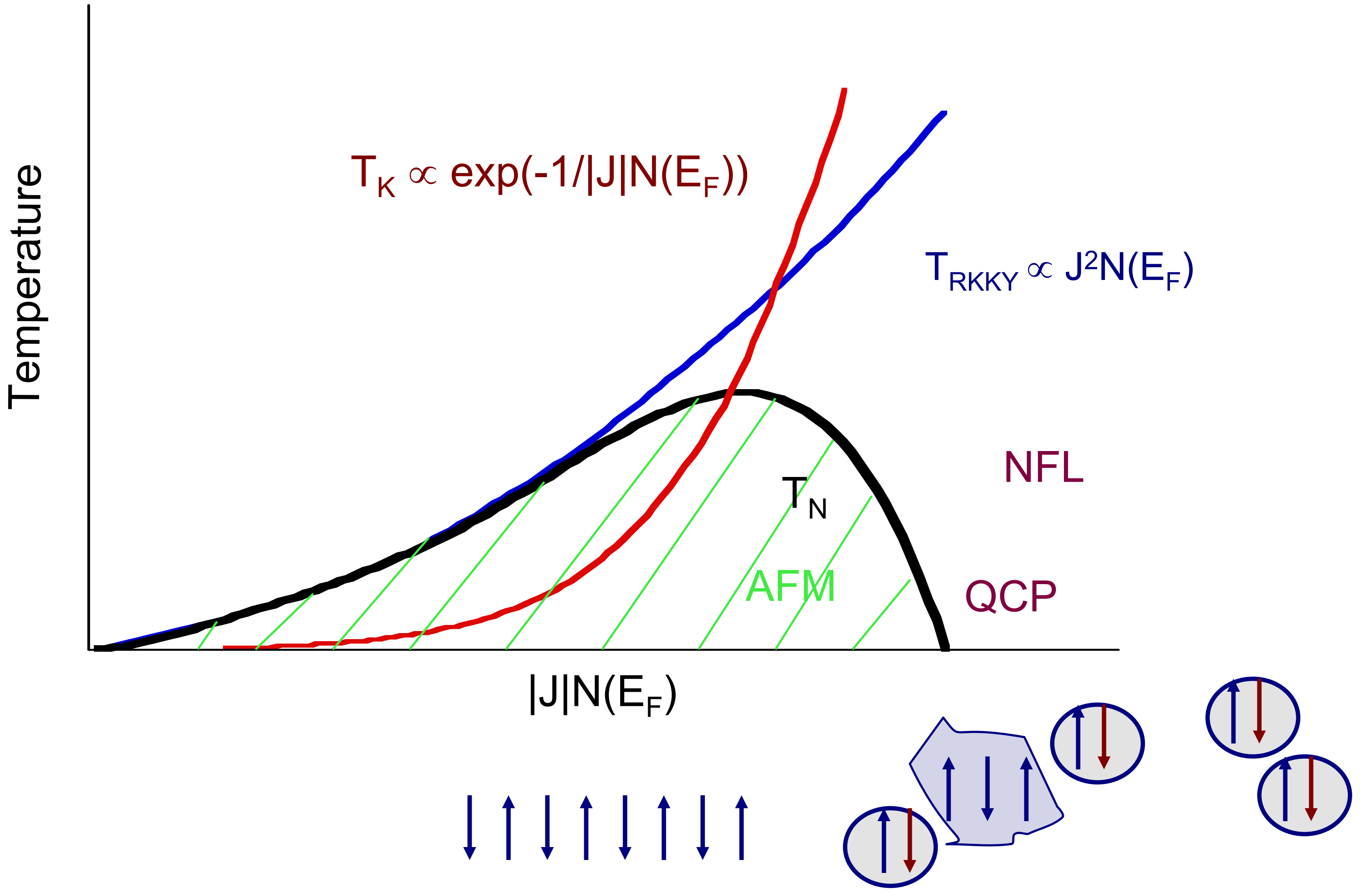
  $R = (\chi_o/\mu_{eff}^2)/(\gamma_o/\pi^2 k_B^2) \approx 1$ (Wilson-Sommerfeld ratio)

  $\rho(T) \propto \rho_{e\text{-}e}(T) \sim AT^2$ with $A \sim \gamma_o^2$

- Heavy FL unstable to SC & magnetic order (RKKY)

Kondo effect vs RKKY interaction

Doniach phase diagram



Superconductivity

- Pairing of electrons with $L > 0$

  $L=1$ (p-wave), $S=1$ (triplet)

  $L=2$ (d-wave), $S=0$ (singlet)

- Pairing mechanism — spin fluctuations
- Anisotropic energy gap $\Delta(\mathbf{k}) \neq$ const.

  $\Delta(\mathbf{k})$ vanishes at points or lines on Fermi surface

- Superconducting properties ~ T^n for $T \ll T_c$

  e.g., $C_e(T) \sim T^n$ ($n=2$, line nodes; $n=3$, point nodes)

  [$\exp(-\Delta/T)$ with const. Δ for BCS superconductor]

- Multiple superconducting states (complex T-x-P

  phase diagrams)

  e.g., UPt_3, $U_{1-x}Th_xBe_{13}$, $PrOs_4Sb_{12}$ (quadrupolar?)

- Sometimes occurs near x or P where $T_M \rightarrow 0$ K

  (quantum critical point — QCP)

Non-Fermi liquid behavior

- Materials: U, Ce, Yb intermetallic compounds
  - Chemically substituted: $Y_{1-x}U_xPd_3$, $UCu_{5-x}Pd_x$, $CeCu_{6-x}Au_x$,…
  - Stoichiometric: (P=0) – UBe_{13}, $CeCoIn_5$,$YbRh_2Si_2$,…;
    (P>0) – $CeIn_3$, $CePd_2Si_2$,…
- Physical properties — weak power law, logarithmic divergences in T at low $T << T_o$
  - $\rho(T) \approx \rho(0)[1 \pm (T/T_o)^n]$ $(1 \leq n \leq 1.5)$
  - $C(T)/T \approx -(1/T_o)\ln(T/T_o)$, $(T/T_o)^{-1+\lambda}$
  - $\chi(T) \approx \chi(0)[1 - (T/T_o)^n]$ $(n \sim 0.5)$, $-(1/T_o)\ln(T/T_o)$,
    $(T/T_o)^{-1+\lambda}$, $C/(T^\alpha + \theta)$
  - $\chi''(\omega,T)$: ω/T scaling
- Appreciable T-dependence below $T_o \Rightarrow$ lower energy scale than Fermi liquid (FL)

Non-Fermi liquid behavior

Experiments ⇒ two routes to NFL behavior:

(1) Single ion & (2) inter-ionic interactions

(1) Single ion models

- Multichannel Kondo effect
- Single channel Kondo effect with disorder — $P(T_K)$

(2) Interacting ion models

- Fluctuations of order parameter above 2nd order phase transition at 0 K
- Griffiths' phase — interplay between disorder & competing Kondo and RKKY interactions

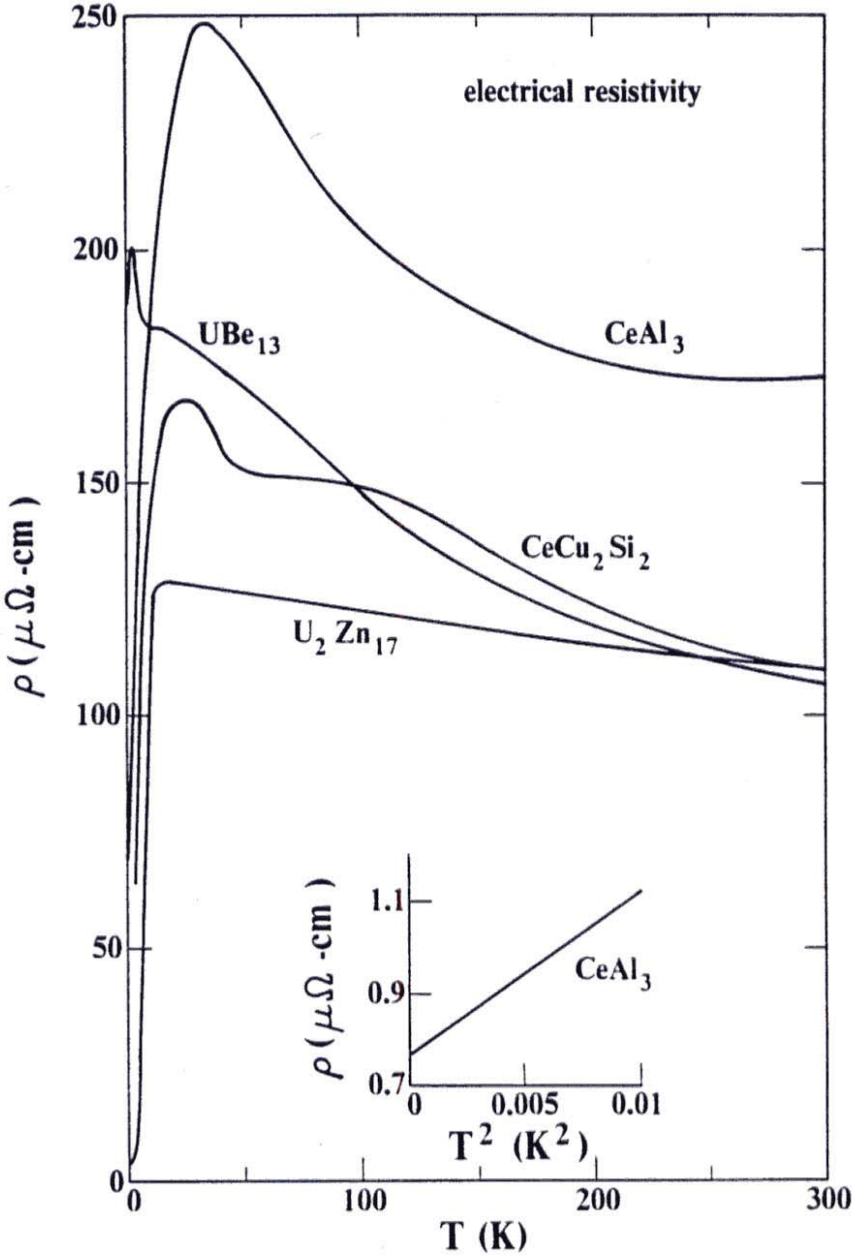
Superconductivity near magnetic QCP apparently mediated by spin fluctuations

This talk

- General overview heavy fermion and non-Fermi liquid behavior in f-electron systems with examples
- Recurring theme: interplay between SC and charge or spin order
    - A few examples of SC occurring in the vicinity of phase boundaries for charge or spin order that suggests that the ordered phase is involved in the SC
        - Several possible scenarios:
            - SCing electron pairing mediated by fluctuations of an order parameter near quantum critical point (QCP)
            - Liberation of the SCing phase through the suppression of the ordered phase which competes with SC
            - Direct participation of the ordered phase in the SCing electron pairing
    - Transition metal oxypnictides (SDW)
    - Lanthanide tritellurides (CDW)
    - URu_2Si_2 (“hidden order” DW phase, various magnetic and NFL phases)
    - SC of the elements

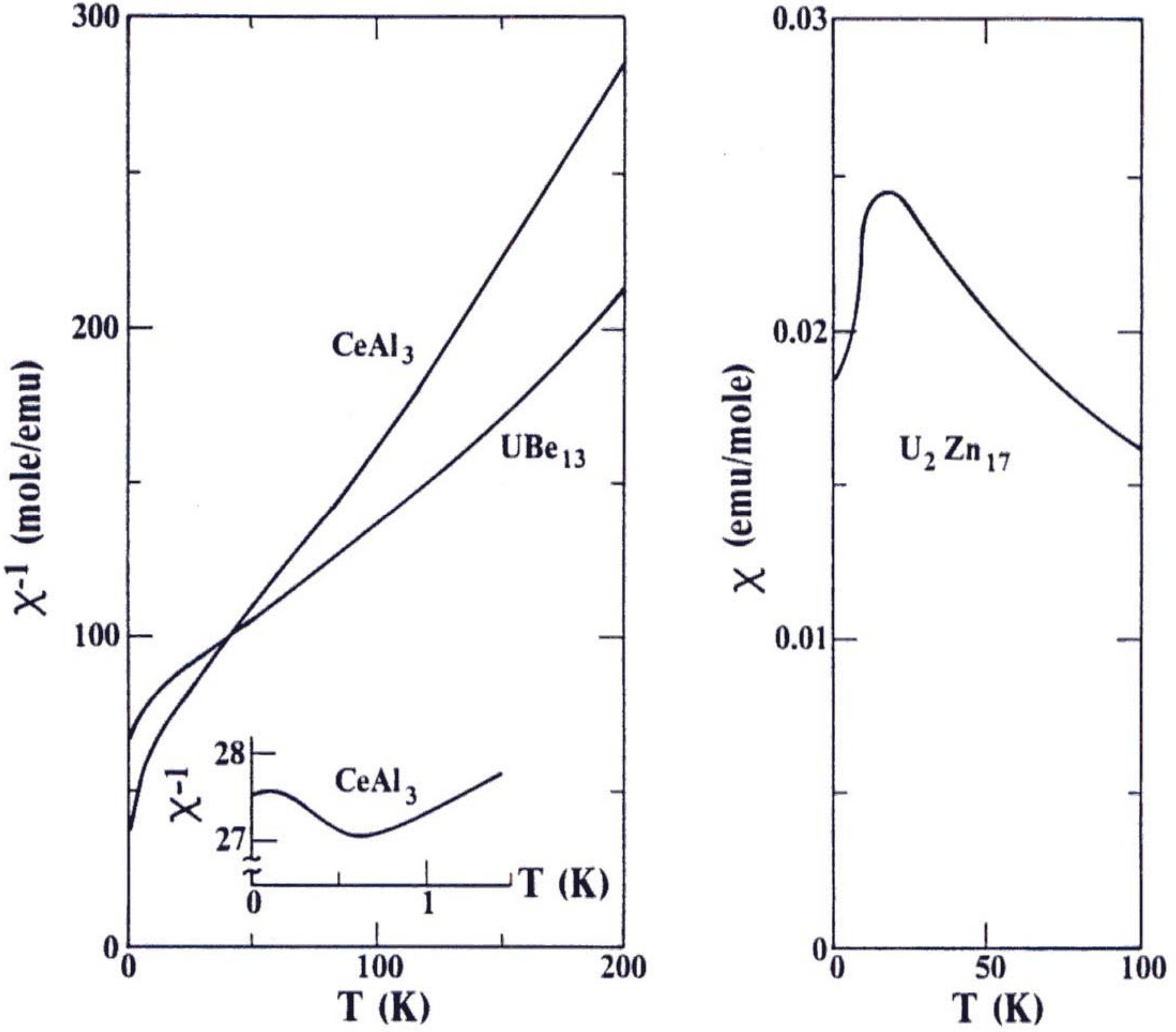
Heavy fermion superconductors

Electrical resistivity of selected heavy fermion compounds



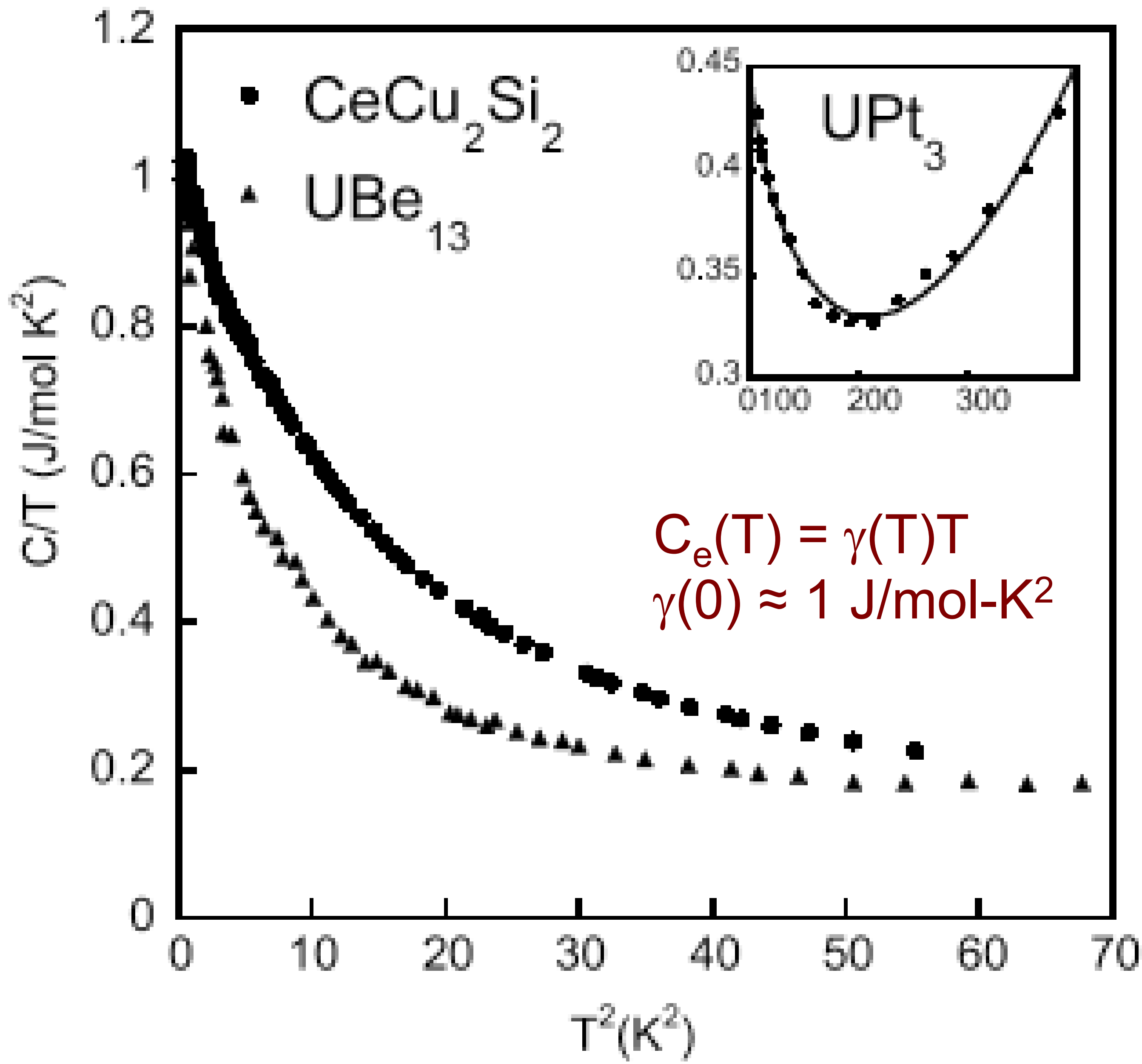
After Fisk, Ott, Rice & Smith 86

Magnetic susceptibility of selected heavy fermion compounds



After Fisk, Ott, Rice & Smith 86

$C/T = \gamma$ vs T^2 for $CeCu_2Si_2$, UBe_{13}, and UPt_3



$C_e(T) = \gamma(T)T$

$\gamma(0) \approx 1$ J/mol-K^2

Fermi liquid aspects of heavy fermion metals

Wilson-Sommerfeld ratio R:

$R = (\chi/\gamma)(\pi^2 k_B^2/\mu_{eff}^2)$; $\mu_{eff}^2 = g_J^2 \mu_B^2 J(J+1)$

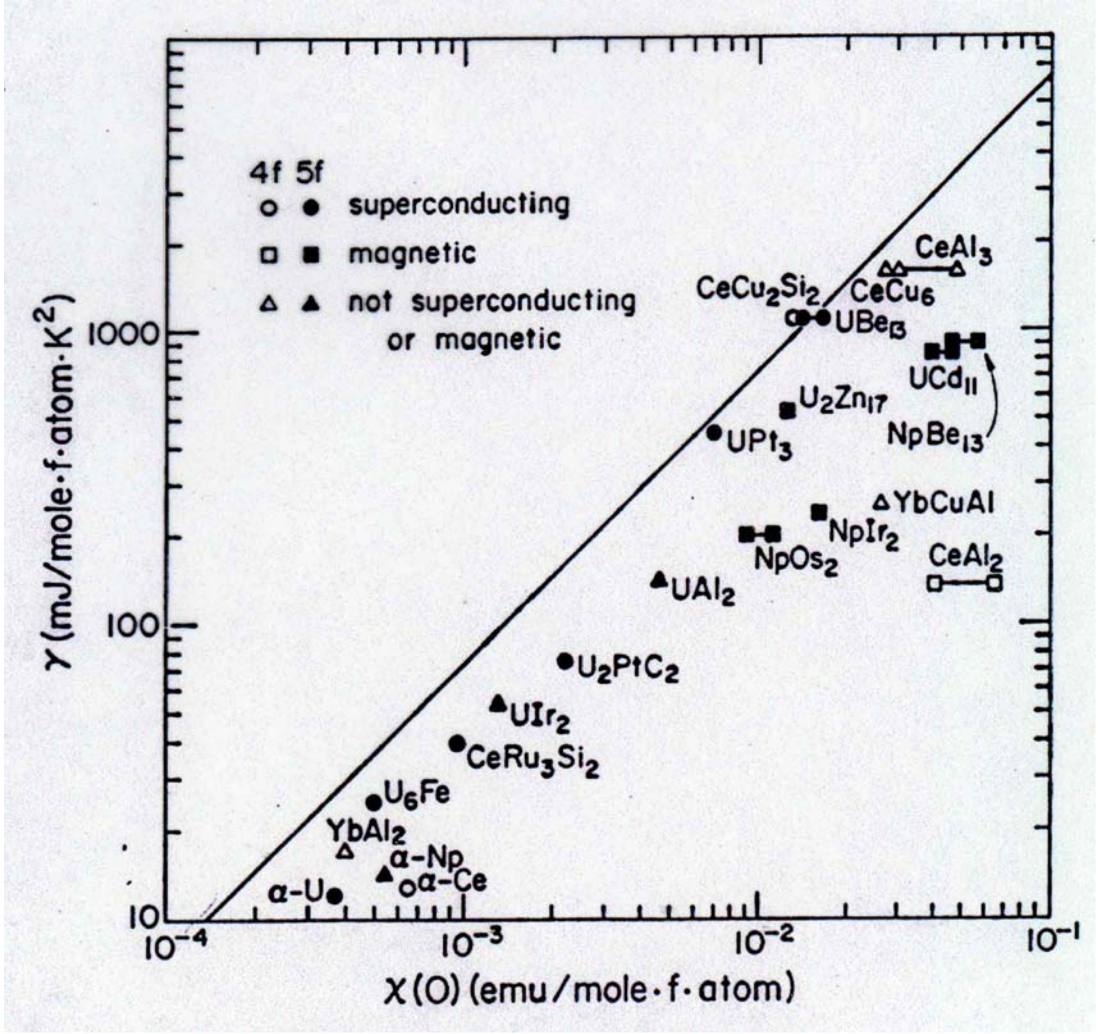
R ≈ 1 for free electrons

Fermi liquid (Landau)

$\gamma = C/T \sim m^* \sim 1/T^*$

$\chi \sim m^*/(1+F_o^a)$

$\Delta\rho = AT^2 \sim (T/T^*)^2 \sim \gamma^2 T^2$



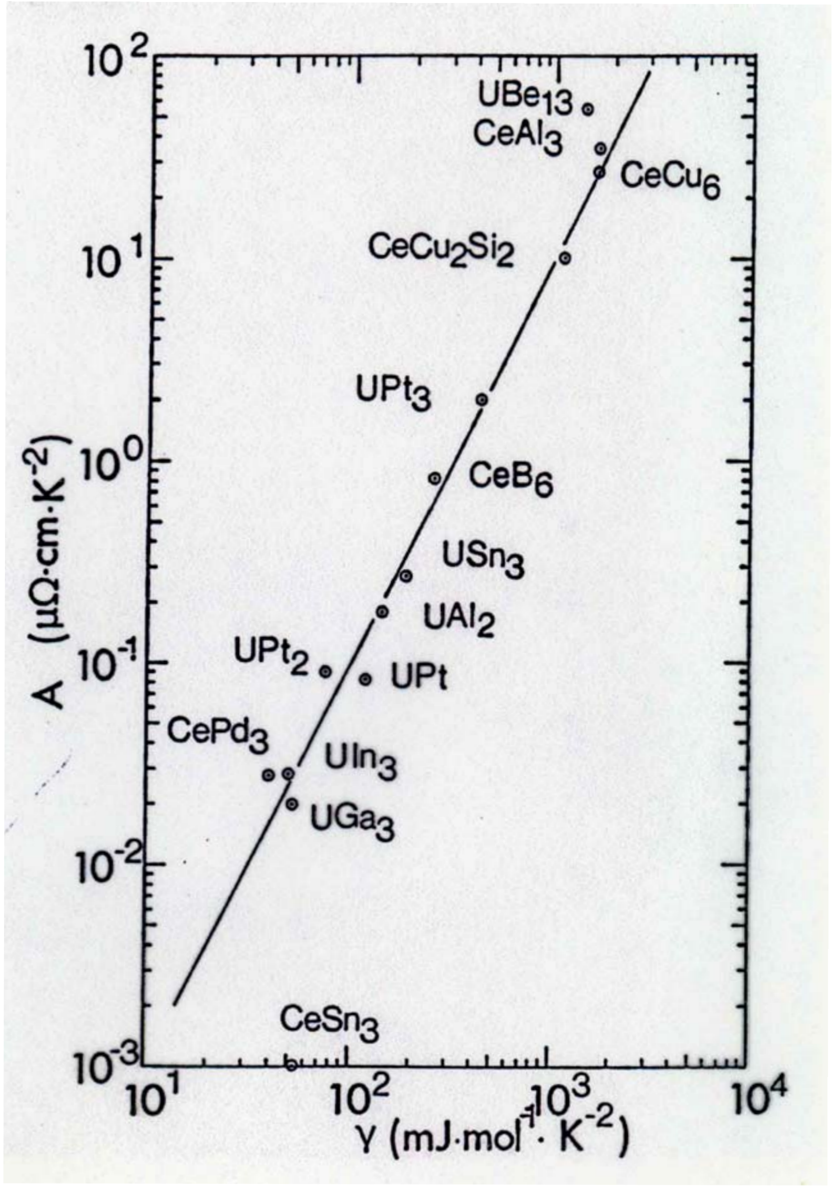
Electron-electron scattering

Heavy quasiparticles:

$\rho_{el\text{-}el}(T) \sim AT^2 \sim (T/T^*)^2 \sim \gamma^2 T^2$

$\Rightarrow A \sim \gamma^2,\ \ln A \propto 2\ln\gamma$

Kadowaki-Woods plot



Heavy fermion superconductors

		T_c (K)
	$CeCoIn_5$	2.3
*	$CeCu_2Si_2$	0.49
	$CeIrIn_5$	0.4
	U_6Fe	3.7
*	UPd_2Al_3	2.0
*	URu_2Si_2	1.5
*	UNi_2Al_3	1.0
	UBe_{13}	0.85
*	UPt_3	0.55
*	URhGe	0.4
	$PuCoGa_5$	18
	$PrOs_4Sb_{12}$	1.8

* Magnetic order

Heavy fermion superconductors

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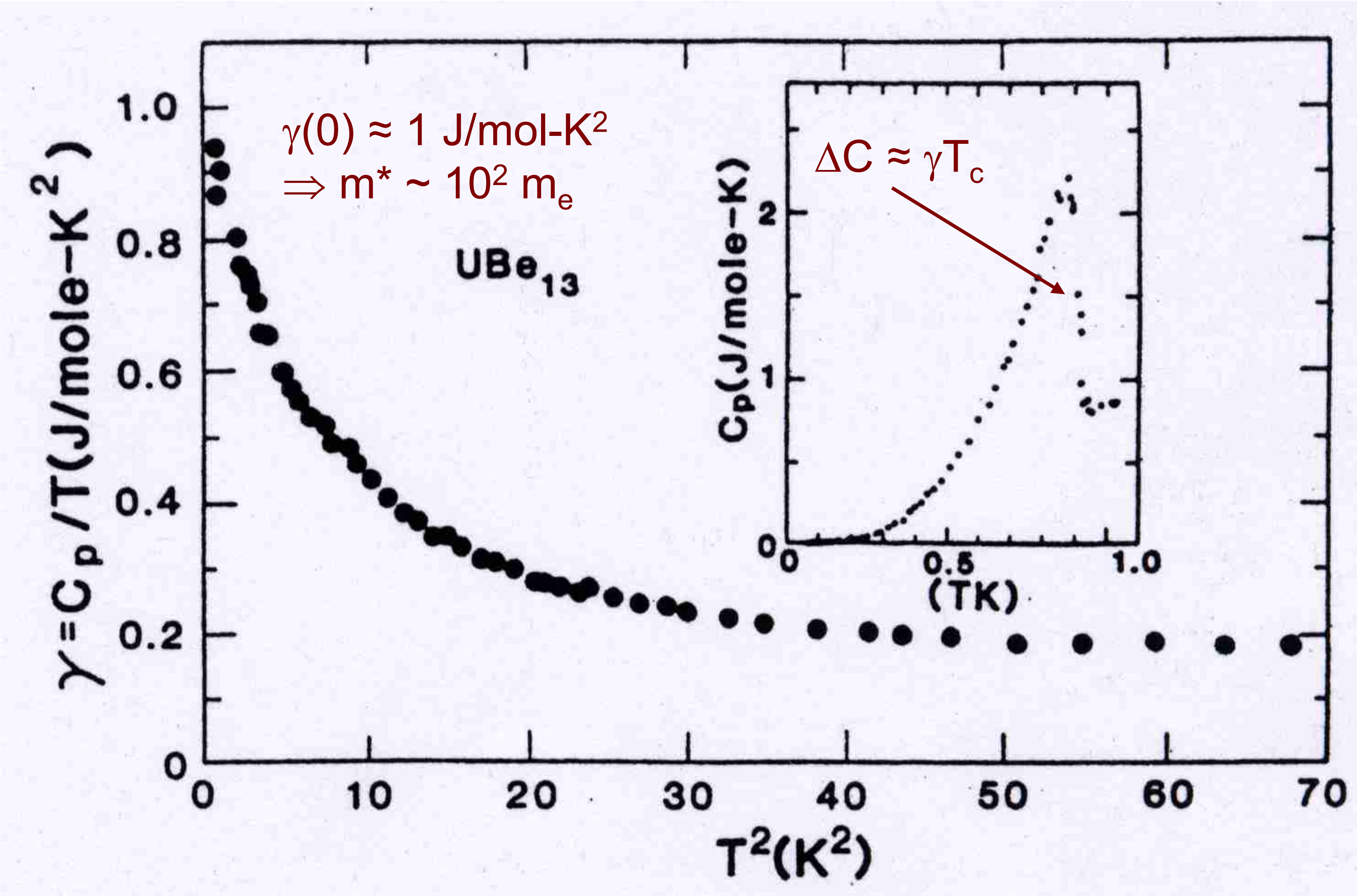
Superconducting under pressure:

		T_c (K)	P (kbar)
*	$CeRhIn_5$	2.2	21
*	Ce_2RhIn_8	2	23
*	$CeCu_2Ge_2$	~2	165
*	$CePd_2Si_2$	0.43	28
*	$CeRh_2Si_2$	0.26	11
	$CeNi_2Ge_2$	0.23	23
*	$CeIn_3$	0.17	25
*	UGe_2	0.7	10

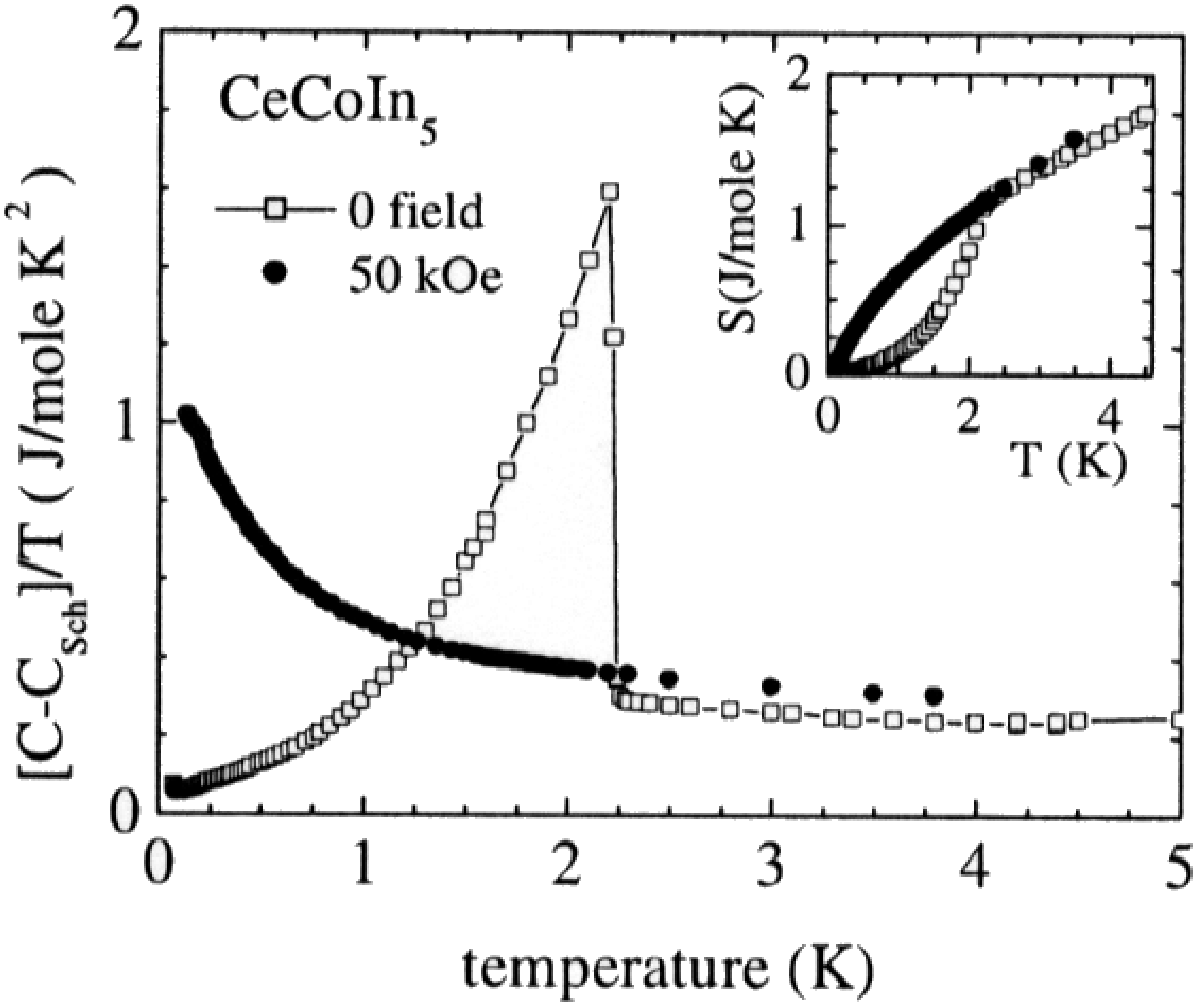
* *Magnetic order*

C/T ≡ γ vs T^2 for UBe_{13}



Origin: Kondo effect

$CeCoIn_5$: heavy fermion superconductor

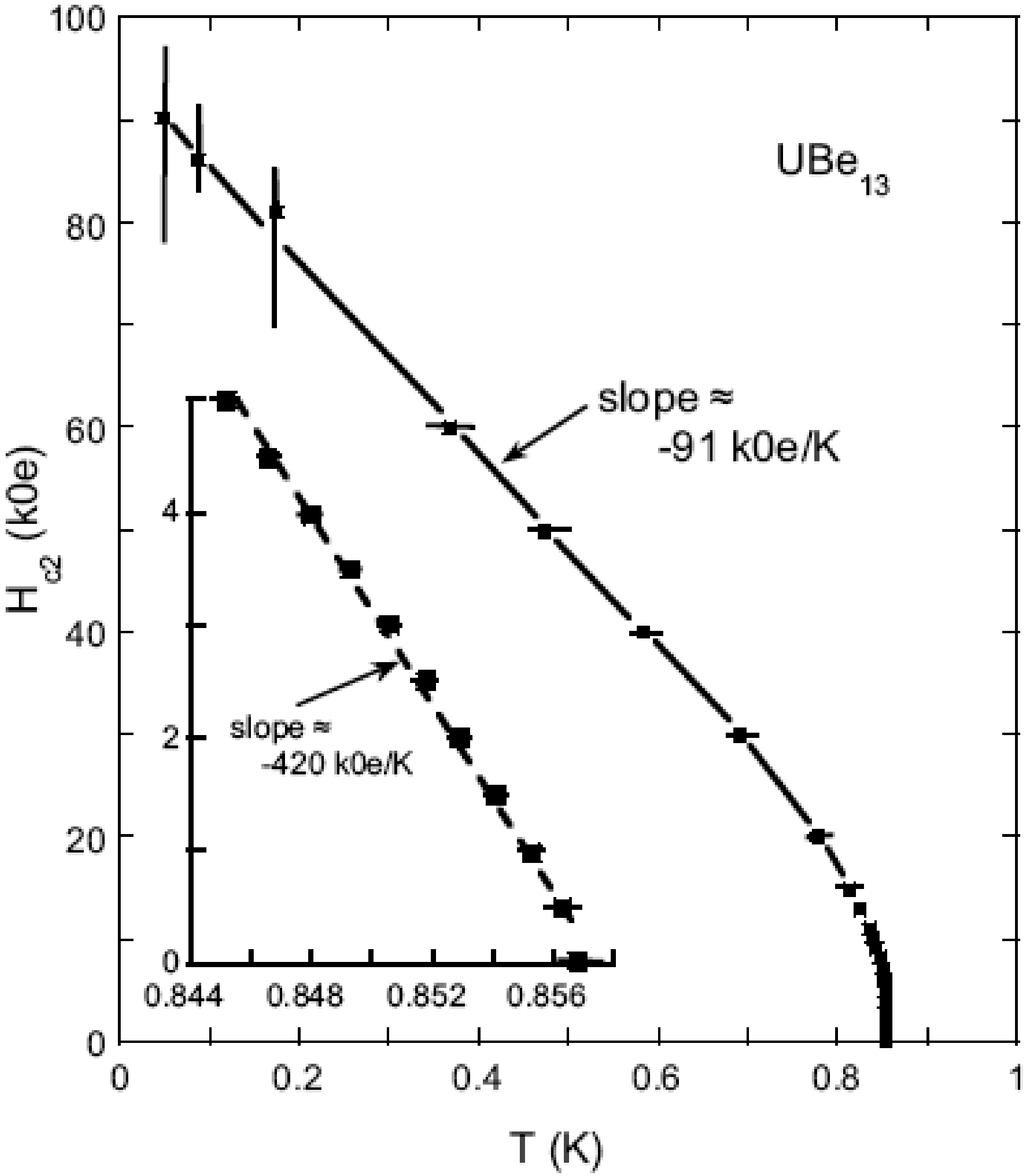


H = 0:
Superconductivity

H = 50 kOe:
NFL behavior

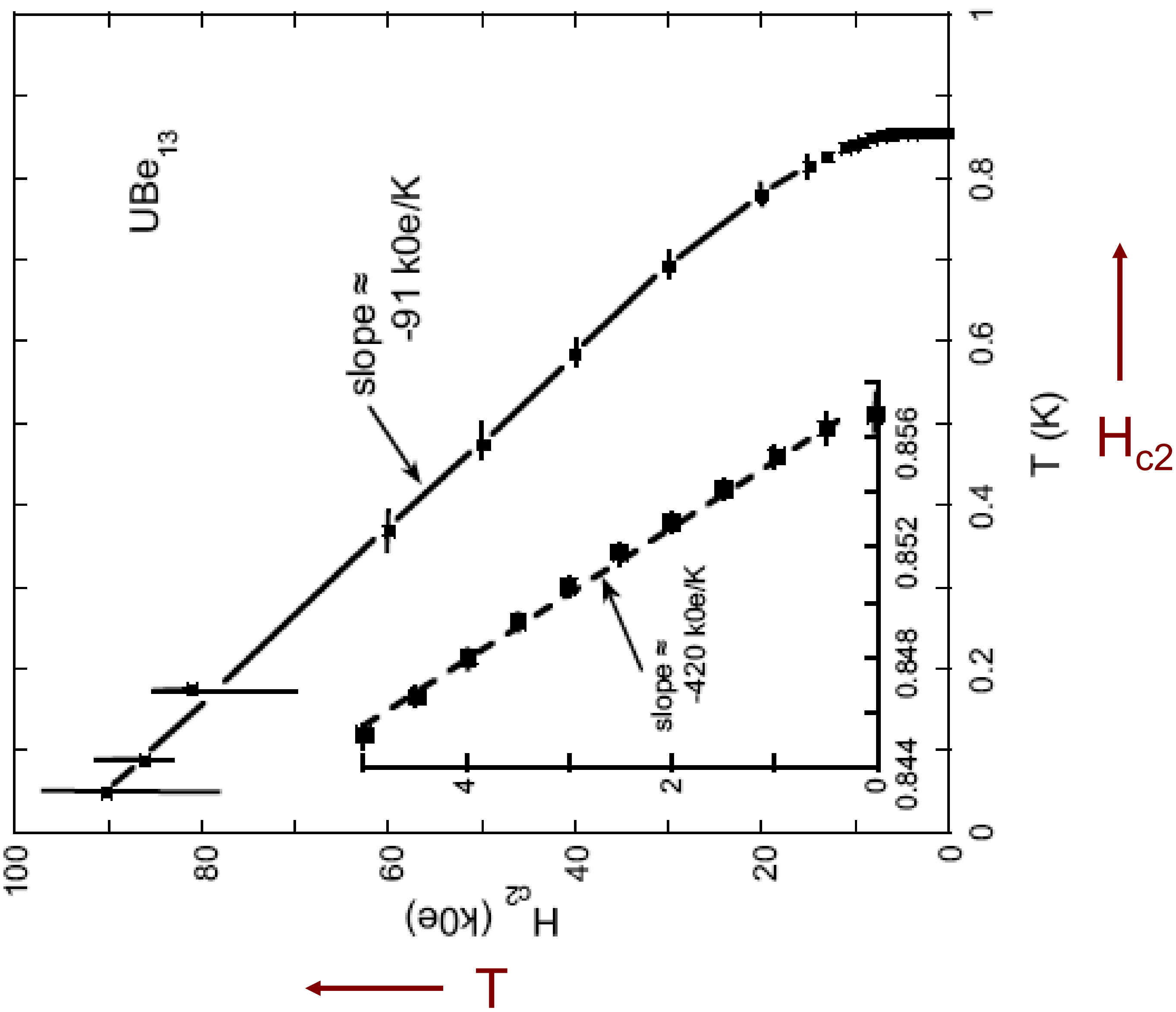
C Petrovic et al., J. Phys.: Condens. Matter ***13*** *(2001) L337*

Upper critical field $H_{c2}(T)$ of UBe_{13}

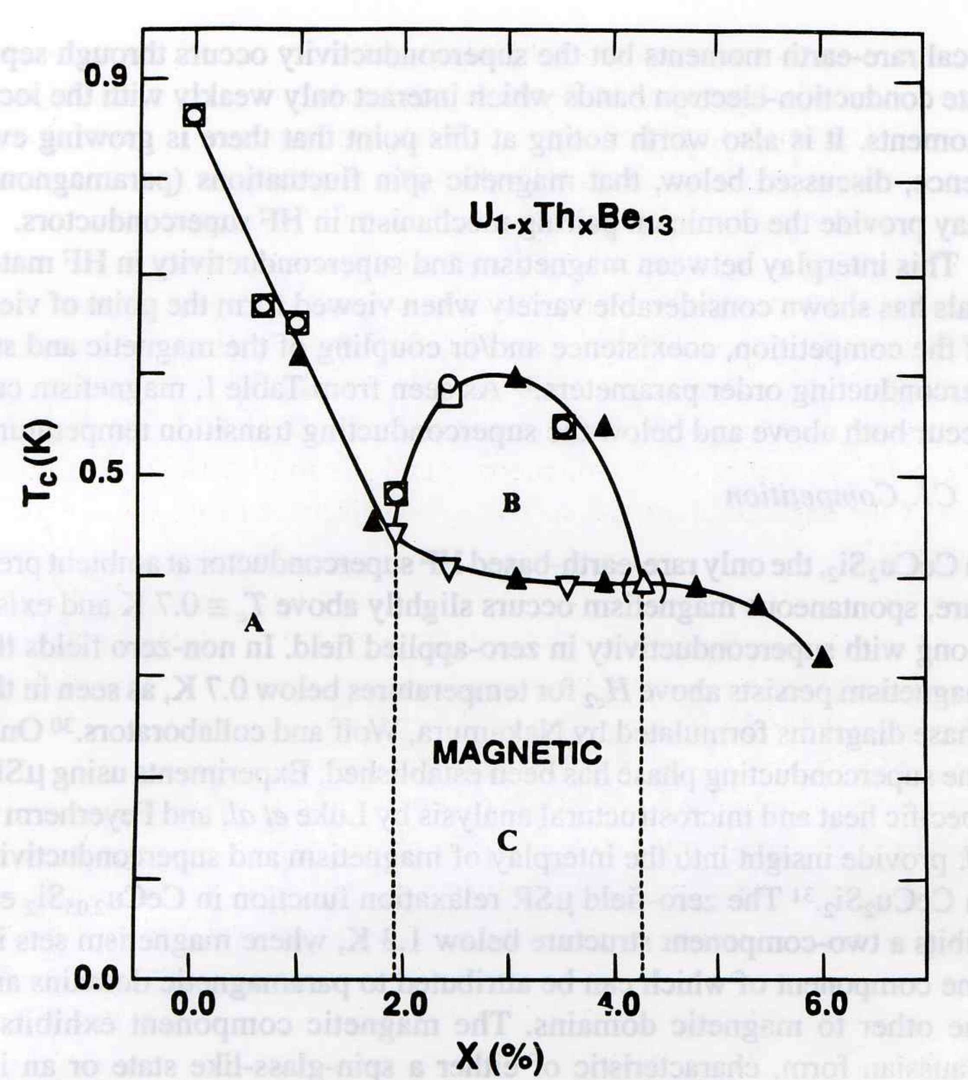


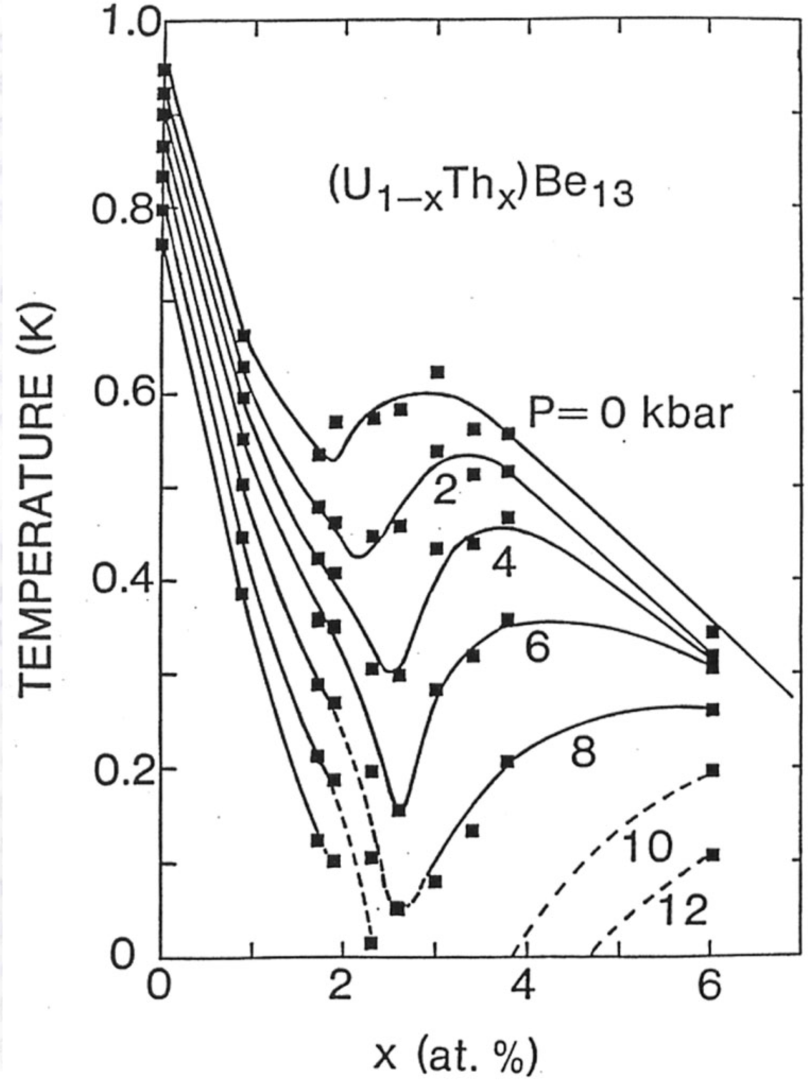
M. B. Maple et al. (85)

$H_{c2}(T)$ for conventional superconductor



Multiple superconducting phases in $U_{1-x}Th_xBe_{13}$

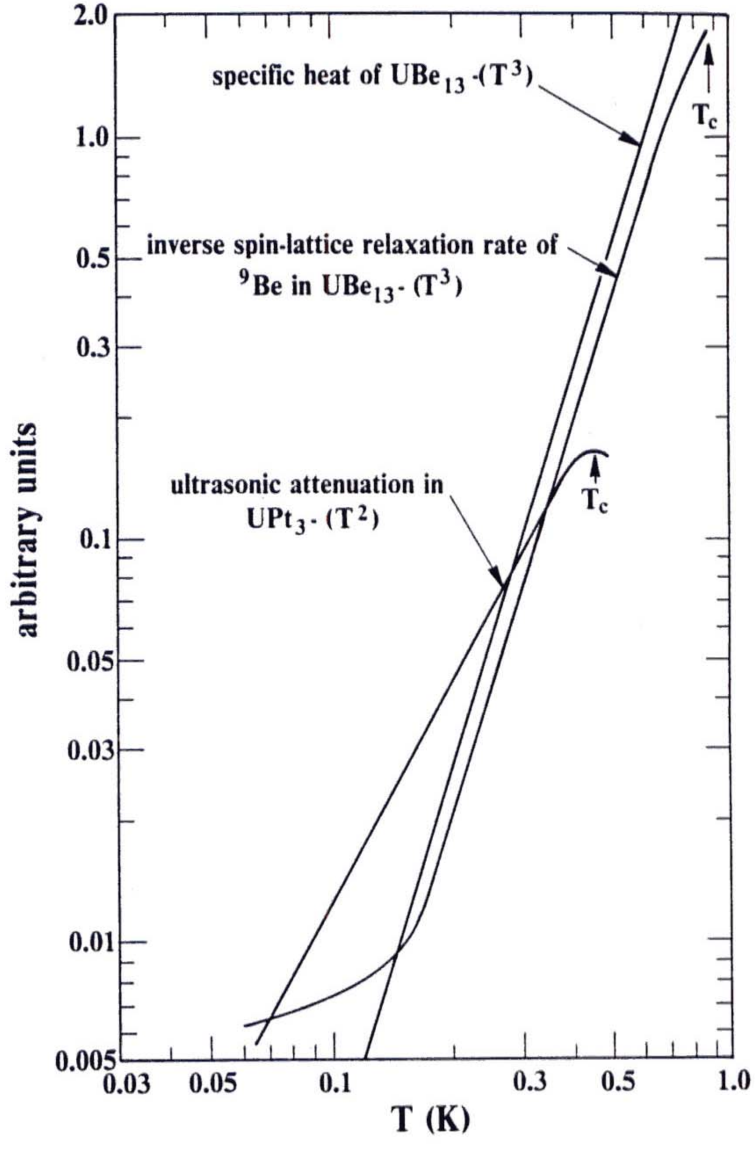




*R. H. Heffner et al., PRL **65** (90)*

*S. E. Lambert et al., PRL **57** (86)*

Power law T-dependence of superconducting properties of heavy fermion superconductors UBe_{13} and UPt_3

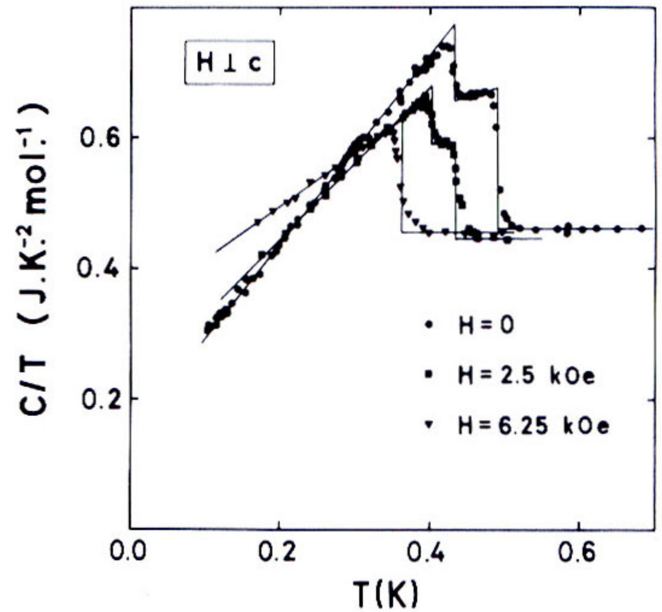


After Fisk, Ott, Rice & Smith 86

Multiple superconducting phases in UPt_3

Two distinct SCing transitions (sensitive to H & P)

Hasselbach, Taillefer, Flouquet 89



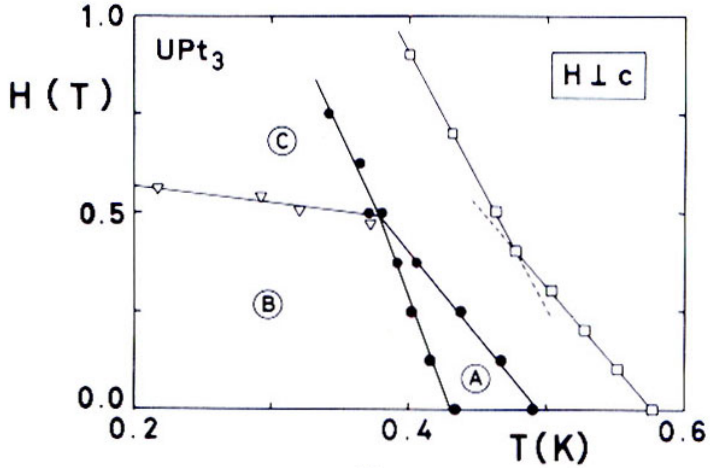
(a)

Coupling between multicomponent SCing OP & AFM OP

AFM: $T_N \approx 5$ K

$\mu \approx 0.02\ \mu_B$/U (basal plane)

Aeppli et al. 88



(b)

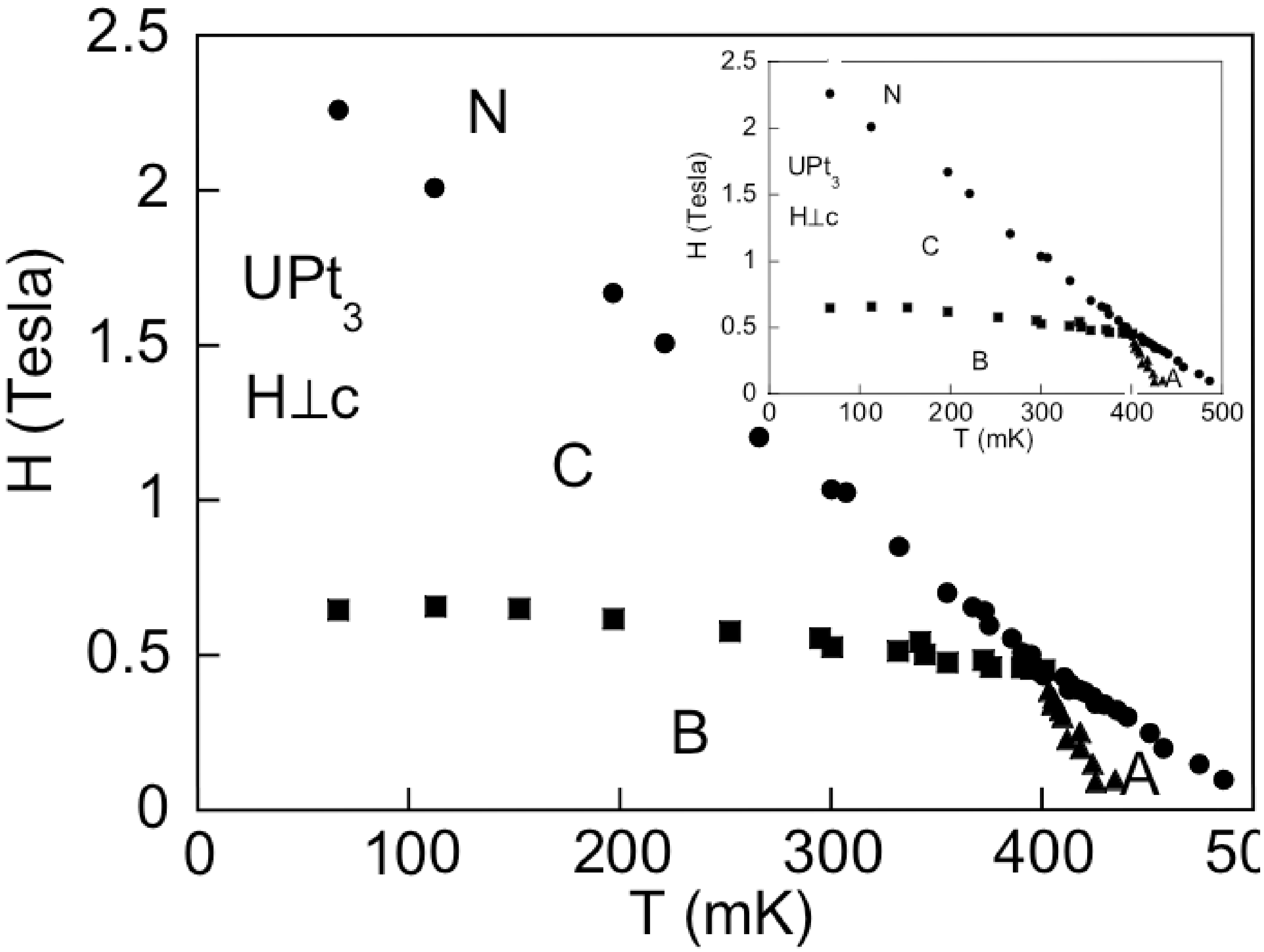
H – T phase diagram of UPt_3

Ultrasonic velocity measurements

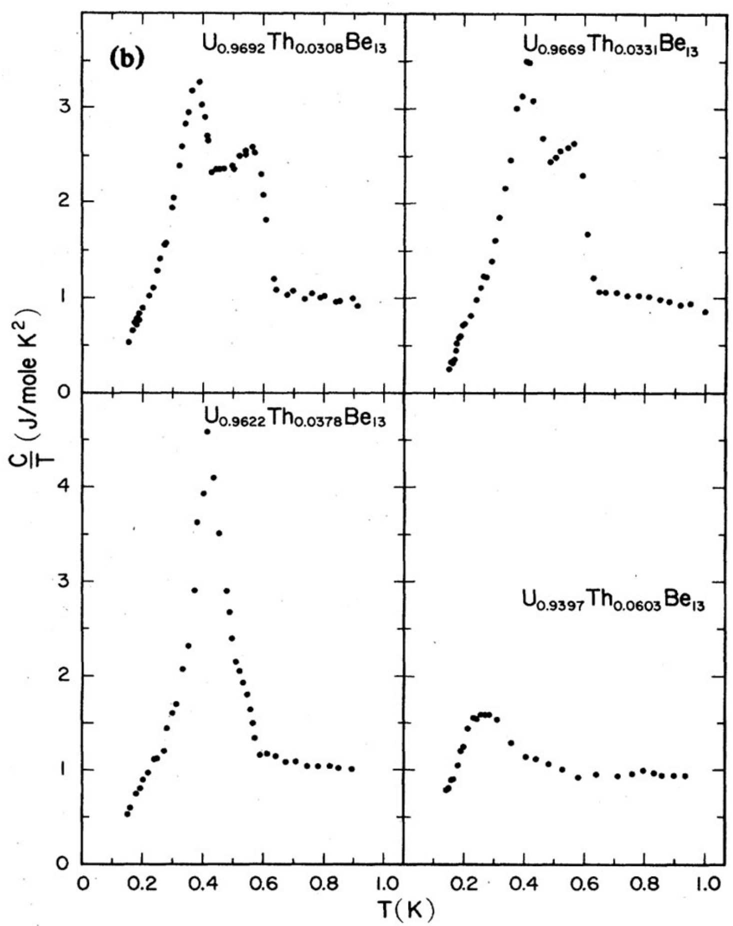
Adenwalla, Ketterson, Yip, Lin, Levy, Sarma 92

B-phase: odd-parity, spin-triplet SCing state

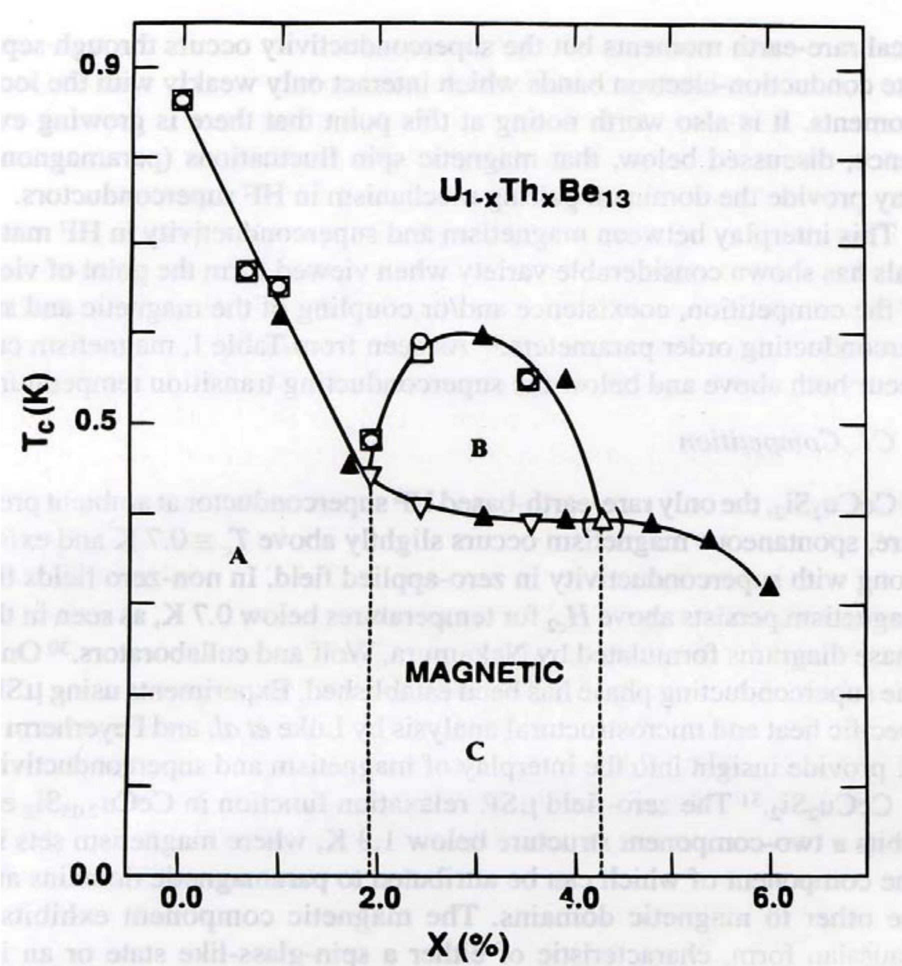
Sauls '94



Multiple superconducting phases in $U_{1-x}Th_xBe_{13}$

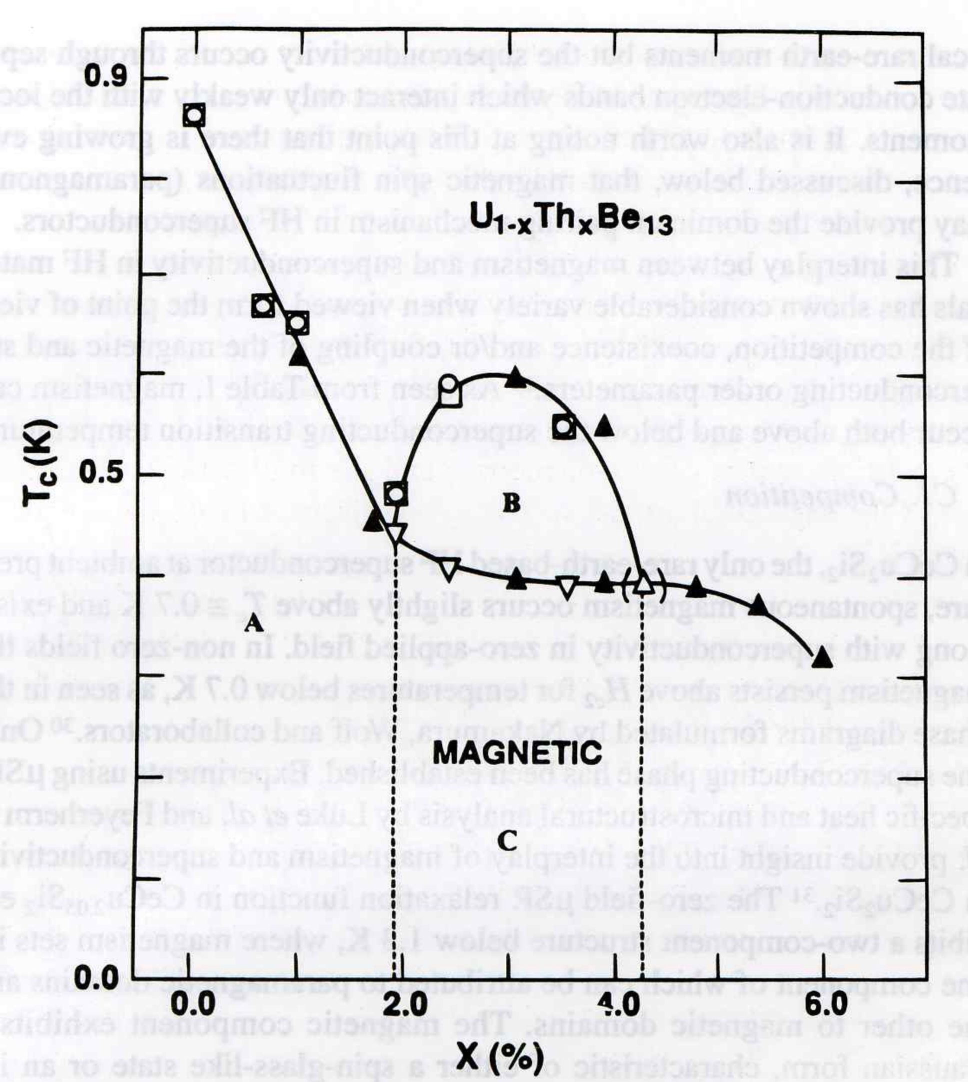


*H. R. Ott et al. PRB **31** '85*

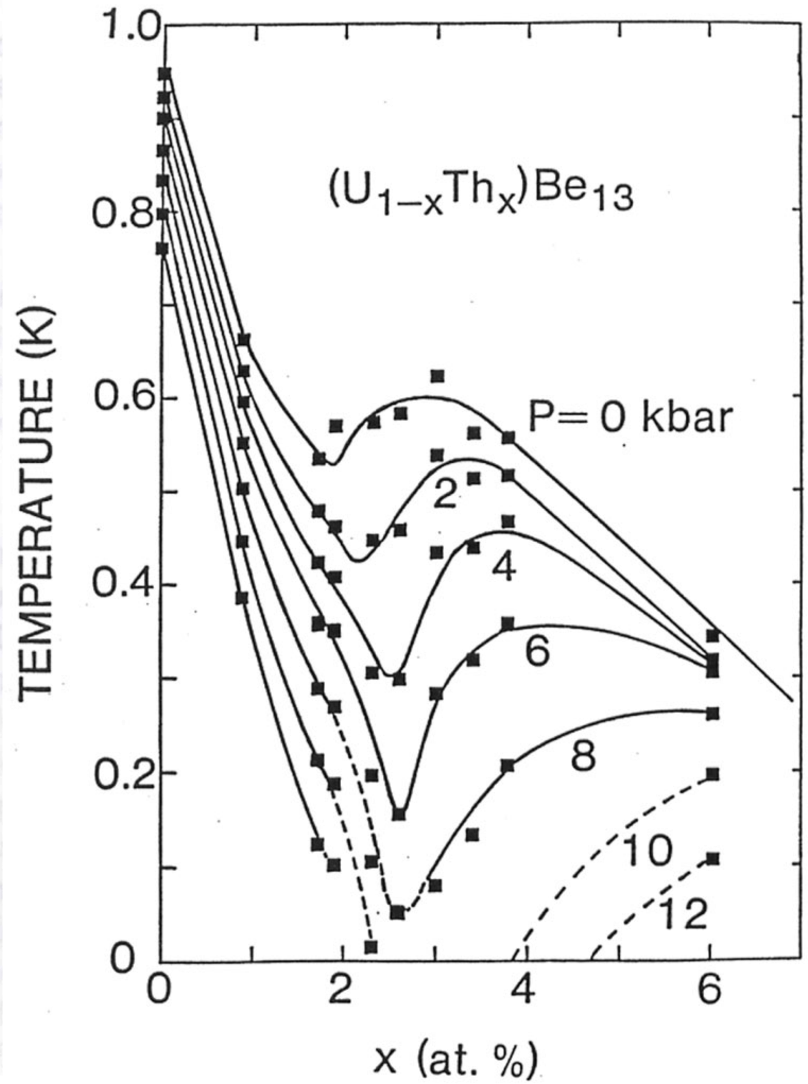


*R. H. Heffner et al. PRL **65** '90*

Multiple superconducting phases in $U_{1-x}Th_xBe_{13}$

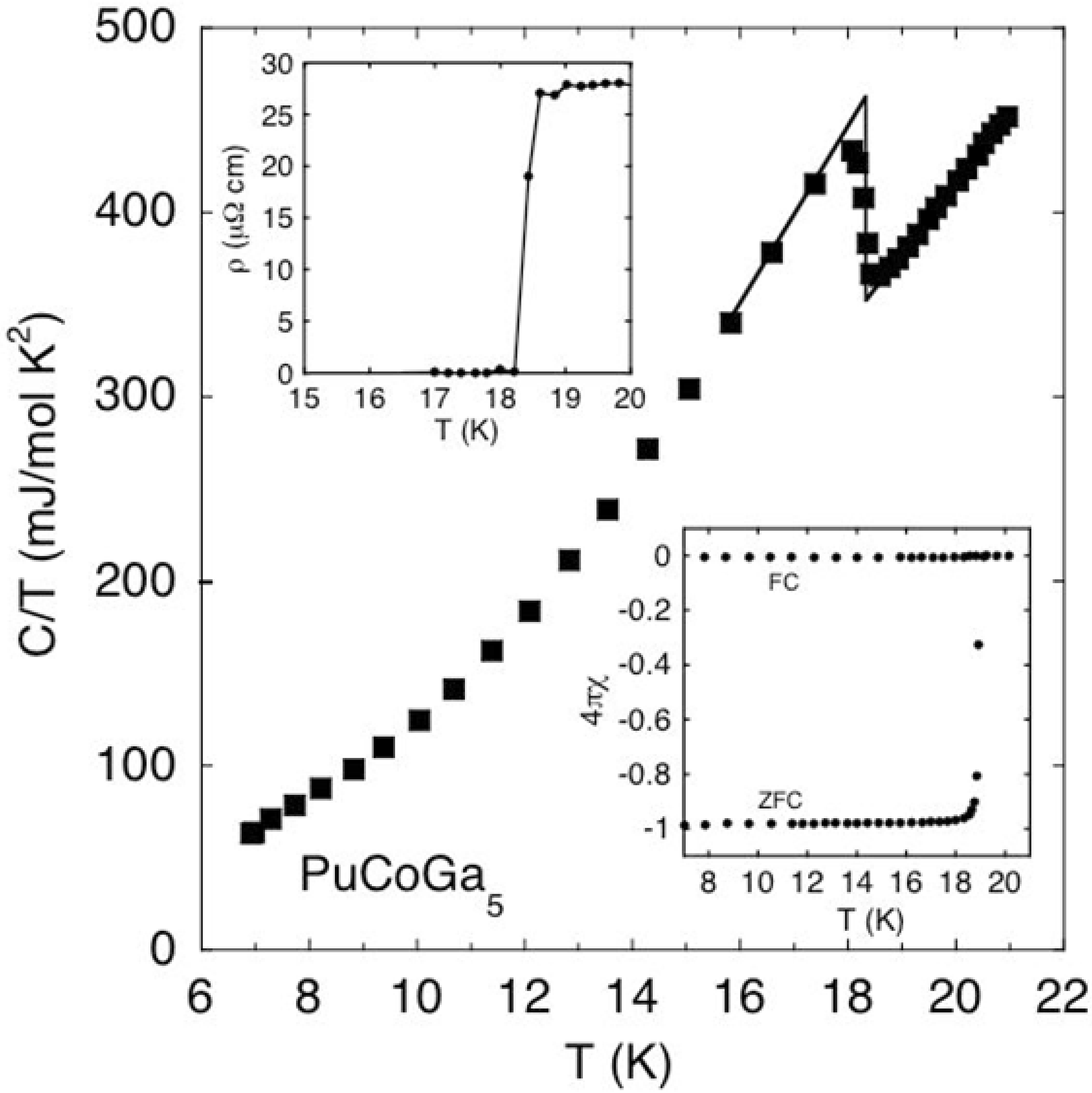


*R. H. Heffner et al. PRL **65** '90*



*S. E. Lambert et al. PRL **57** '86*

High temperature superconductivity in $PuCoGa_5$



Sarrao et al. '02

Non-Fermi liquid behavior in f-electron systems

Non-Fermi liquid behavior in $M_{1-x}U_xPd_3$ (M = Y, Sc) systems

$Y_{1-x}U_xPd_3$: First f-electron system in which NFL behavior observed

*C. L. Seaman, M. B. Maple, B. W. Lee, S. Ghamaty, M. S. Torikachvili, K. N. Yang, L. Z. Liu, J. W. Allen, D. L. Cox, PRL **67**, 2882 '91*

$\rho(T,H)$, $C(T)$, $M(T,H)$ for $0 < x \leq 0.2$

Interpreted NFL behavior in terms of quadrupolar Kondo effect

*B. Andraka, A. M. Tsvelik, PRL **67**, 2886 '91*

$\rho(T,H)$, $C(T,H)$, $M(T,H)$ for $x = 0.2$

Interpreted NFL behavior in terms of OP fluctuations associated with magnetic transition suppressed to 0 K

Extensive investigations $\Rightarrow$ situation much more complex

INS studies: U^{4+} energy level scheme in cubic CEF not yet established

Fluctuations in U concentration x on ~10 μm scale (*S. Süllow et al. '91*)

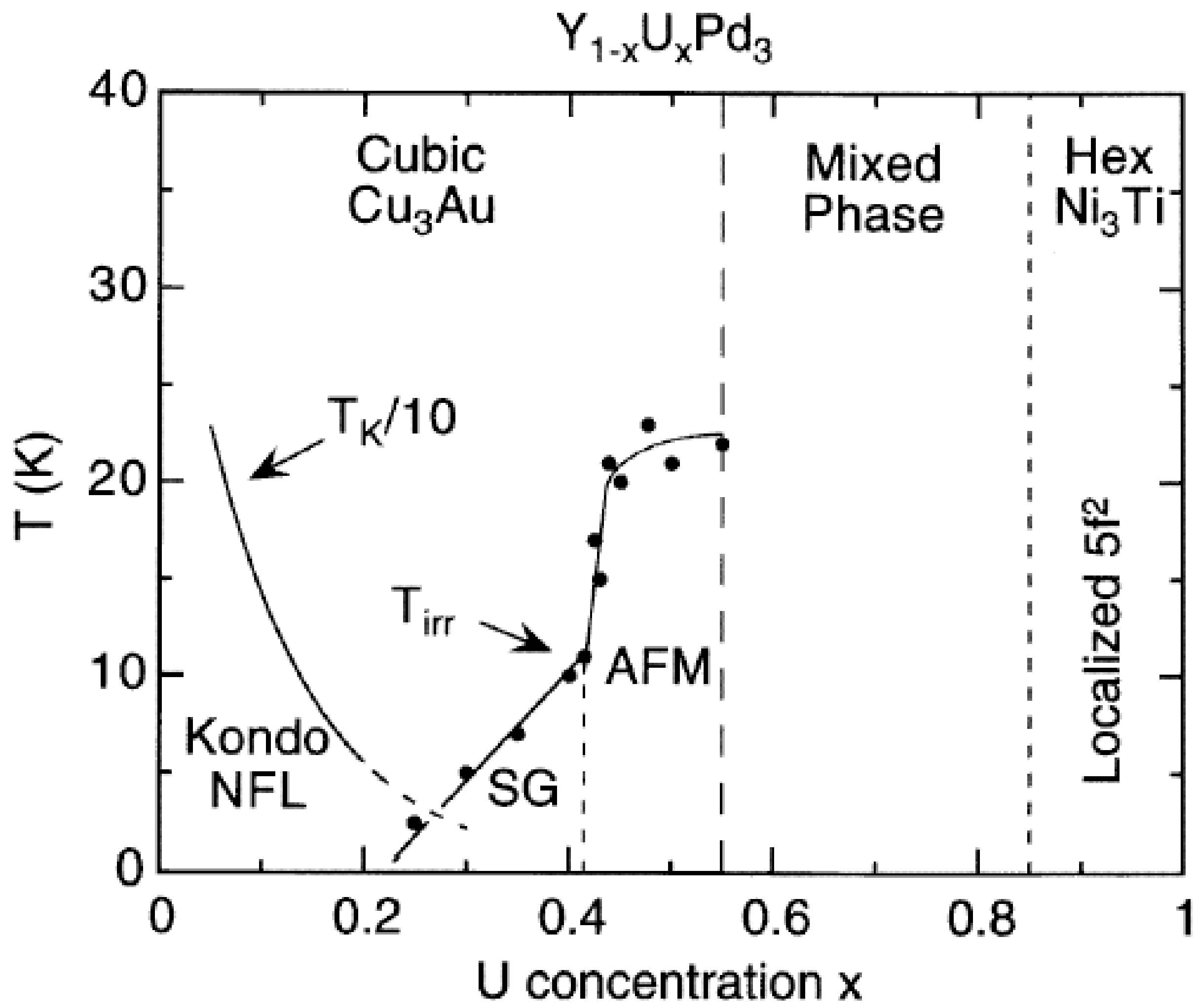
$Sc_{1-x}U_xPd_3$: NFL behavior similar to that observed in $Y_{1-x}U_xPd_3$ system

More homogeneous distribution of U

INS studies: ω/T scaling of $\chi''(\omega,T)$

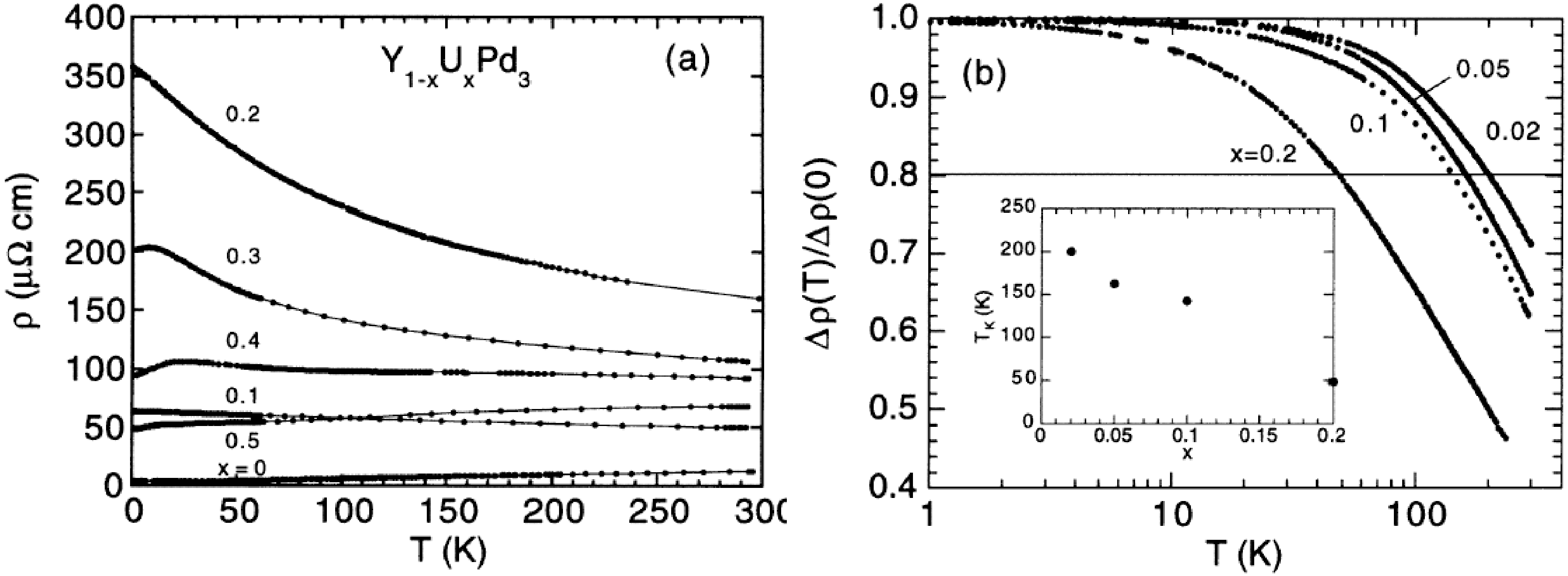
Interplay between Kondo effect, disorder, AFM interactions

T-x phase diagram of $Y_{1-x}U_xPd_3$ system



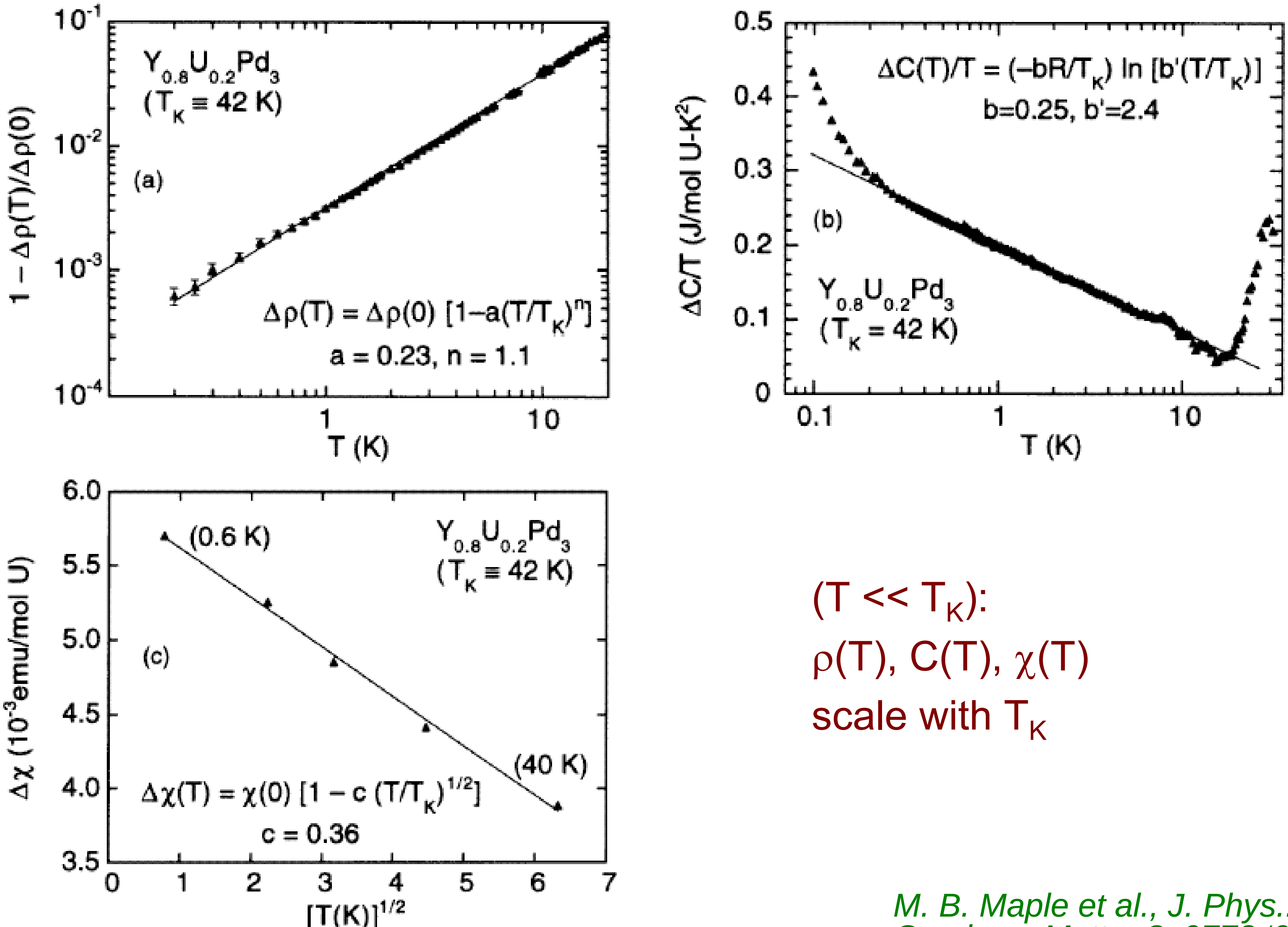
*C. L. Seaman et al., PRL **67**, 2882 ‘91*
*D. A. Gajewski, N. R. Dilley, R. Chau, M. B. Maple, J. Phys.: Condens. Matter **8**, 9793 ‘96*

Electrical resistivity ρ vs T for the $Y_{1-x}U_xPd_3$ system (high T)



M. B. Maple, R. P. Dickey, J. Herrmann, M. C. de Andrade, E. J. Freeman, D. A. Gajewski, R. Chau, J. Phys.: Condens. Matter ***8****, 9773 '96*

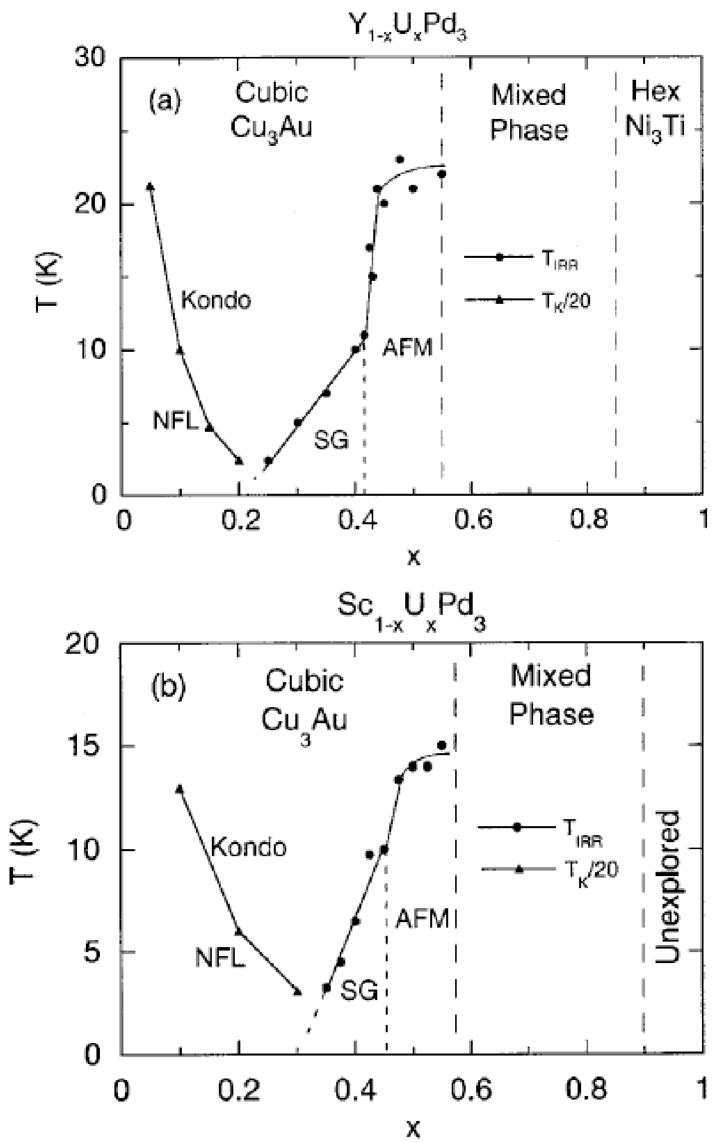
Low-T NFL behavior in ρ(T), C(T), χ(T) for $Y_{1-x}U_xPd_3$



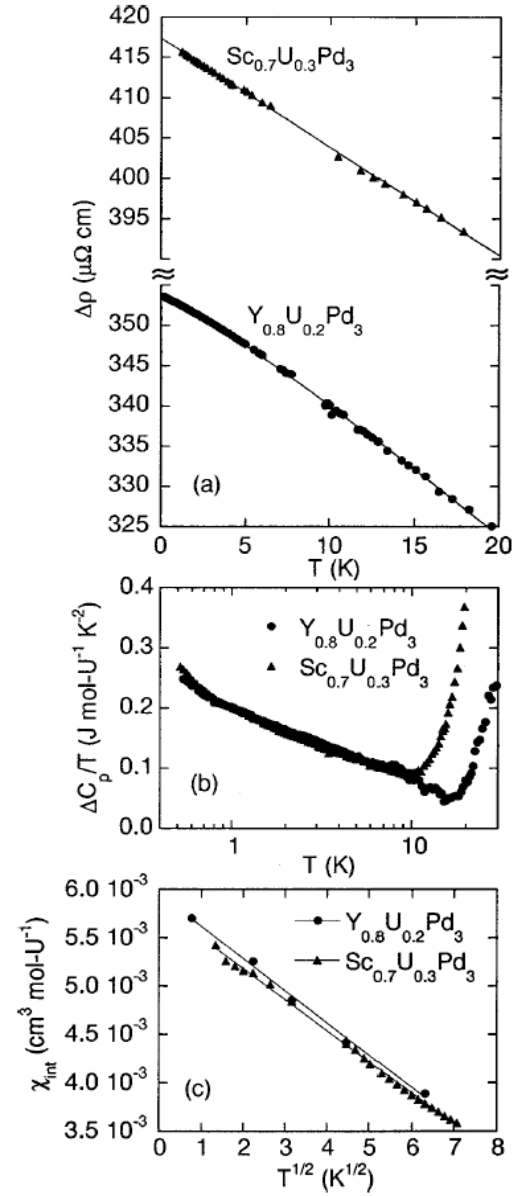
($T \ll T_K$):
ρ(T), C(T), χ(T)
scale with T_K

M. B. Maple et al., J. Phys.: Condens. Matter ***8****, 9773 '96*

T-x phase diagrams & low T properties of $M_{1-x}U_xPd_3$ (M = Y,Sc)

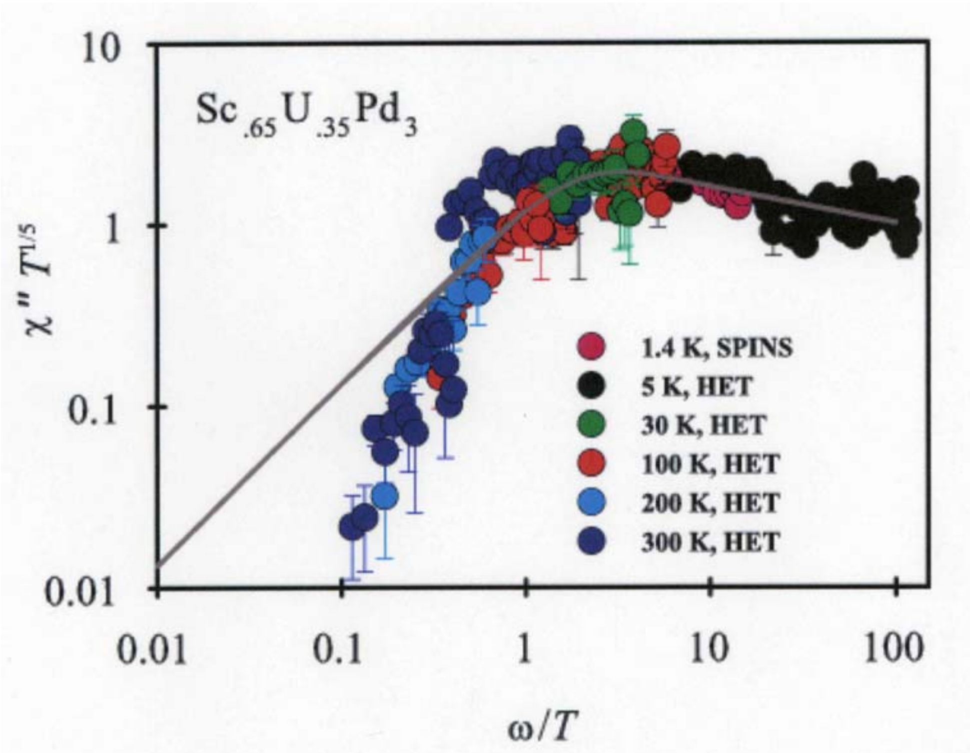


- Similar T-x phase diagrams & low T NFL characteristics in ρ(T), C(T)/T, χ(T)
- T_K decreases with x
- AFM & SG phases
- SG QCP
  M=Y: $x_c \approx 0.2$
  M=Sc: $x_c \approx 0.3$
- NFL behavior associated with SG QCP



*D. A. Gajewski, R. Chau, M. B. Maple, PRB **62**, 5496 '00*

Inelastic neutron scattering on $Sc_{1.65}U_{0.35}Pd_3$: ω/T scaling



$\chi''(q,\omega,T) = 1/[AT^{\alpha}F(\omega/T)]$
with $\alpha = 1/5$

No q-dependence $\Rightarrow$ no evidence for U-U correlations

Single impurity critical scaling associated with spin glass transition suppressed to 0 K

Solid line: $F(\omega/T) = \exp[\alpha\Psi(1/2 - i\omega/2\pi T)]$
Proposed for AFM QCP — *Q. Si et al., Nature **413**, 804 '01*

S. D. Wilson, P. Dai, D. T. Androja, S.-H. Lee, J.-H. Chung, J. W. Lynn, N. P. Butch, M. B. Maple, PRL '05 (in press)

Inelastic neutron scattering on $Sc_{1.65}U_{0.35}Pd_3$: ω/T scaling

Similar behavior: $\chi''(q,\omega,T) \propto 1/AT^{\alpha}F(\omega/T)$

$UCu_{5-x}Pd_x$ (x = 1, 1.5); $\alpha = 1/3$

Near AFM QCP

*M. Aronson et al., PRL **75**, 725 '95; PRL **87**,197205 '01*

$CeCu_{5.9}Au_{0.1}$; $\alpha = 0.75$

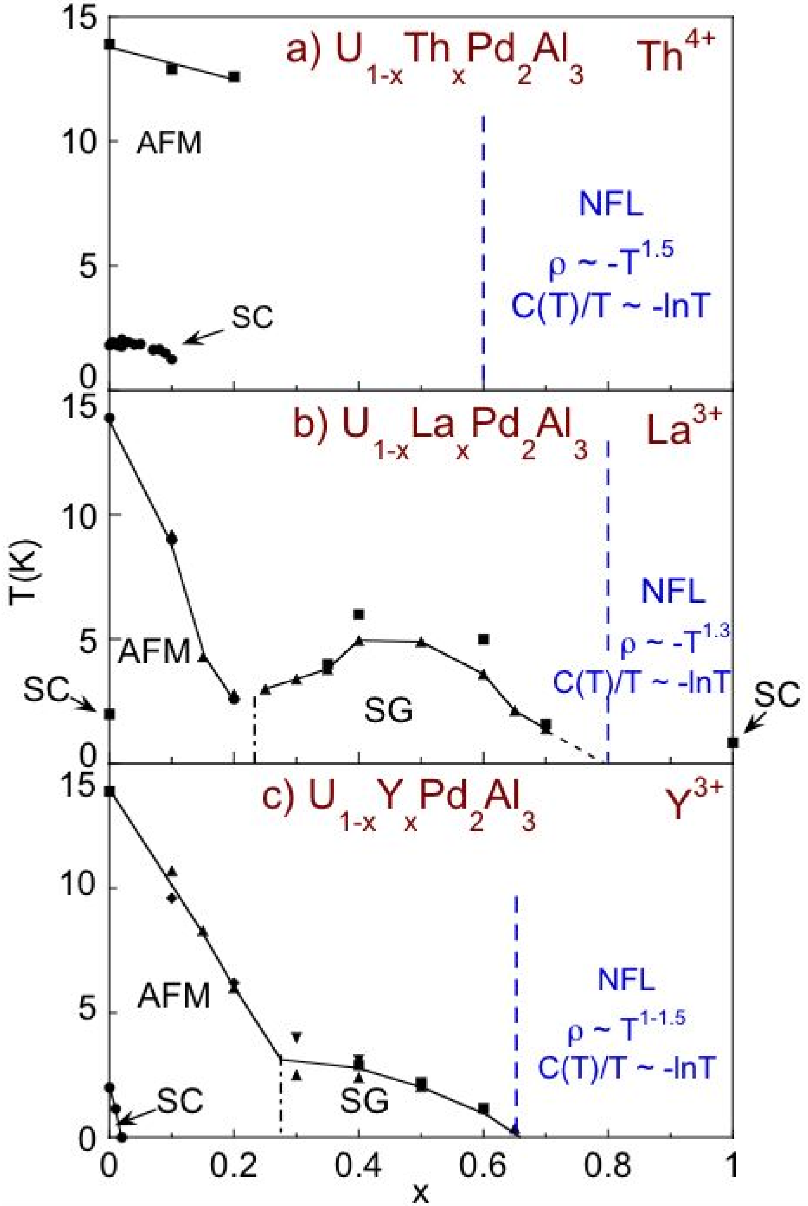
*A. Schröder et al., PRL **80**, 5623 '98*

Near AFM QCP

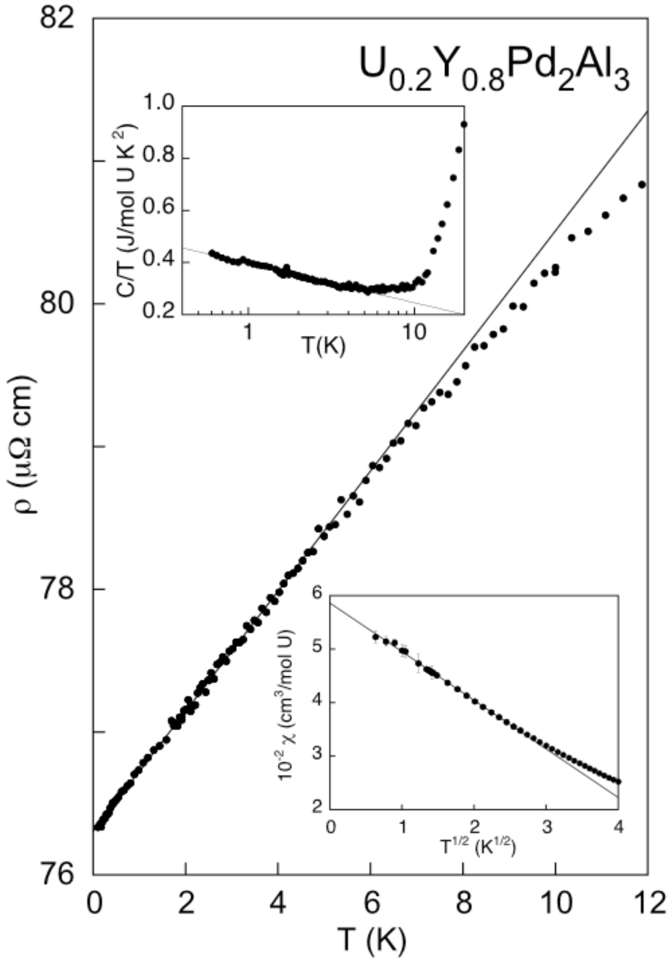
Absence of energy scale, other than T itself
Microscopic origin of the NFL behavior involves individual ions near 0 K spin glass phase transition

Although impurities may play role, they do not appear to be primary cause

T-x phase diagrams for $U_{1-x}M_xPd_2Al_3$ (M = Th, Y, La)



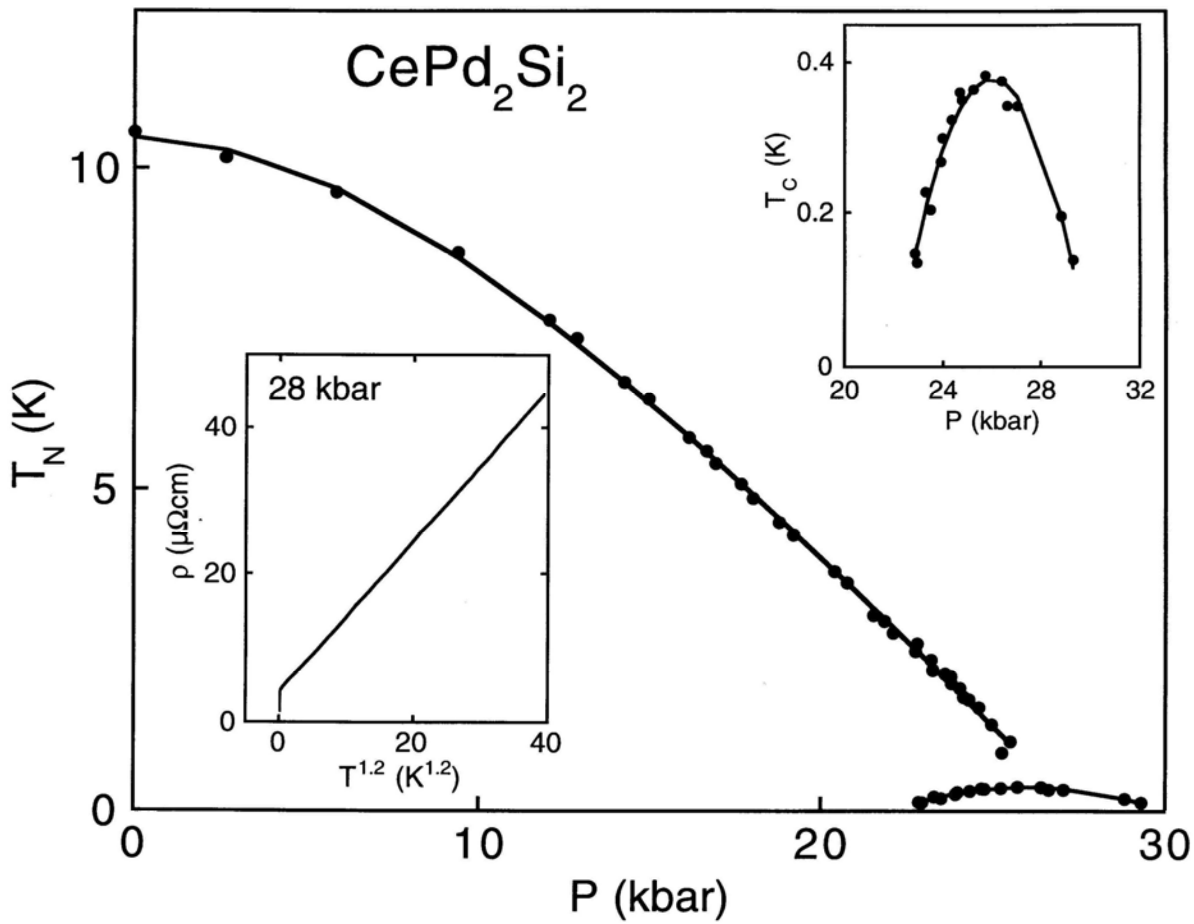
NFL behavior in $U_{1-x}Y_xPd_2Al_3$ near QCP at $x_c \approx 0.65$



UCSD '01

Superconductivity near quantum critical points

Superconductivity near pressure-induced AFM QCP



AFM QCP:
$P_c \approx 28$ kbar
$\rho(T) \approx \rho_o + AT^{1.2}$
$T_c \leq T \leq 40$ K
$T_c(\max) \approx 0.4$ K
Similar behavior for $CeIn_3$ under P

Julian, Lonzarich et al. (98)

Suggests AFM spin fluctuations responsible for NFL behavior in $\rho(T)$ and SCing electron pairing

Superconductivity within the ferromagnetic state in UGe_2

- First P-induced FM-SC

  Saxena et al. (00)

  High purity crystal (l >> ξ)

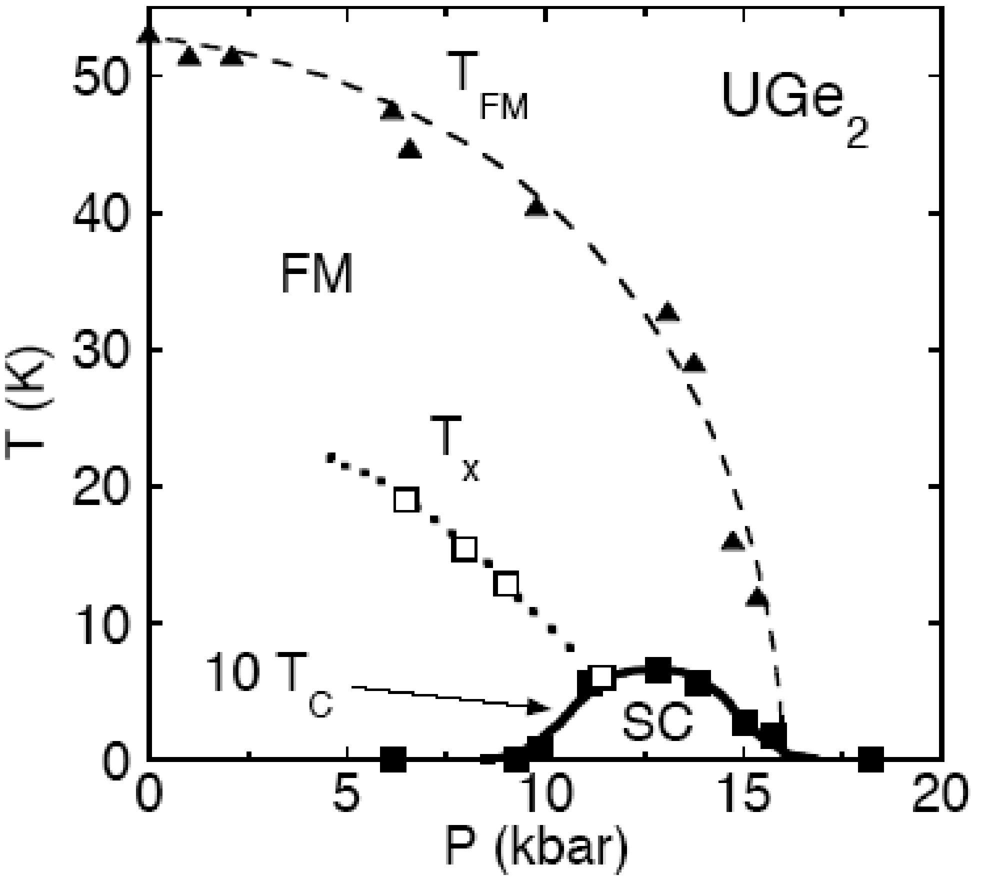
  ⇒ microscopic coexistence of triplet-spin SC & FM?
- Itinerant electron FM

  θ_C = 53 K (P = 0)
- $\gamma \approx$ 35 mJ/mol K^2

  $m^* \approx 20\ m_e$

  Onuki et al. (93)
- $\theta_C \to$ 0 K at $P_c \approx$ 16 kbar

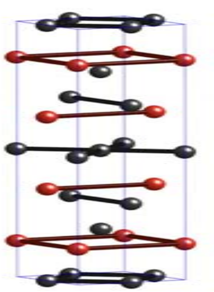
  Oomi et al. (98)



- Experiments on polycrystalline UGe_2 (l ≈ ξ)

  Inhomogeneous state: coexistence of singlet-spin SC regions & FM regions?

  Bauer, Zapf, Ho, Maple (01)



Crystal structure of filled skutterudites

Filled skutterudites: MT_4X_{12} (M = alkali metal, alkaline earth, rare earth, actinide; T = Fe, Ru, Os; X = P, As, Sb)

Binary skutterudites: TX_3 (T = Co, Rh, Ir; X = P, As, Sb)

Prototype $CoAs_3$: Discovered in Skutterud, Norway

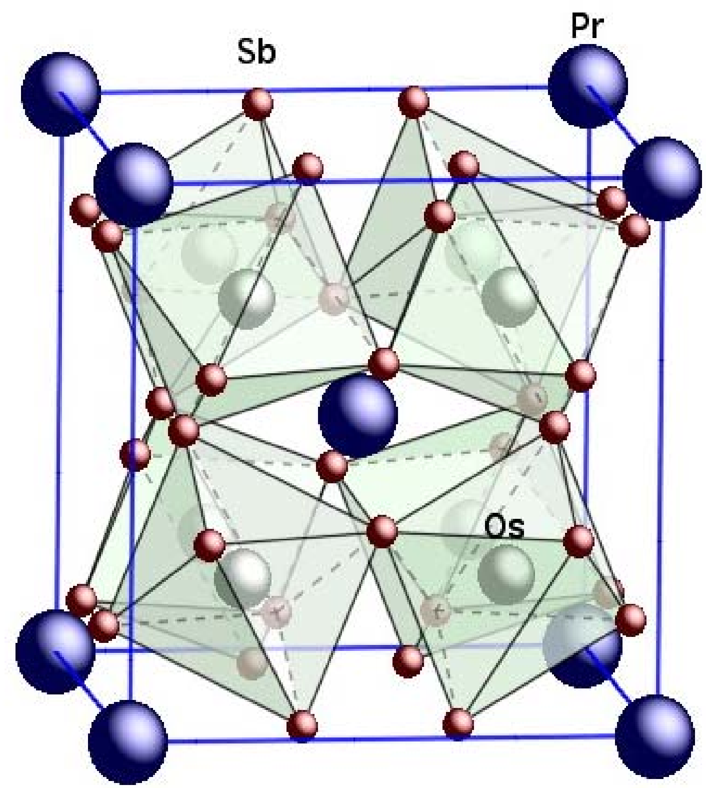
M cations — bcc sublattice (atomic cages — undergo large amplitude vibrations)

T cations — sc sublattice

X anions — distorted corner sharing octahedra centered by T cation

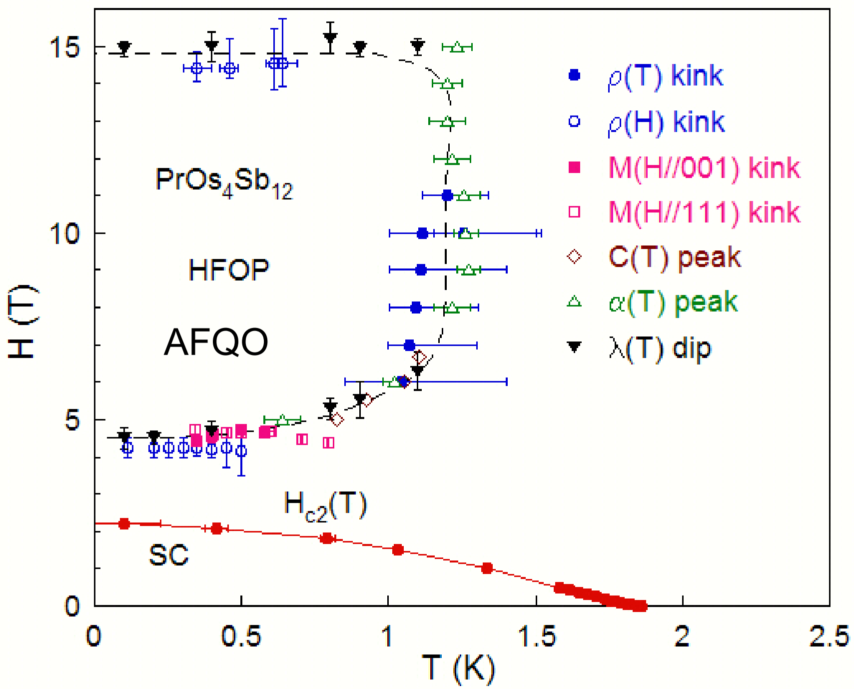
bcc structure (*Im-3*)
a = 9.3068 Å

W. Jeitschko & D. J. Braun 77



$PrOs_4Sb_{12}$

Heavy fermion superconductor $PrOs_4Sb_{12}$



Ho et al., 03

Pr^{3+} energy levels in CEF

INS measurements

Goremychkin et al., 04

———	Γ_3 doublet	(~200 K)
———	Γ_4 triplet	(~130 K)
———	Γ_5 triplet	(~7 K)
———	Γ_1 singlet	(0 K)

- First Pr-based heavy fermion (HF) superconductor (SC) ($m^* \sim 50\ m_e$; $T_c = 1.85$ K) *Bauer et al., 01; Maple et al., 02*
- Unconventional type of strong-coupling SC
- Evidence for several distinct SCing phases, point nodes in $\Delta(\mathbf{k})$, time reversal symmetry breaking
- Possible candidate for triplet SC
- Evidence for multiband SC *Sayfarth et al., 06*
- HFOP identified with antiferro-quadrupolar order (AFQO): neutron diffraction *Kohgi et al., 03*
- SC in vicinity of AFQ QCP!
- HF behavior and unconventional SC may be mediated by electric quadrupole fluctuations
- Off-center rattling and tunneling of Pr filler ion may play role in HF behavior and SC

Goto et al., 04

High temperature superconductivity and spin density waves in transition metal pnictides

High temperature superconductivity and spin density waves in transition metal pnictides

Coworkers

University of California, San Diego

R. E. Baumbach, N. M. Crisosto, J. J. Hamlin, T. A. Sayles, L. Shu, D. A. Zocco

D. N. Basov, M. M. Qazilbash

Oak Ridge National Laboratory

R. Jin, D. Mandrus, M. A. McGuire, B. C. Sales, A. S. Sefat

D. J. Singh, Lijun Zhang

Lawrence Livermore National Laboratory

J. R. Jeffries, S. T. Weir

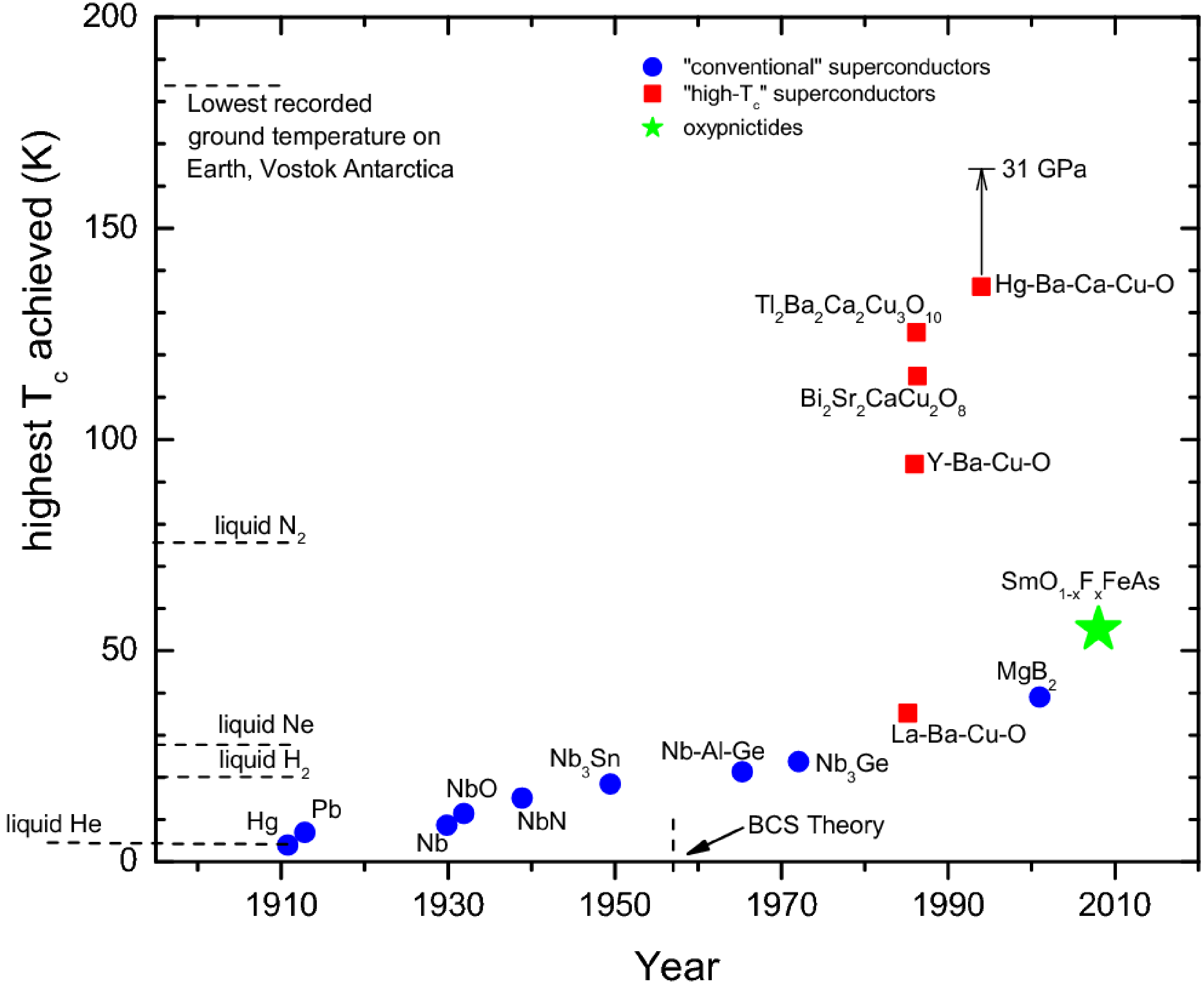
University of Alabama, Birmingham

Y. K. Vohra

Superconductivity in transition metal pnictides

- Recent developments in high T_c superconductivity
  - $LnFeAsO_{1-x}F_x$
    - Maximum $T_c \approx 55$ K (Ln = Sm)
    - Structure type: ZrCuSiAs (1:1:1:1)
  - $A_{1-x}M_xFe_2As_2$
    - Maximum $T_c \approx 38$ K (A = Sr, M = K, Cs; A = Ba, M = K)
    - Structure type: $ThCr_2Si_2$ (1:1:2) (structure of many HF SCs)
- Superconductivity
  - Periodically declared to be "all over"
  - Resuscitated by developments in materials; e.g.,
    - Magnetic superconductors (~75)
    - Organic superconductors (~75)
    - Heavy fermion superconductors (~80)
    - High T_c cuprate superconductors (~86)
    - High T_c transition metal pnictide superconductors (~08)

Highest T_c achieved vs year



Iron-pnictide high-temperature superconductors

Wolfgang Jeitschko

Journal of Alloys and Compounds 302 (2000) 70–74

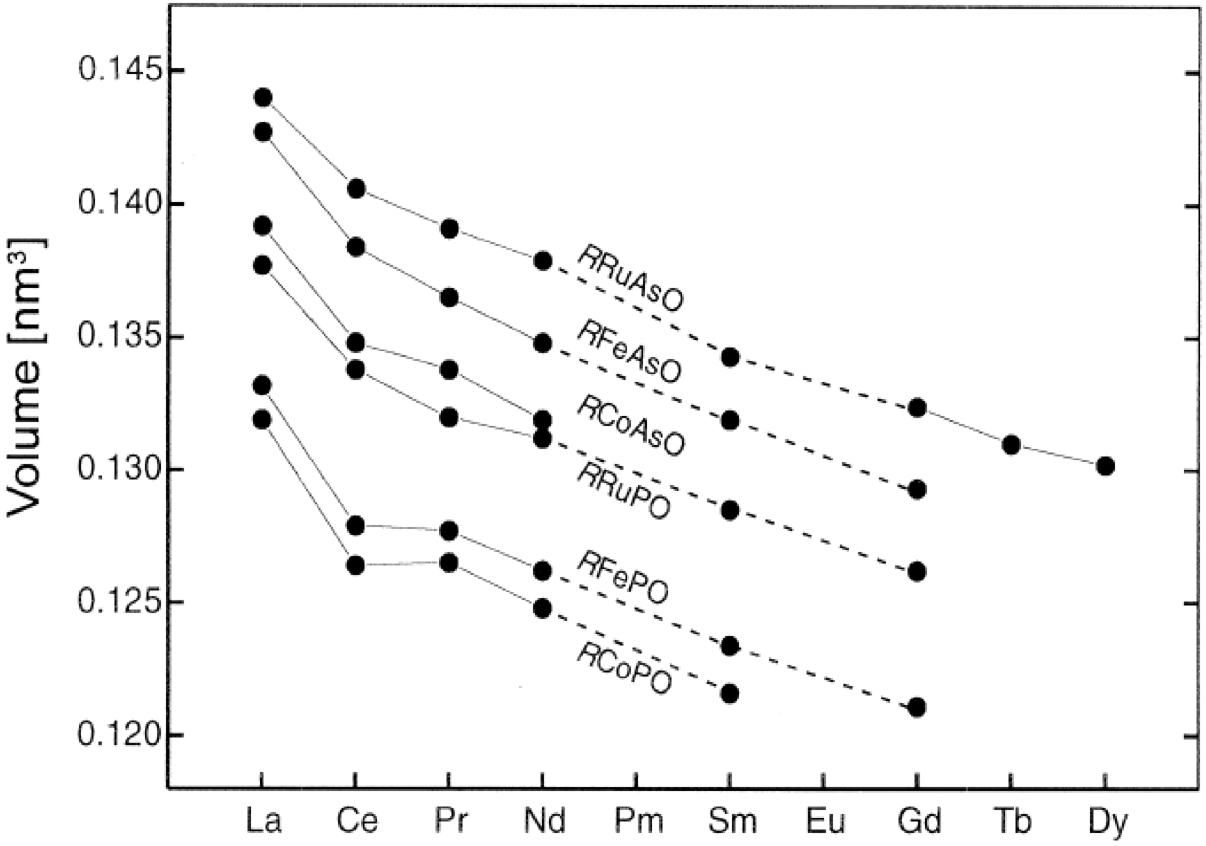


Fig. 1. Cell volumes of compounds *RTPn*O (*T* = Fe, Ru, Co; *Pn* = P, As) with ZrCuSiAs type structure.

J|A|C|S
COMMUNICATIONS

Published on Web 07/15/2006

Iron-Based Layered Superconductor: LaOFeP

Yoichi Kamihara,† Hidenori Hiramatsu,† Masahiro Hirano,†,‡ Ryuto Kawamura,§ Hiroshi Yanagi,§
Toshio Kamiya,†,§ and Hideo Hosono*,†,‡

LaFePO	$T_c \sim 3$ K
$LaFePO_{1-x}F_x$	$T_c \sim 5$ K

J|A|C|S
COMMUNICATIONS

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LaFePO	$T_c \sim 3$ K
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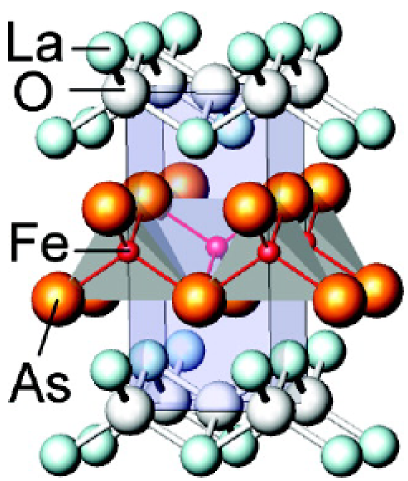
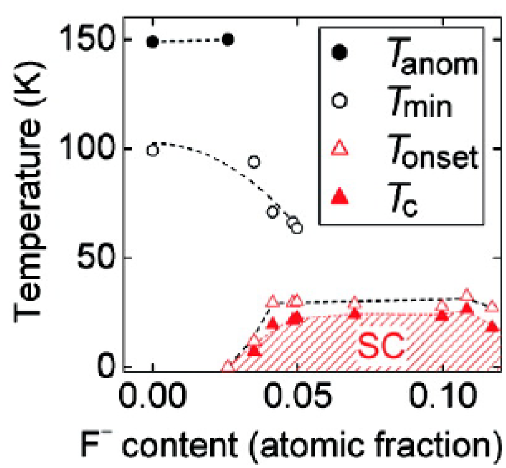
Iron-Based Layered Superconductor La[OF]FeAs (x = 0.05–0.12) with T = 26 K

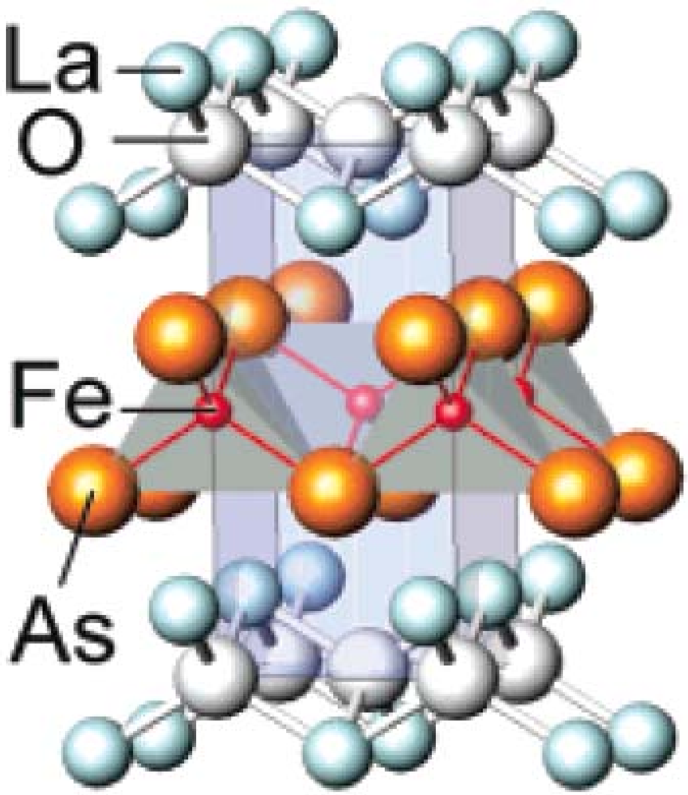
Yoichi Kamihara, Takumi Watanabe, Masahiro Hirano, and Hideo Hosono

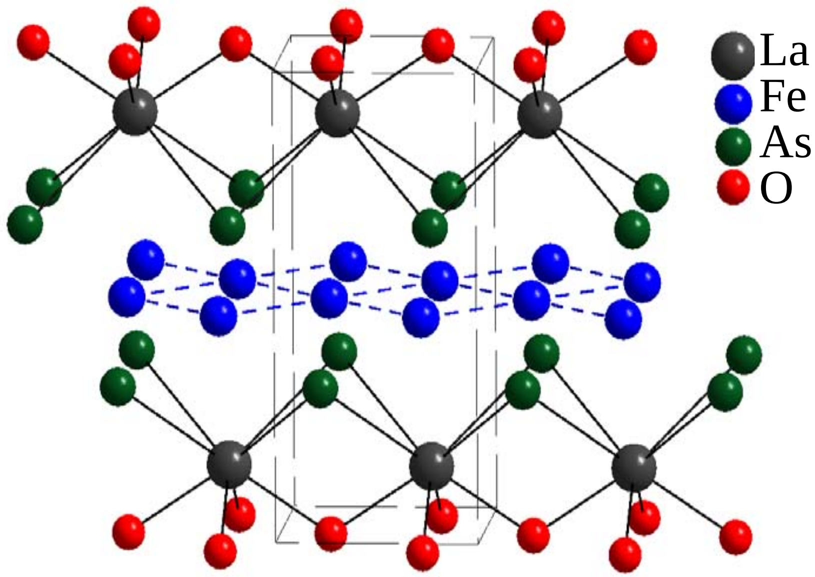
J. Am. Chem. Soc., **2008**, 130 (11), 3296-3297• DOI: 10.1021/ja800073m • Publication Date (Web): 23 February 2008





Structure of LaFeAsO

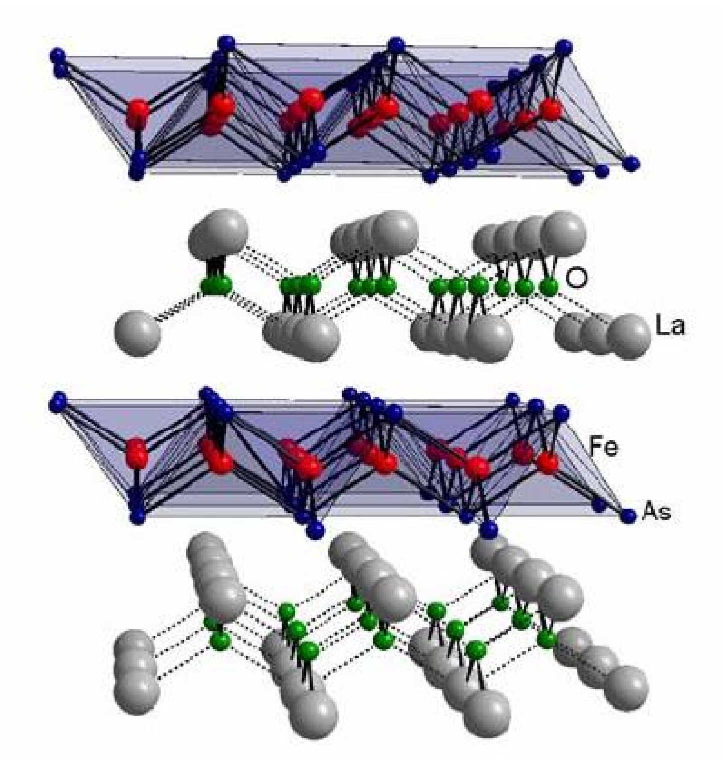




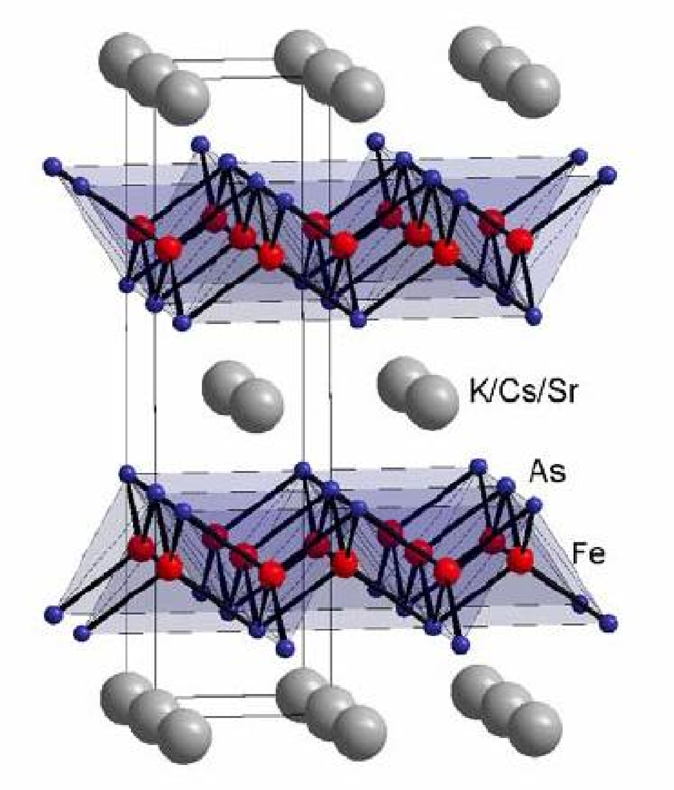
- Tetragonal ZrCuSiAs prototype
- Edge sharing $FeAs_4$ tetrahedra
- Fe atoms form square nets
- Fe-Fe bonding likely to be important
- a = 4.034 Å, c = 8.745 Å

Structure of TM pnictide 1:1:1:1 and 1:2:2 superconductors

Structure type: ZrCuSiAs



Structure type: $ThCr_2Si_2$

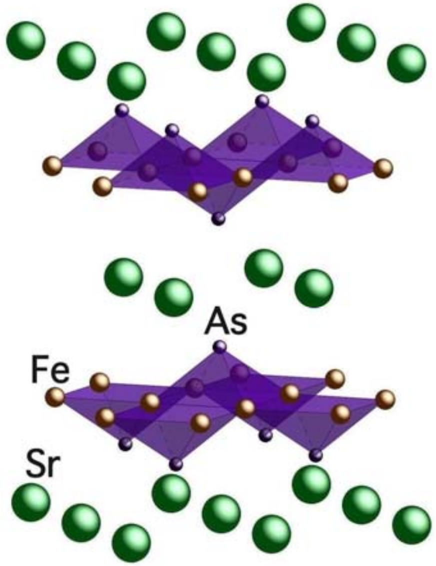


K. Sasmal, B. Lv, B. Lorenz, A. Guloy, F. Chen, Y. Xue, C. W. Chu, arXiv:0806.1301 (2008)

$LaFeAsO_{1-x}F_x$	26 K
$CeFeAsO_{1-x}F_x$	40 K
$NdFeAsO_{1-x}F_x$	50 K
$SmFeAsO_{1-x}F_x$	55 K

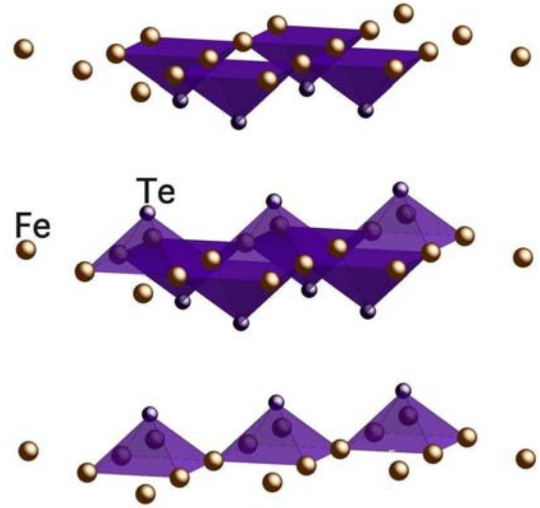
$Ba_{1-x}K_xFe_2As_2$	38 K
$Ca_{1-x}Na_xFe_2As_2$	40 K

Transition metal-pnictogen structures

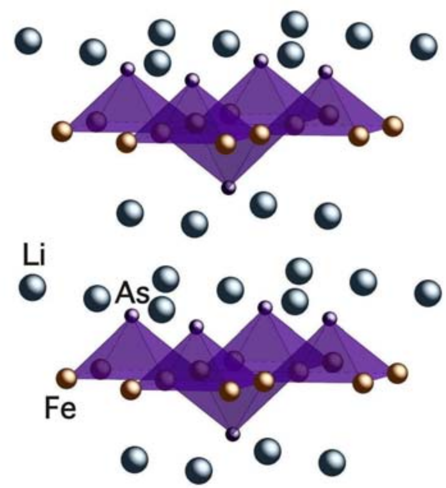


$SrFe_2As_2$

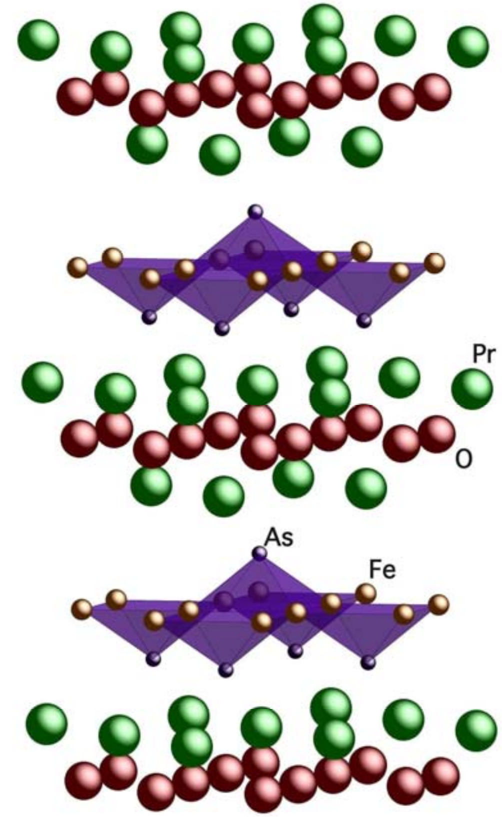
“122”



FeTe “11”



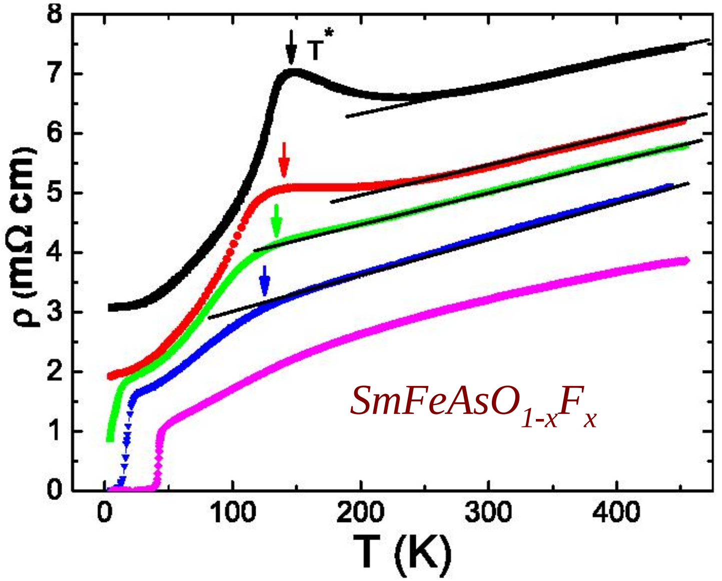
LiFeAs “111”

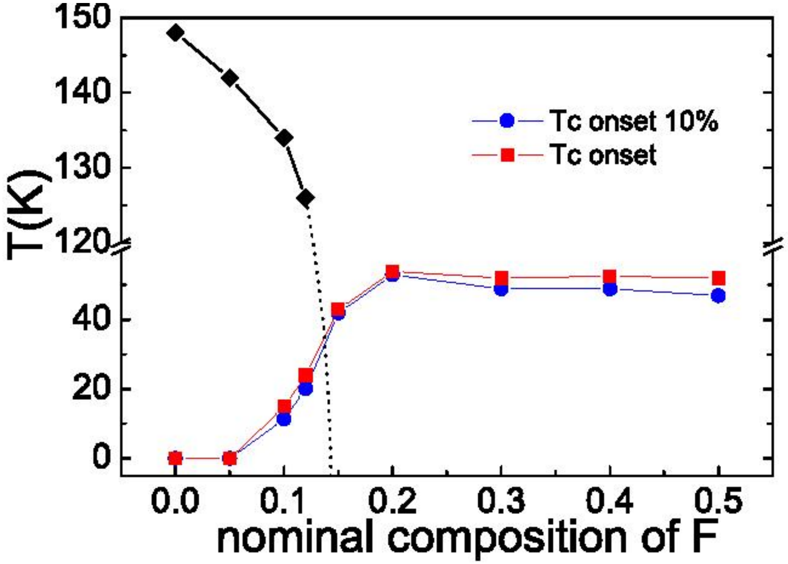


PrFeAsO

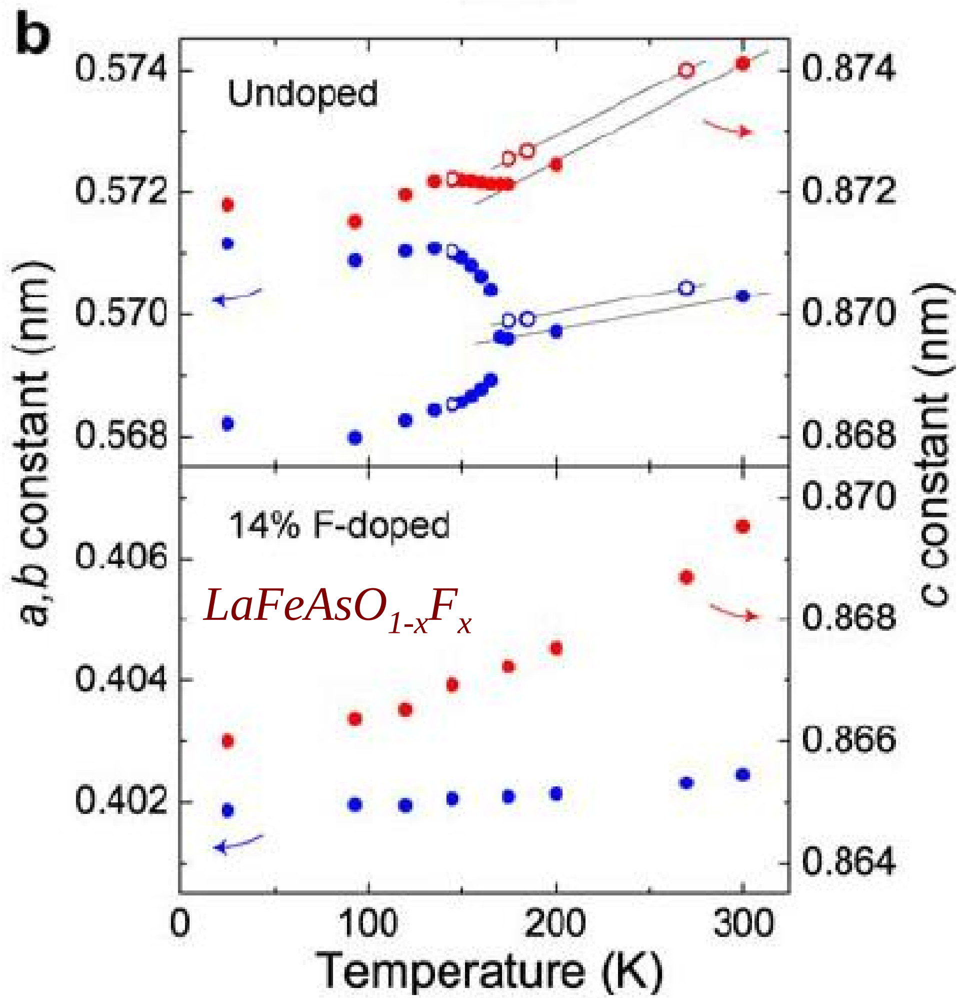
“1111”

$LnFeAsO_{1-x}F_x$ high T_c superconductors



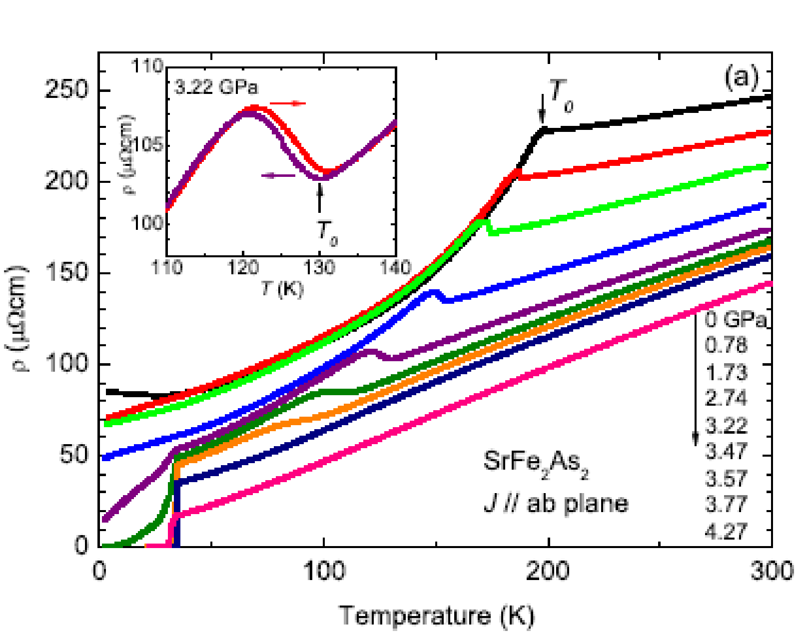


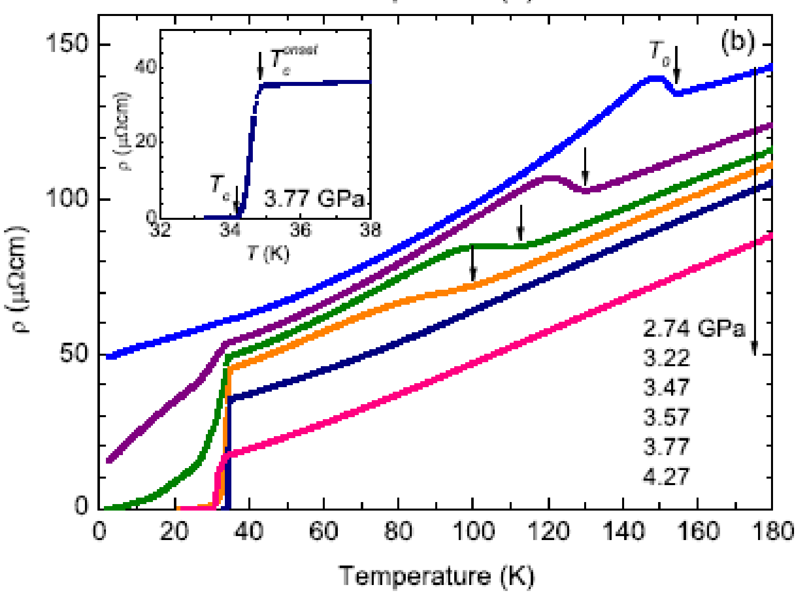
Liu et al. arxiv:0804.2105

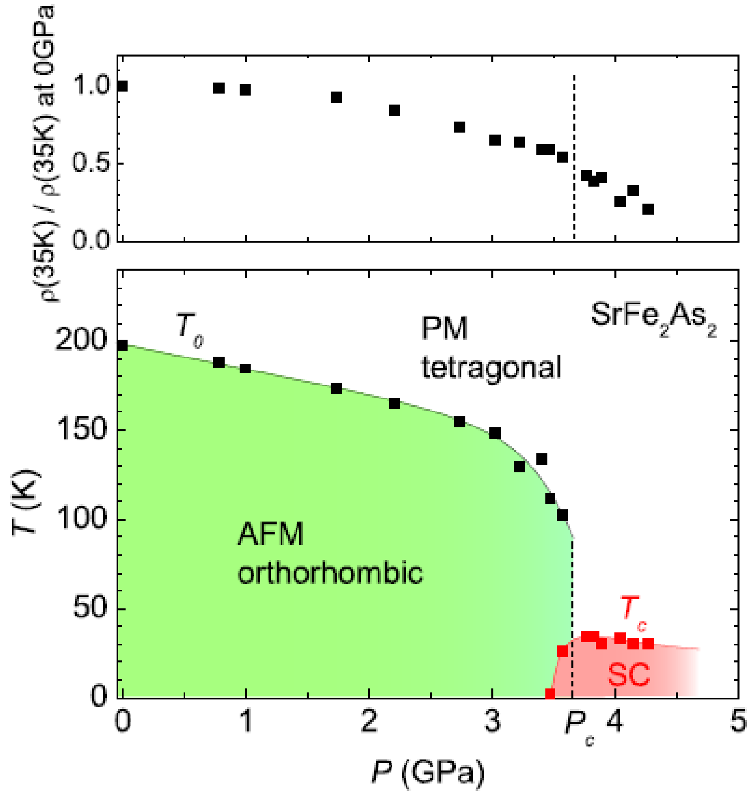


Nomura et al. arxiv:0804.3569

SDW and SC under pressure $SrFe_2As_2$



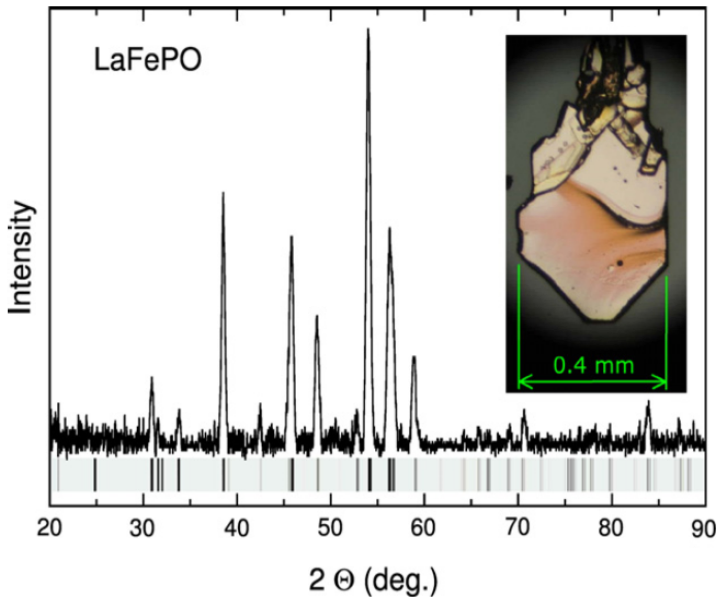


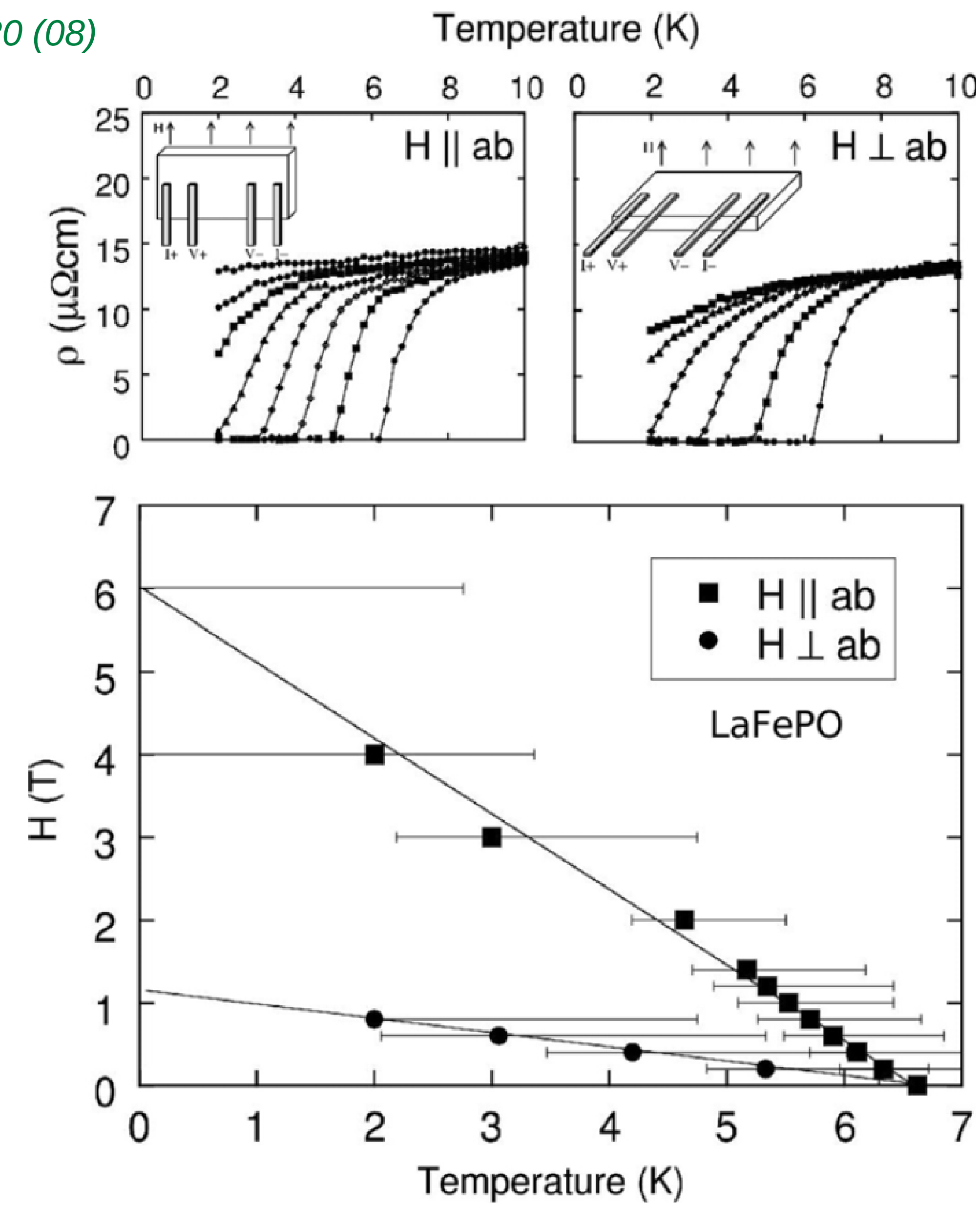


Tou et al., arXiv: 0810.4856v1 (08)

Superconductivity in LaFePO

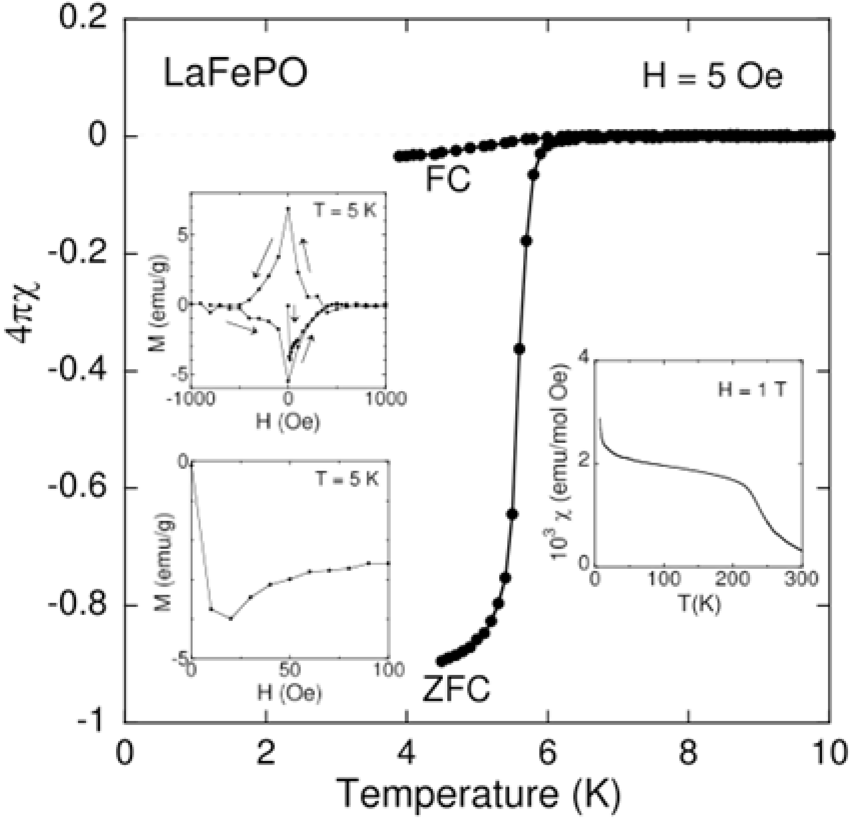
*J. J. Hamlin, R. E. Baumbach, D. A. Zocco, T. A. Sayles, M. B. Maple, JPCM **20**, 365220 (08)*

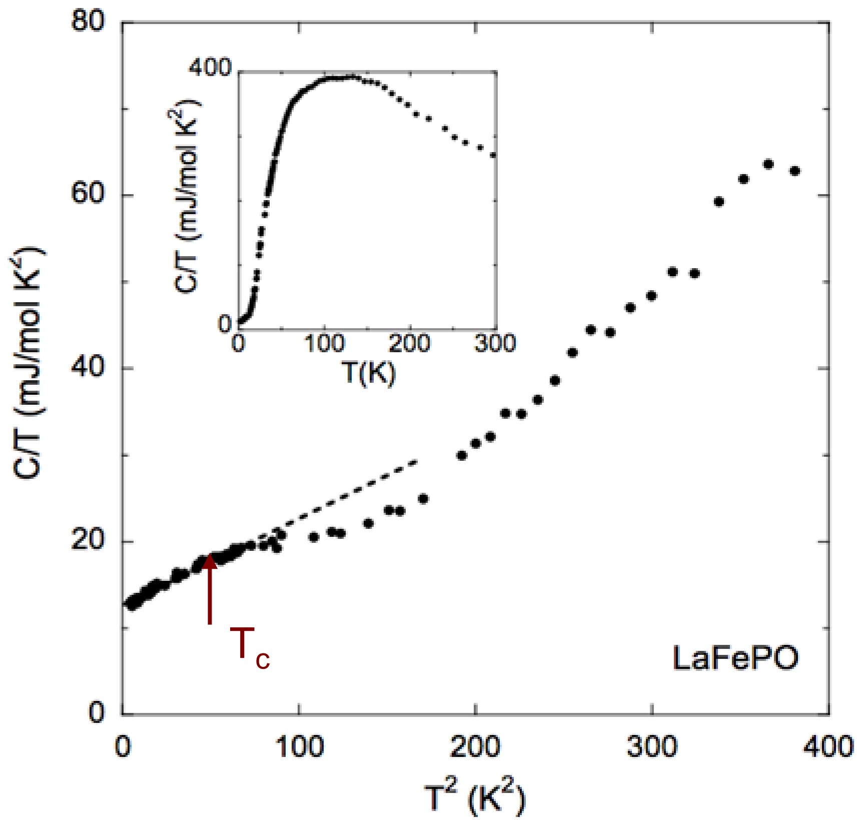




- LaFePO single crystals grown in Sn flux
- Superconductivity: T_c = 6.7 K
- Significant anisotropy of resistively determined $H_{c2}(T)$ curves

Superconductivity in LaFePO



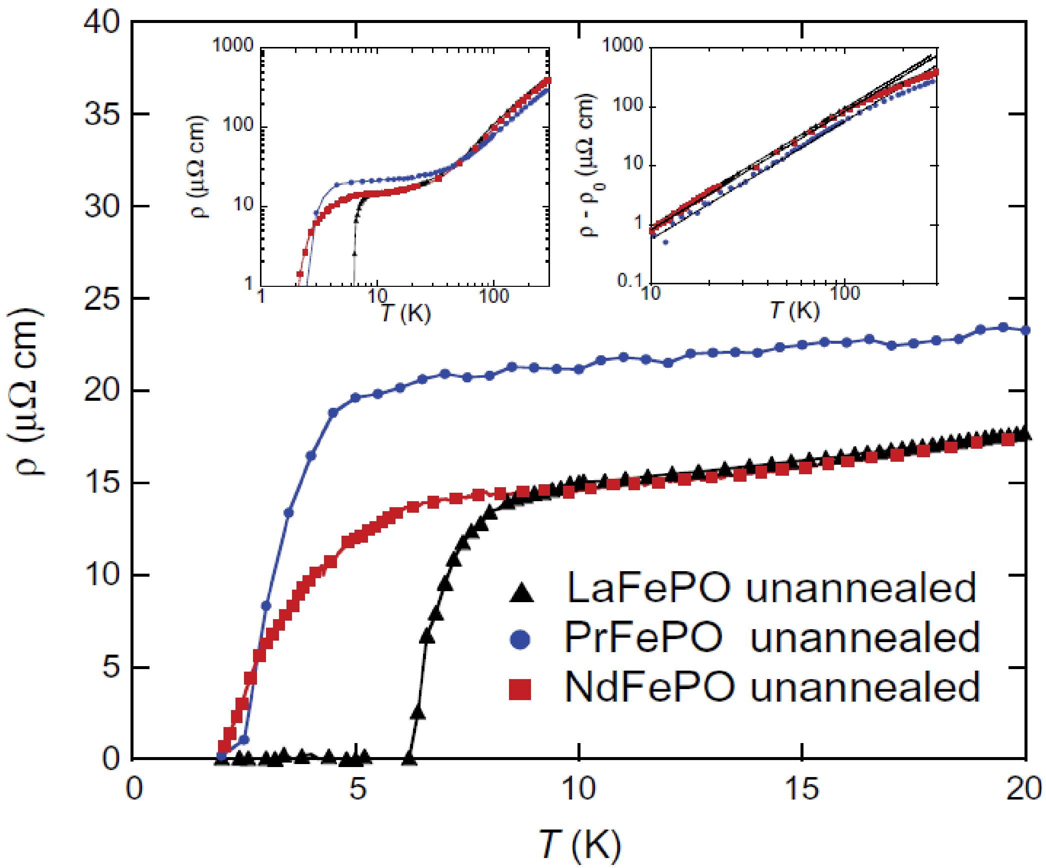


- No measurable jump in C(T) at T_c = 6.7 K
- Very small superconducting fraction?
- Superconductivity due to oxygen defects?

J. J. Hamlin, R. E. Baumbach,
D. A. Zocco, T. A. Sayles,
*M. B. Maple, JPCM **20**, 365220 (08)*

Superconductivity in LnFePO (Ln = La, Pr, Nd)

LnFePO (Ln = La, Pr, Nd)

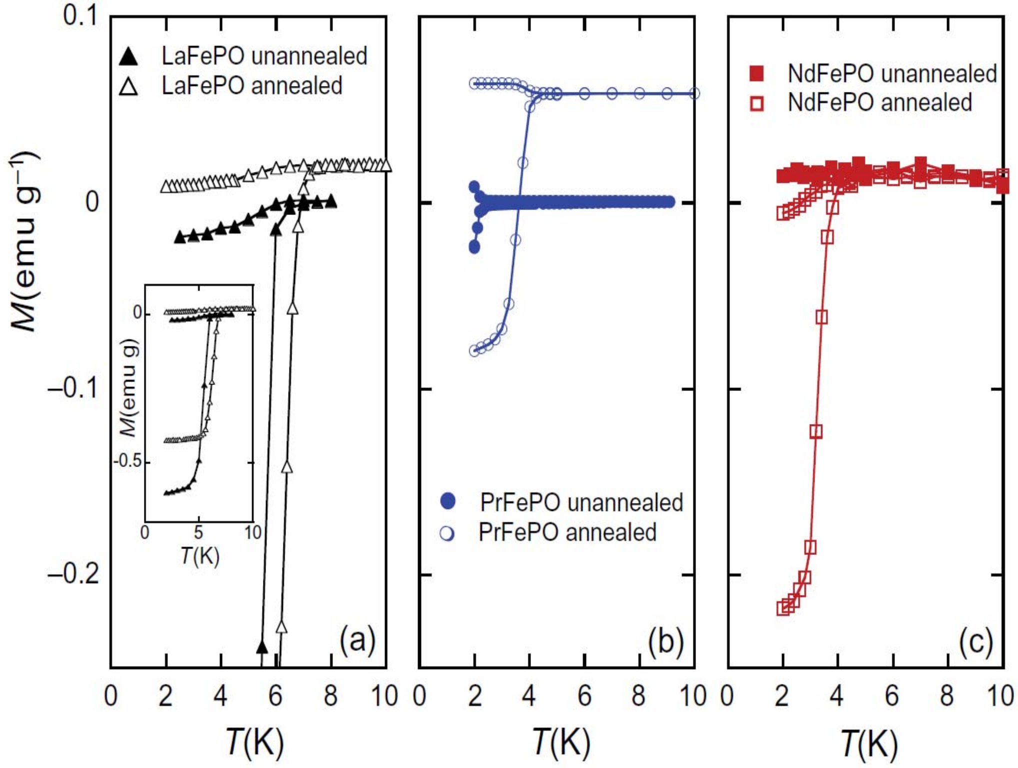


- T_c lower for Ln = Pr, Nd
- Metallic behavior
- No evidence for SDW
- $\rho \approx T^2$ for 10 K < T < 100 K
- Large RRR values

Ln	T_c (K)	RRR
La	6.6	32
Pr	3.2	14
Nd	3.1	28

*R. E. Baumbach, J. J. Hamlin, L. Shu, D. A. Zocco, N. M. Crisosto, and M. B. Maple, NJP **11**, 025018 (09)*

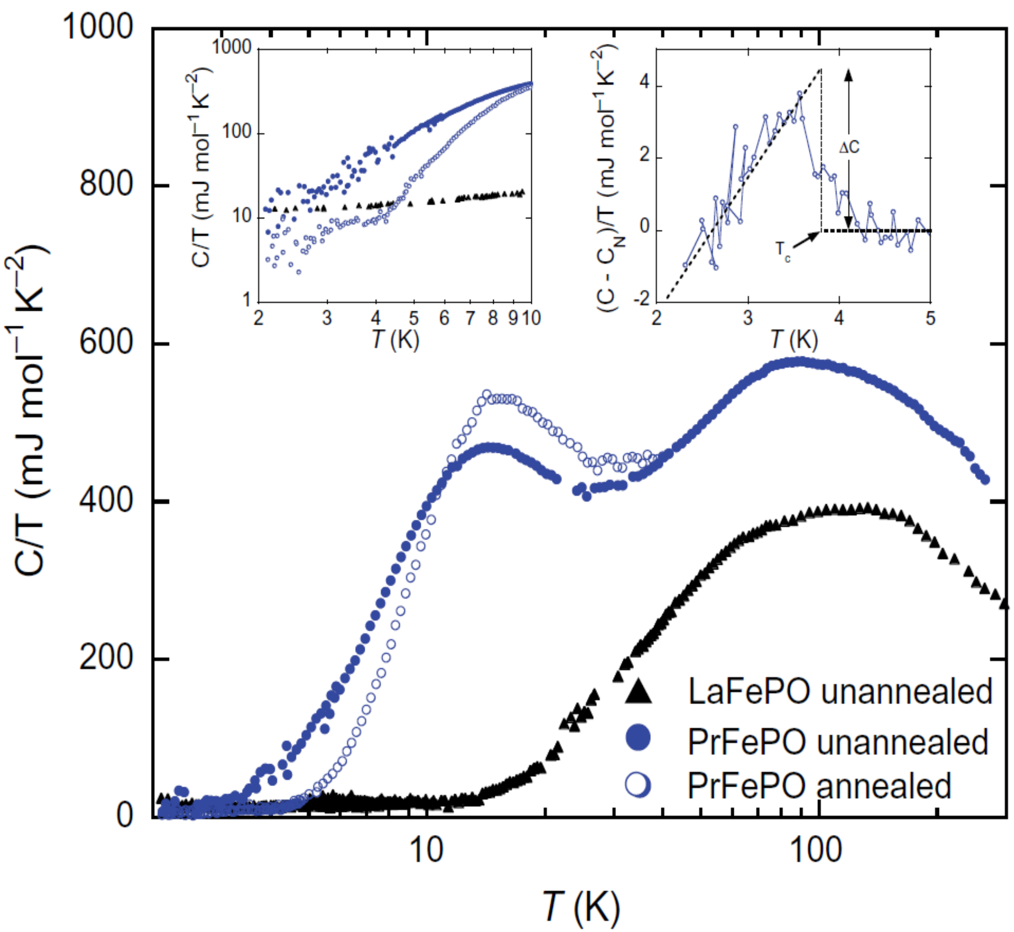
O_2 annealing for LnFePO (Ln = La, Pr, Nd)



- Unannealed PrFePO and NdFePO show nearly zero shielding
- O_2 annealing improves shielding for PrFePO and NdFePO
- O_2 annealing slightly increases T_c for LaFePO

*R. E. Baumbach, J. J. Hamlin, L. Shu, D. A. Zocco, N. M. Crisosto, and M. B. Maple, NJP **11**, 025018 (09)*

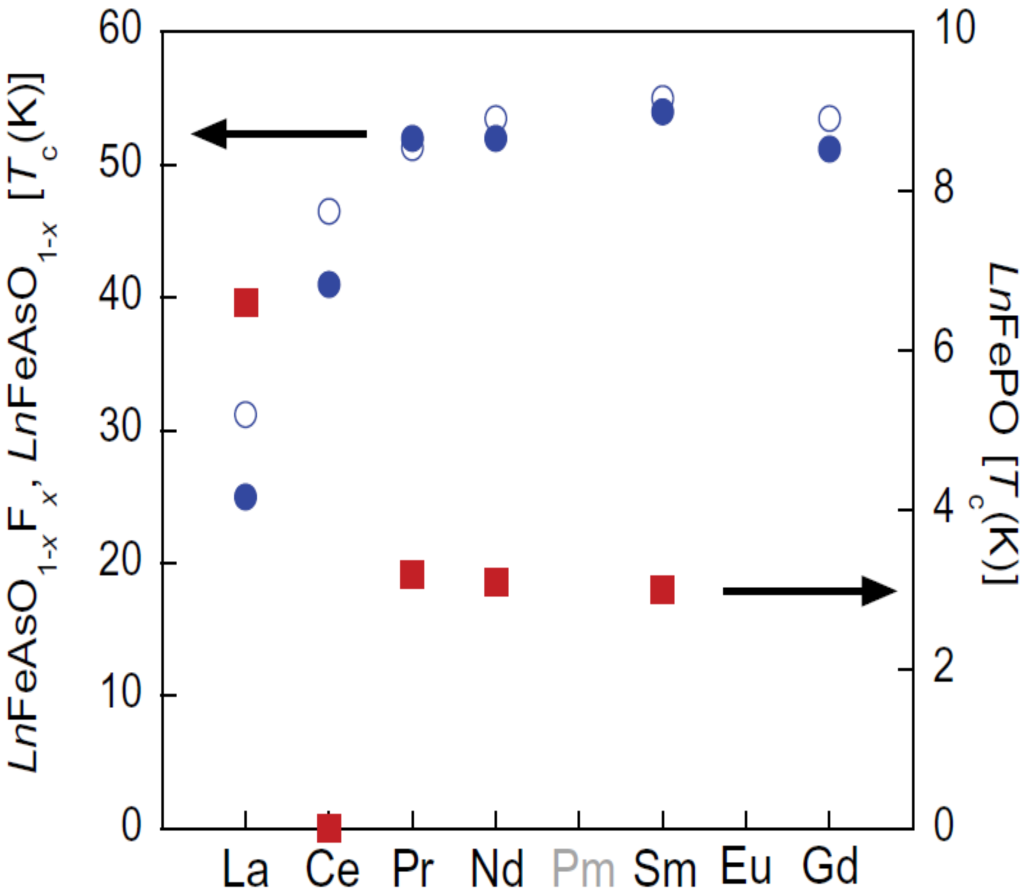
C(T) for LnFePO (Ln = La, Pr) single crystals



*R. E. Baumbach, J. J. Hamlin, L. Shu, D. A. Zocco, N. M. Crisosto, and M. B. Maple, NJP **11**, 025018 (09)*

- No specific heat jump for as-grown LaFePO and PrFePO
- Specific heat jump observed for O_2 annealed PrFePO
- Jump is nearly 100 % of value expected from BCS for O_2 annealed PrFePO
- Schottky anomaly for PrFePO
- CEF fits suggest nonmagnetic ground state and first excited state: $\Delta \sim 41$ K
- Annealing sharpens CEF peak – may indicate reduced spread of CEF splittings
- Small γ for LaFePO and PrFePO

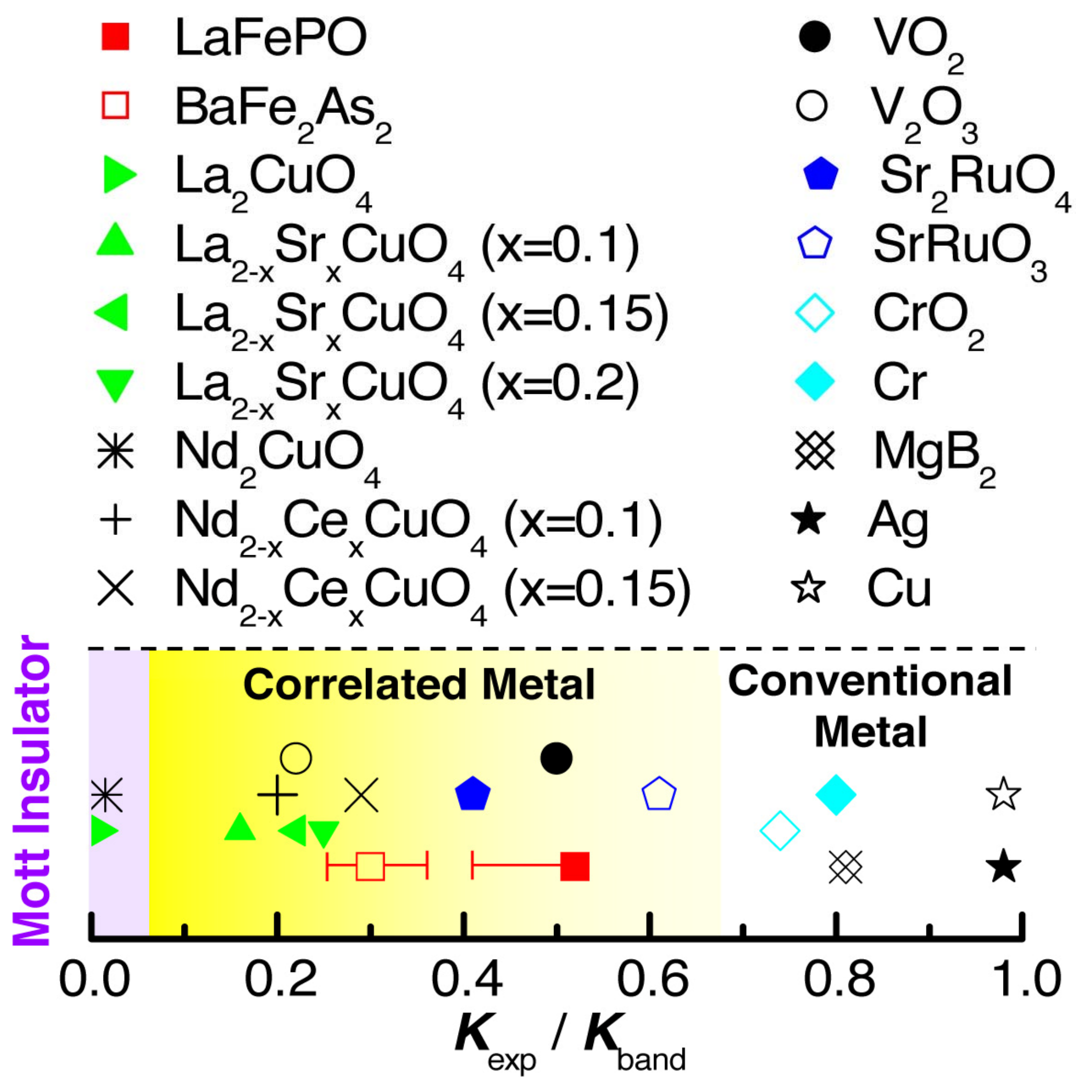
Comparison of T_c vs Ln for Pn = P, As



R. E. Baumbach, J. J. Hamlin, L. Shu, D. A. Zocco, N. M. Crisosto, and M. B. Maple, NJP **11**, 025018 (09)

- Solid circles: $LnFeAsO_{1-x}F_x$
- Open circles: $LnFeAsO_{1-x}$
- Solid squares: LnFePO
- Much lower T_c for Pn = P
- For Pn = As: T_c enhanced with magnetic Ln
- For Pn = P: T_c depressed with magnetic Ln
- Possible magnetic pair-breaking for LnFePO
- Suggests that arsenides may be unconventional SCs, while phosphides are conventional (BCS) superconductors!

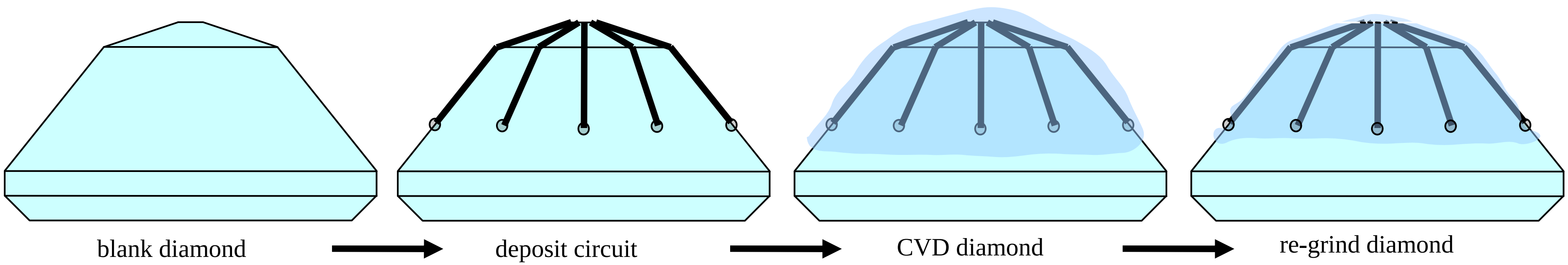
Ratio of kinetic energy from experiment and band theory K_{exp}/K_{band} for iron pnictides and various other materials

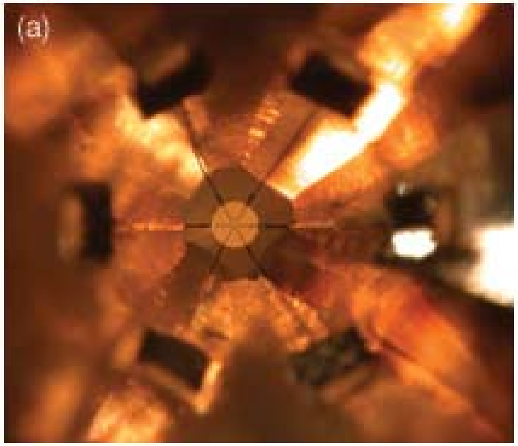


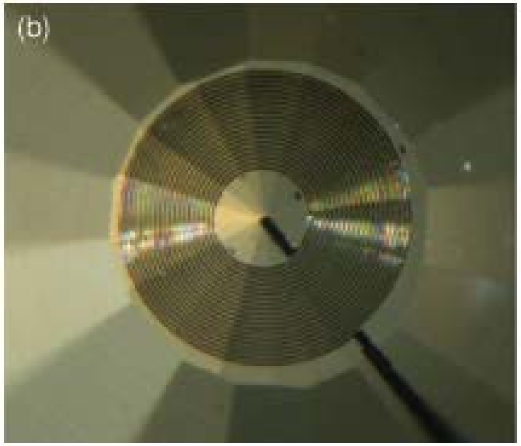
Infrared and optical study of electronic correlations in LaFePO

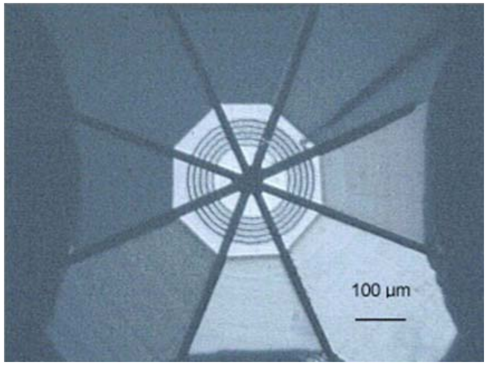
M. M. Qazilbash, J. J. Hamlin, R. E. Baumbach, Lijun Zhang, D. T. Singh, M.B. Maple amd D. N. Basov, Nature Physics (09)

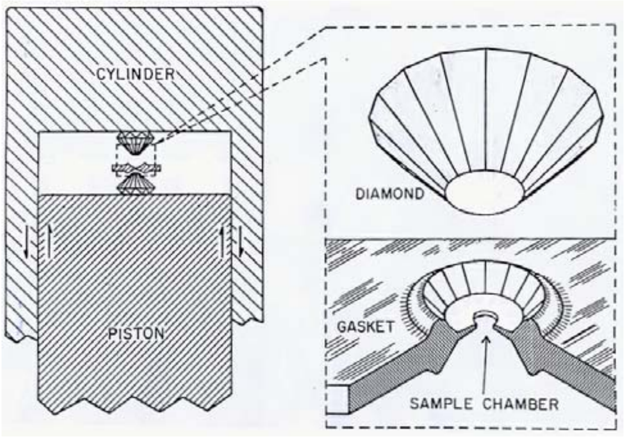
Designer Diamond Anvils



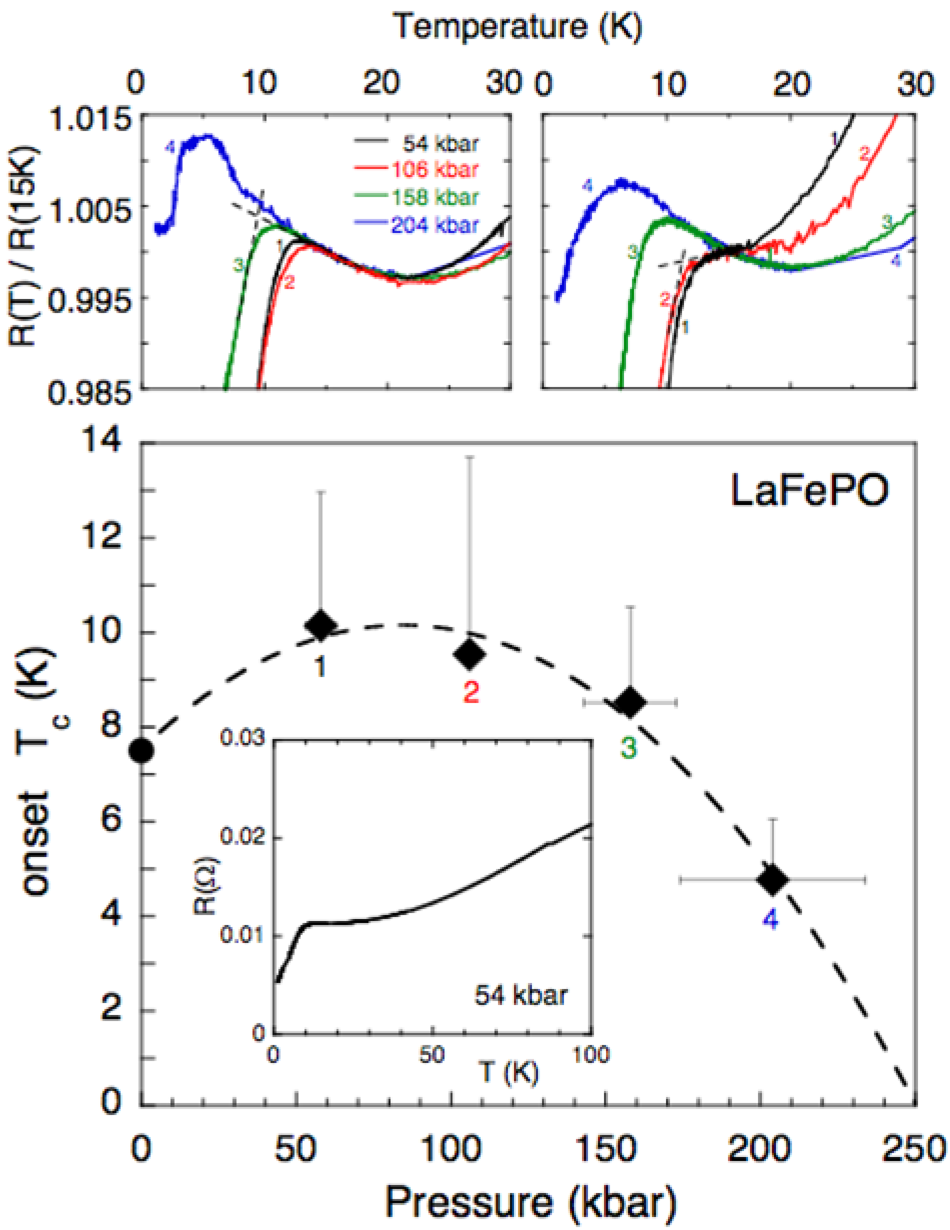








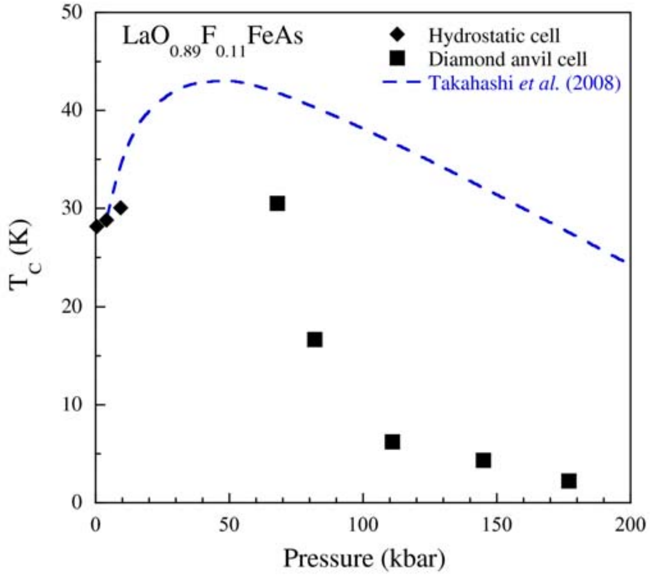
$\rho(T, P)$ for LaFePO

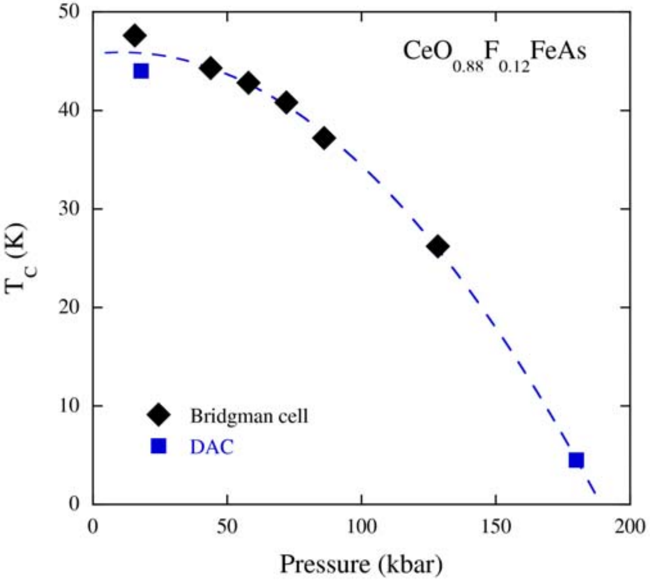


- Dome-shaped $T_c(P)$ curve
- $T_{c,\ max}$ ~ 14 K
- Similar behavior of $T_c(P)$ for optimally doped $LaFeAsO_{1-x}F_x$ and $CeFeAsO_{1-x}F_x$
- Reminiscent of $T_c(x)$ in cuprates and $T_c(P)$ in heavy fermion f-electron compounds

*J. J. Hamlin, R. E. Baumbach, D. A. Zocco, T. A. Sayles, M. B. Maple, JPCM **20**, 365220 (08)*

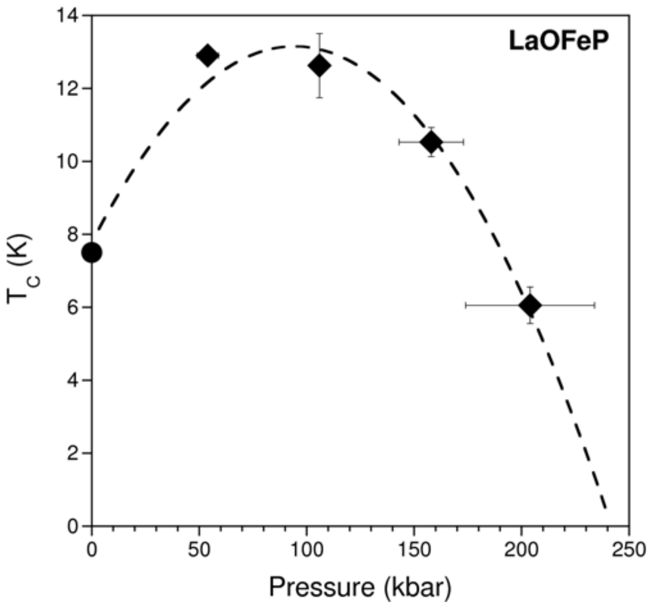
Oxypnictide superconductors: pressure dependence of T_c





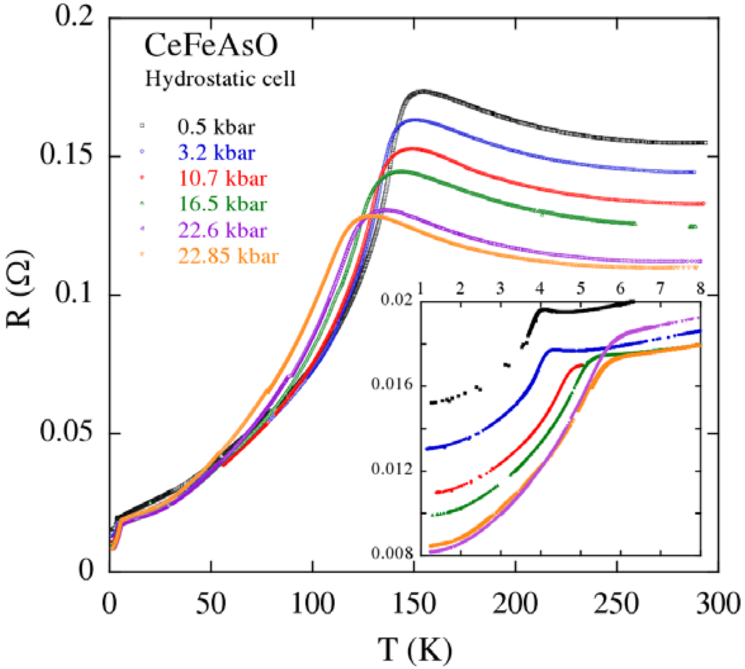
D. A. Zocco, J. J. Hamlin, R. E. Baumbach, M. B. Maple, M. A. Maguire, B. C. Sales, A. S. Sefat, R. Jin, D. Mandrus, J. R. Jeffries, S. T. Weir, Y. K. Vohra, J. Superconductivity 08

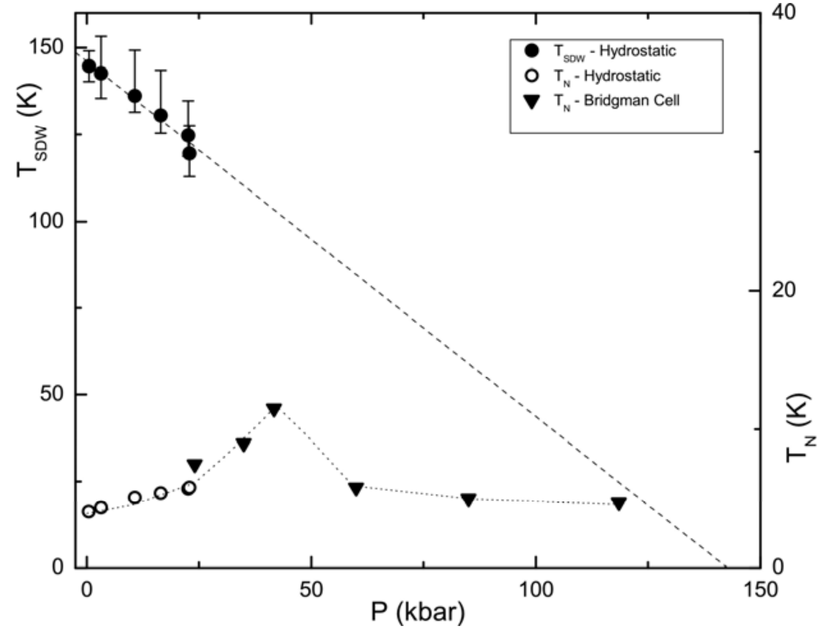
J. J. Hamlin, R. E. Baumbach, D. A. Zocco, T. A. Sayles, M. B. Maple, J. Phys.: Cond. Matt. 08



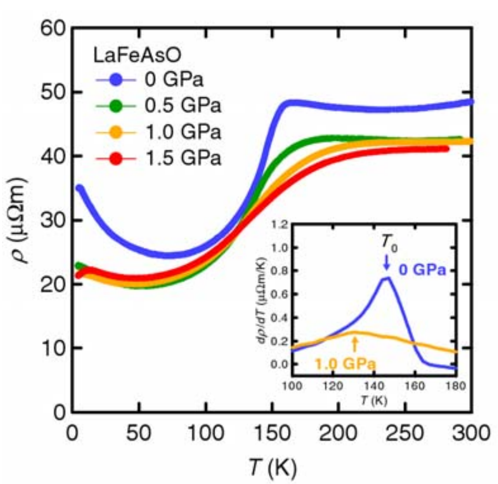
- Dome-shaped dependence of T_c on P
- Reminiscent of T_c vs x in cuprates and T_c vs P in heavy fermion compounds

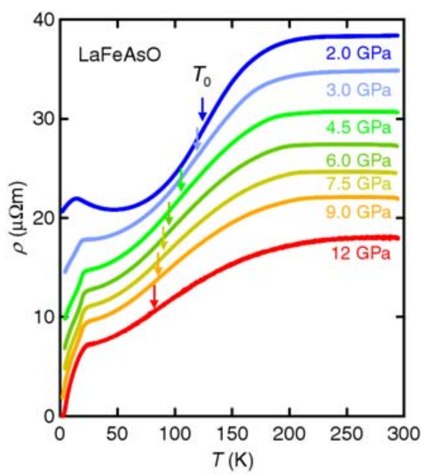
Electrical resistivity of CeFeAsO under pressure

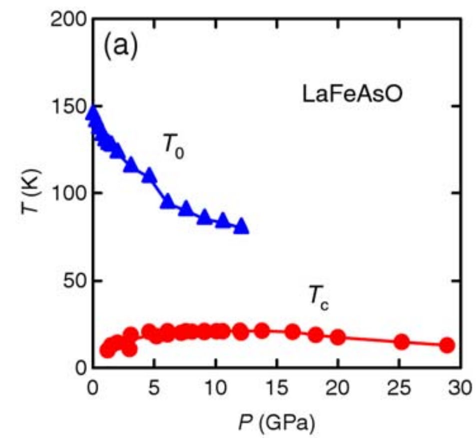




D. A. Zocco, J. J. Hamlin, M. B. Maple, unpublished (09)





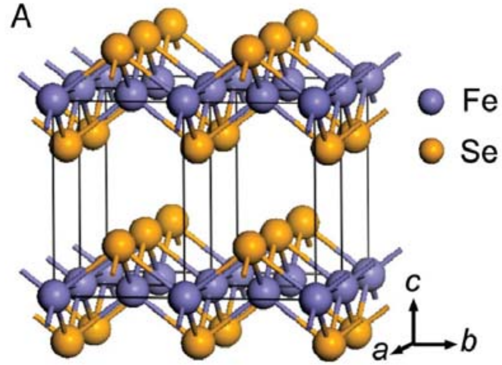


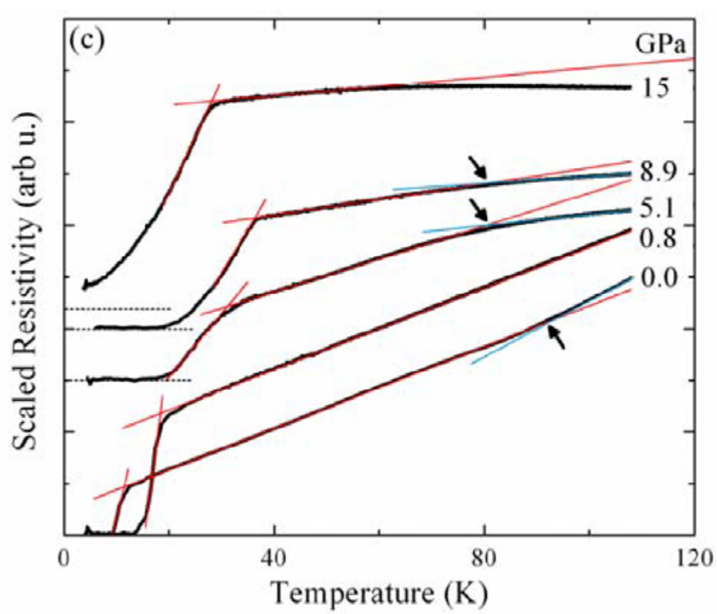
*Okada et al., JPSJ **77**, 113712 (2008)*

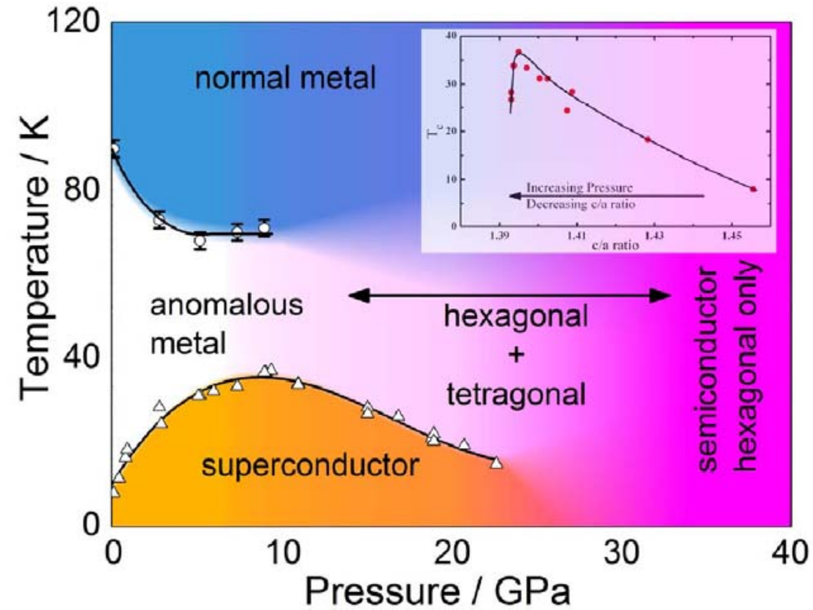
Superconductivity at 36 K in β-$Fe_{1.01}$Se with the Compression of the Interlayer Separation Under Pressure

S. Medvedev[1,2], T. M. McQueen[3], I. Trojan[2,4], T. Palasyuk[2,5], M. I. Eremets[2], R. J. Cava[3], S. Naghavi[1], F. Casper[1], V. Ksenofontov[1], G. Wortmann[6] and C. Felser[1]

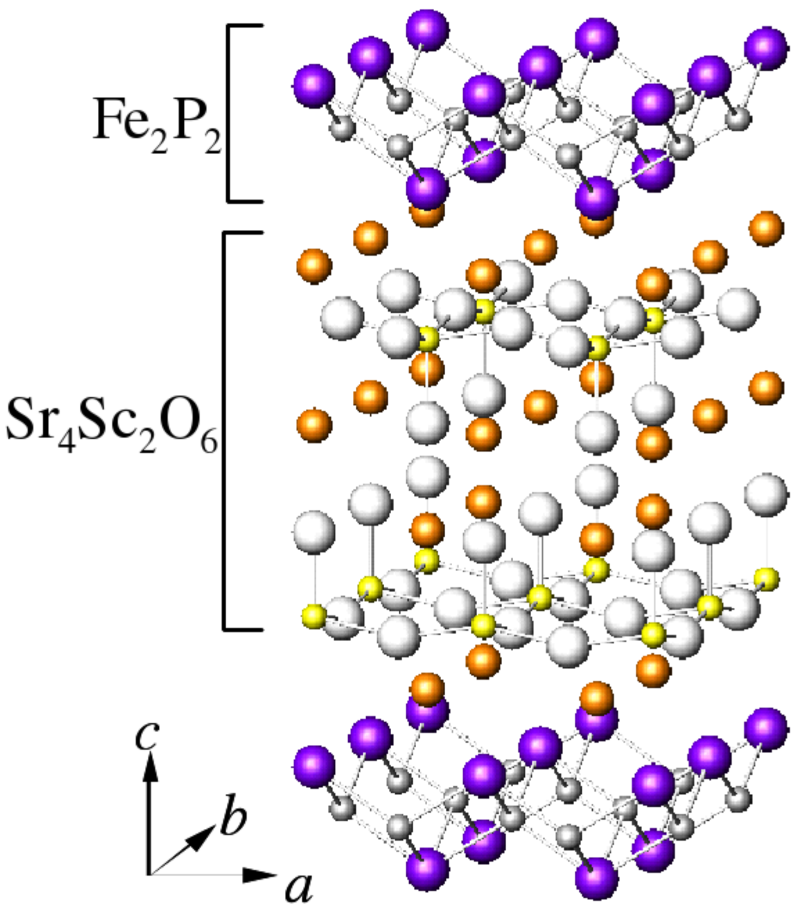
arXiv:0903.2143 (2009)



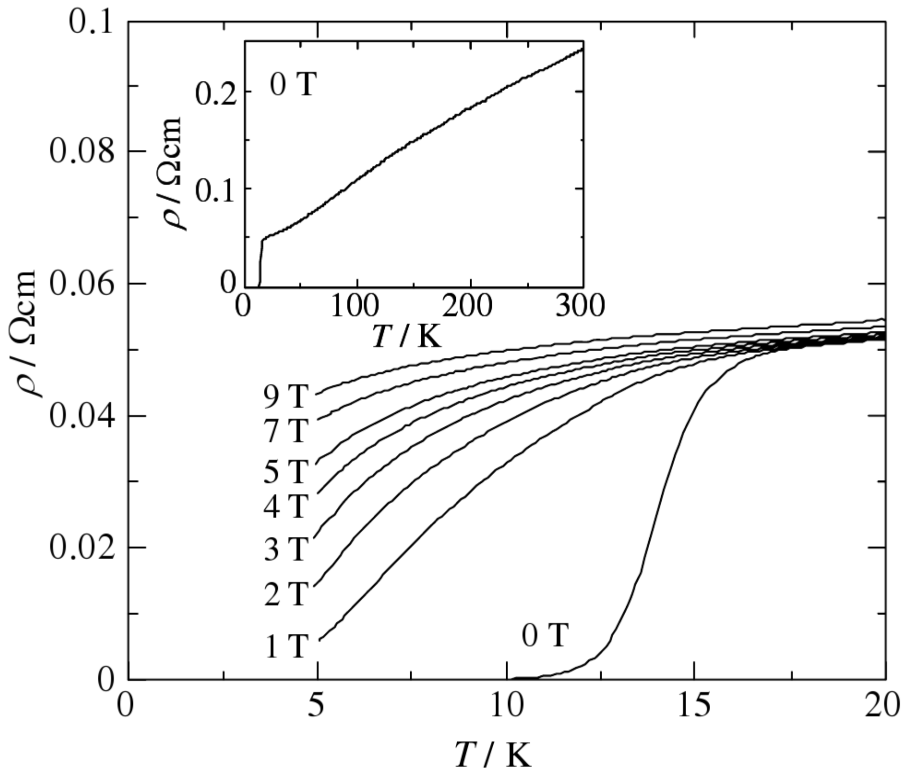




Superconductivity at 17K in $Sr_4Sc_2Fe_2P_2O_6$: new superconducting layered oxypnictides with thick perovskite oxide layer



Ogino et al., arXiv:0903.3314



Interplay of superconducting, charge density wave, and magnetic order in rare earth tritelluride RTe_3 compounds

Interplay of superconducting, charge density wave, and magnetic order in rare earth tritelluride RTe_3 compounds

<u>Coworkers</u>

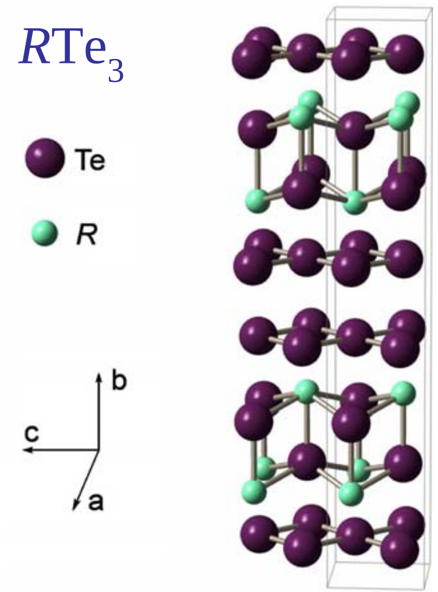
University of California, San Diego

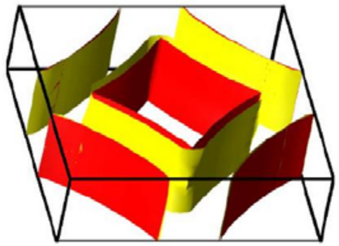
J. J. Hamlin, T. A. Sayles, D. A. Zocco

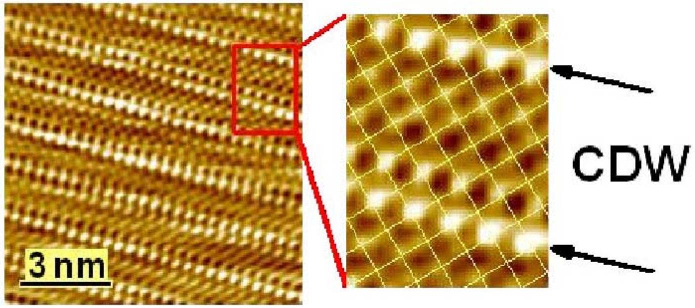
Stanford University

J.-H. Chu, I. R. Fischer

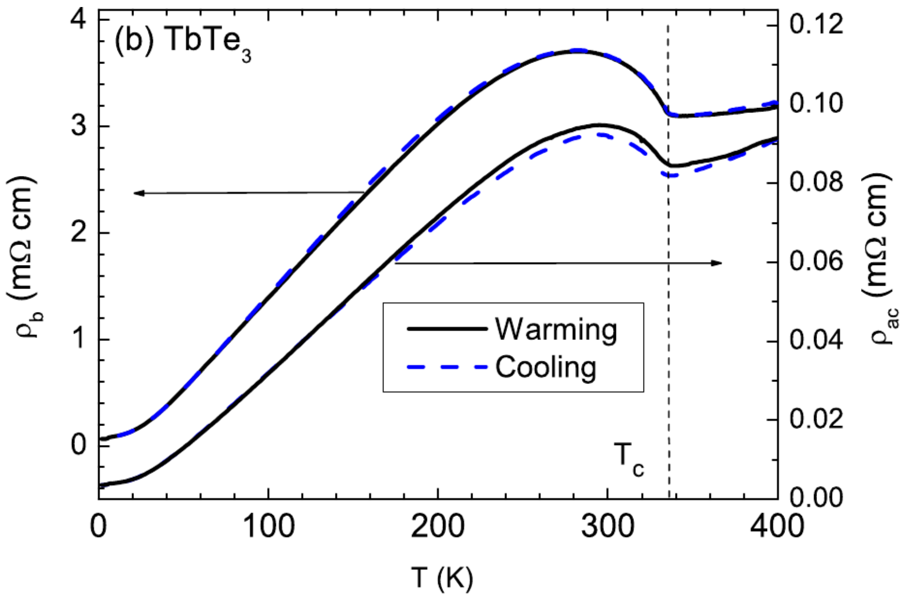
"2D" charge density waves

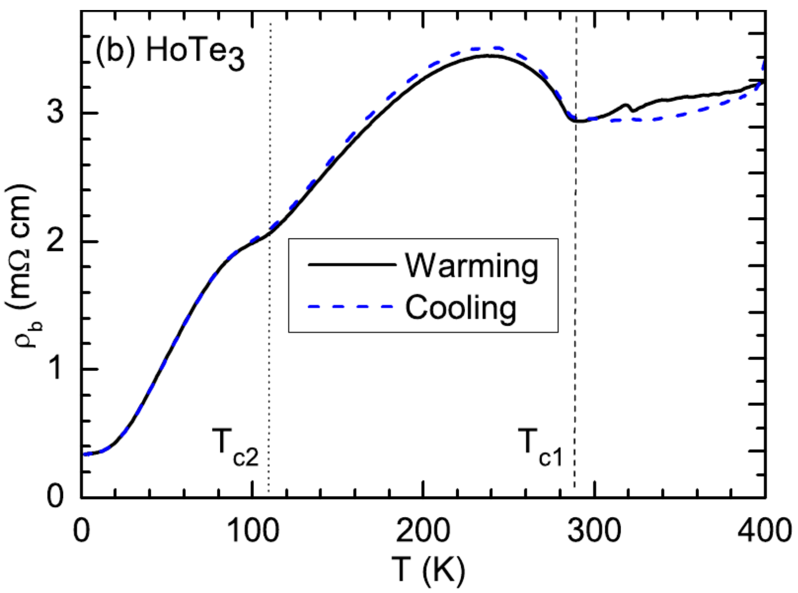




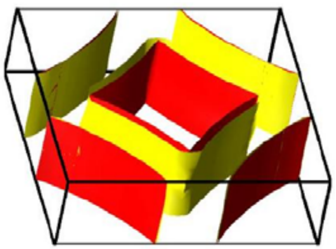


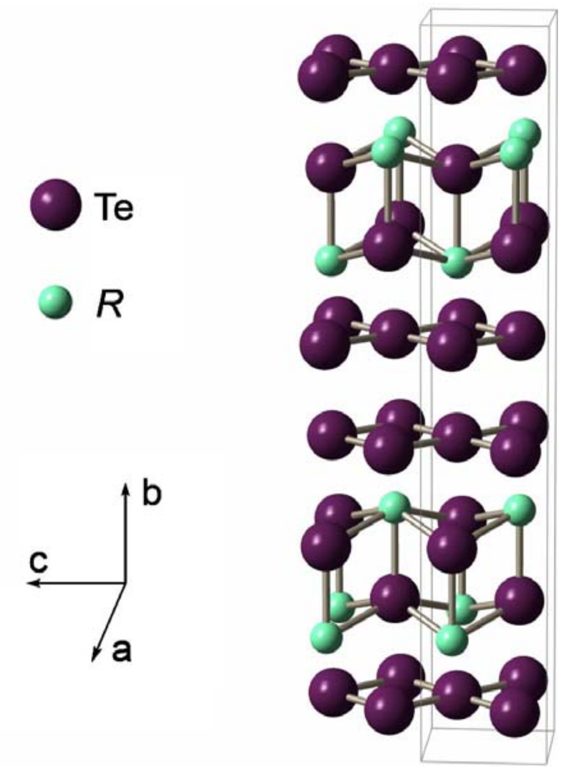
- *Kim et al., PRL **96**, 226401 (2006)*
- *Ru et al., PRB **77**, 035114 (2008)*

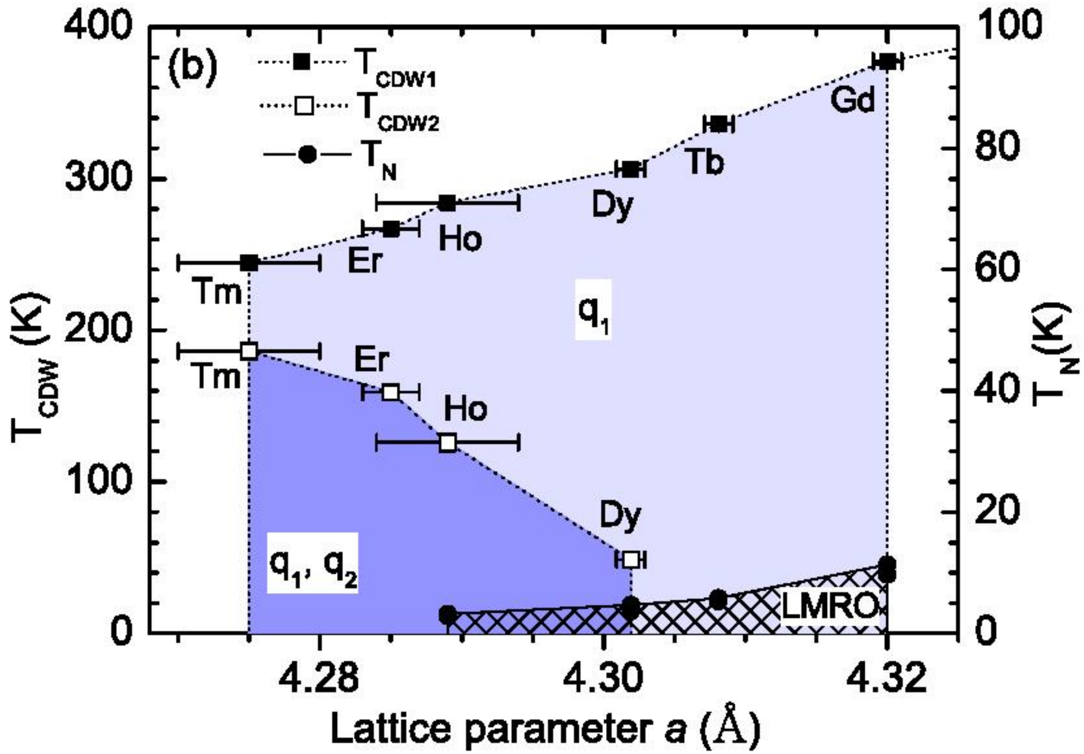




Rare earth tritellurides: RTe_3





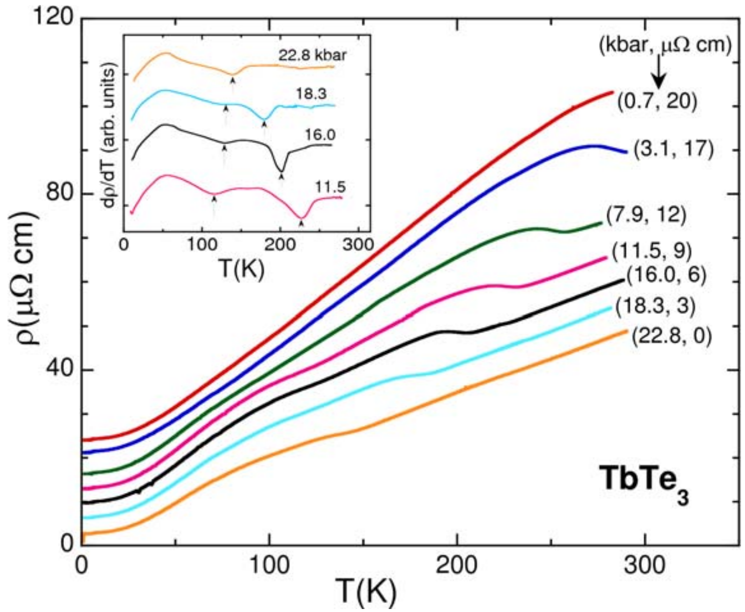


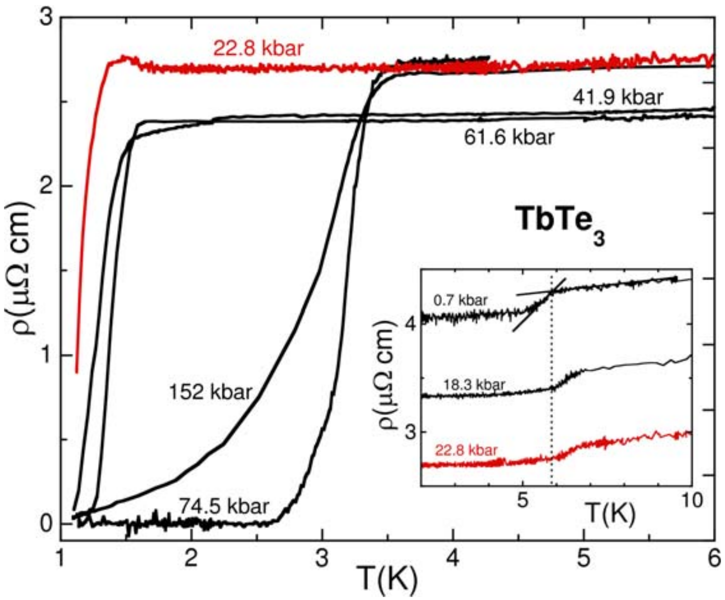
- T_{CDW1} decreases and T_{CDW2} increases with decreasing a
- Suggests similar dependence of T_{CDW1} and T_{CDW2} on P

□ ρ(T) measurements under P

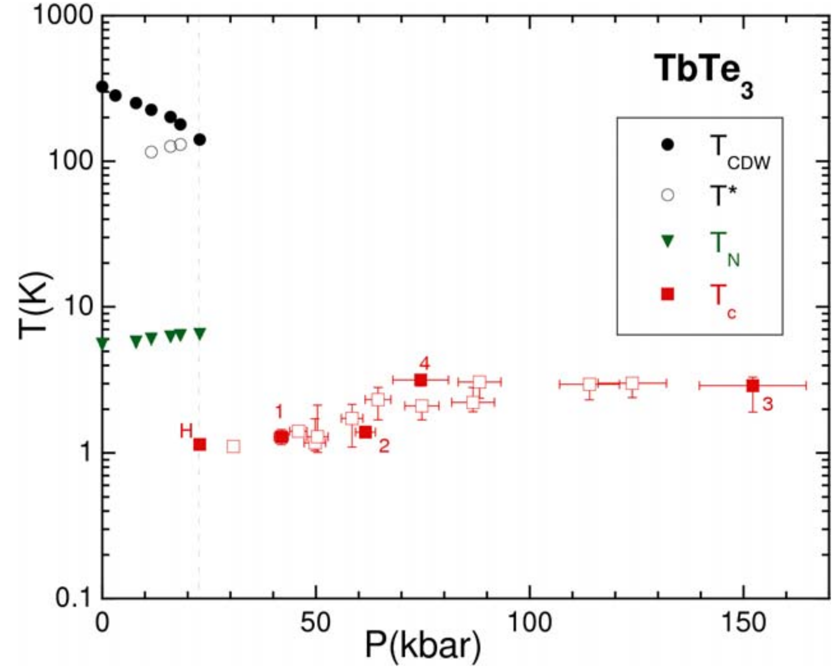
E. DiMasi, M. C. Aronson, J. F. Mansfield, B. Foran, S. Lee, PRB **52**,14516 (1995)
N. Ru, I. R. Fisher, PRB **73**, 033101 (2006)
J. Lavarock, S. B. Dugdale, Zs. Major, M. A. Alam, N. Ru, I. R. Fisher, G. Santi, E. Bruno, PRB **71**, 0851144 (2005)
N. Ru, J. -H. Chu, I. R. Fisher, PRB **78**, 012410 (2008)

SCing, CDW, and AFM order under pressure in $TbTe_3$



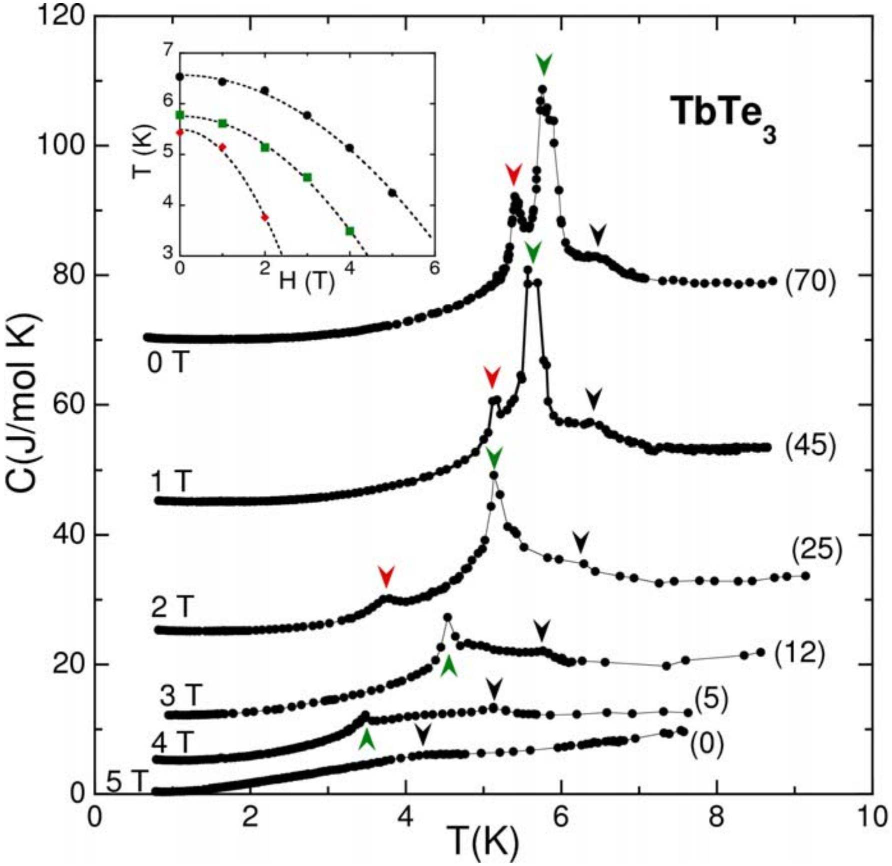


J. J. Hamlin, D. A. Zocco,
T. A. Sayles, M. B. Maple,
J.-H. Chu, I. R. Fischer, 08

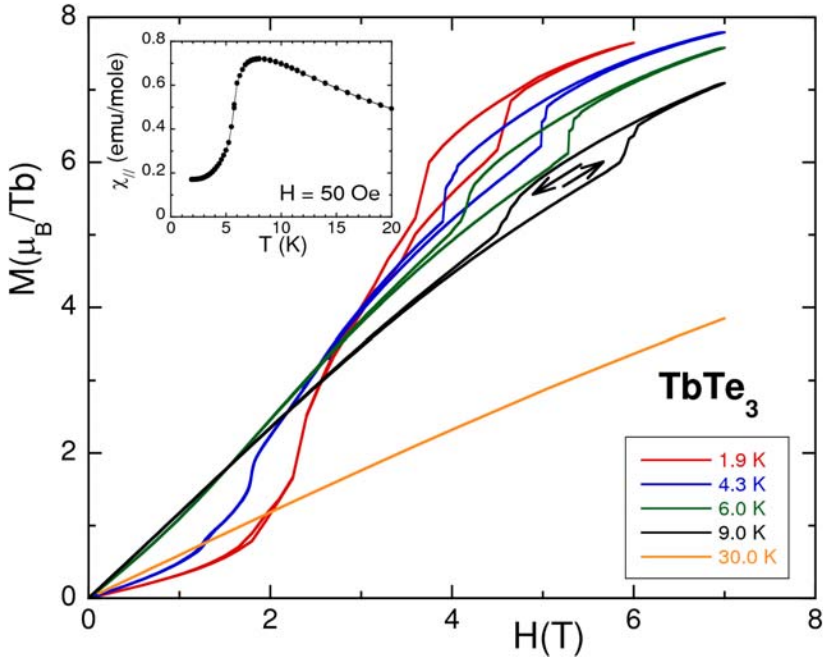


- Interplay of CDW, AFM & SC
- P-induced SC
- CDW, AFM, SC coexist (~23 kbar)
- Opportunity to study magnetically ordered SCs under P

Evidence for antiferromagnetic ordering of Tb^{3+} ions in $TbTe_3$



J. J. Hamlin, D. A. Zocco, T. A. Sayles, M. B. Maple, J.-H. Chu, I. R. Fischer, 08



Interplay of hidden, magnetic, and superconducting order in URu_2Si_2

Coworkers

University of California,
San Diego

N. P. Butch	*U. Maryland*
J. R. Jeffries	*Lawrence Livermore National Laboratory*
T. A. Sayles	*Quantum Design, San Diego*
B. T. Yukich	*UCSD, SOM*
D. A. Zocco	

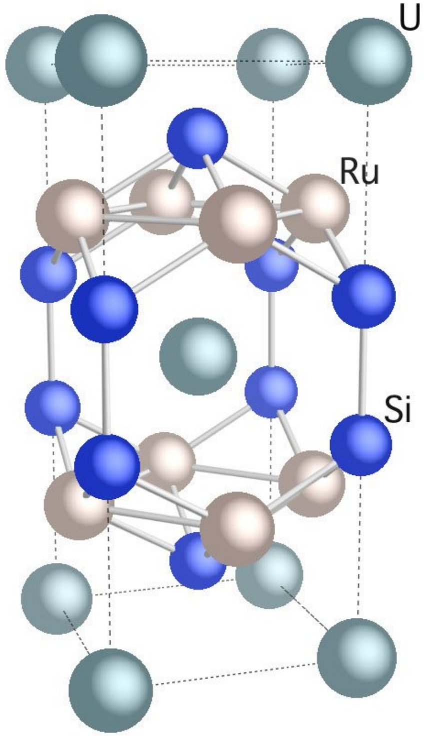
$URu_{2-x}Re_xSi_2$: Introduction

- Delicate interplay between competing interactions in URu_2Si_2 produces a wide variety of correlated electron phenomena
- Phenomena include
  - Superconductivity (SC)
  - A "hidden order" (HO) phase that apparently forms a gap over portion of Fermi surface (like SDW or CDW)
  - Antiferromagnetism (AFM)
  - Ferromagnetism (FM)
  - Heavy fermion (HF) behavior
  - Non-Fermi liquid (NFL) behavior
- Interactions "tuned" via P, H, chemical substitution
- Experiments on suppression of HO phase and onset of FM in URu_2Si_2 via Re substitution — objectives:
  - Investigate physical properties of single crystals of $URu_{2-x}Re_xSi_2$ ($0 \le x \le 1$)
  - Allows the anisotropy of physical properties to be determined — advantage over previous studies on polycrystalline specimens of $URu_{2-x}Re_xSi_2$
  - Establish T-x phase boundaries and locations of quantum phase transitions (QPTs) for HO and FM phases
  - Characterize NFL behavior associated with quantum critical points (QCPs)

URu_2Si_2: Initial experiments

- Heavy fermion superconductivity (polycrystalline specimens)
  Schlabitz, Baumann, Pollit, Rauchschwalbe, Mayer, Alheim, Bredl, ZP (86)
- Anisotropy of physical properties (single crystal specimens)
  Palstra, Menovsky, van den Berg, Dirkmaat, Kes, Nieuwenhuys, Mydosh, PRL (85)
- Partial gapping of the FS by CDW or SDW (polycrystalline specimens)
  Maple, Dalichaouch, Kohara, Rossel, Torikachvili, McElfresh, Thompson, PRL (86)
- SM-AFM – neutron scattering experiments (single crystal specimens)
  Broholm, Kjems, Buyers, Matthews, Palstra, Menovsky, Mydosh, PRL (86)
- Followed by an enormous amount of experimental and theoretical work, especially devoted to establishing the identity of the so-called "hidden order" phase
- Review some of history that led us to the Re substitution experiments in URu_2Si_2

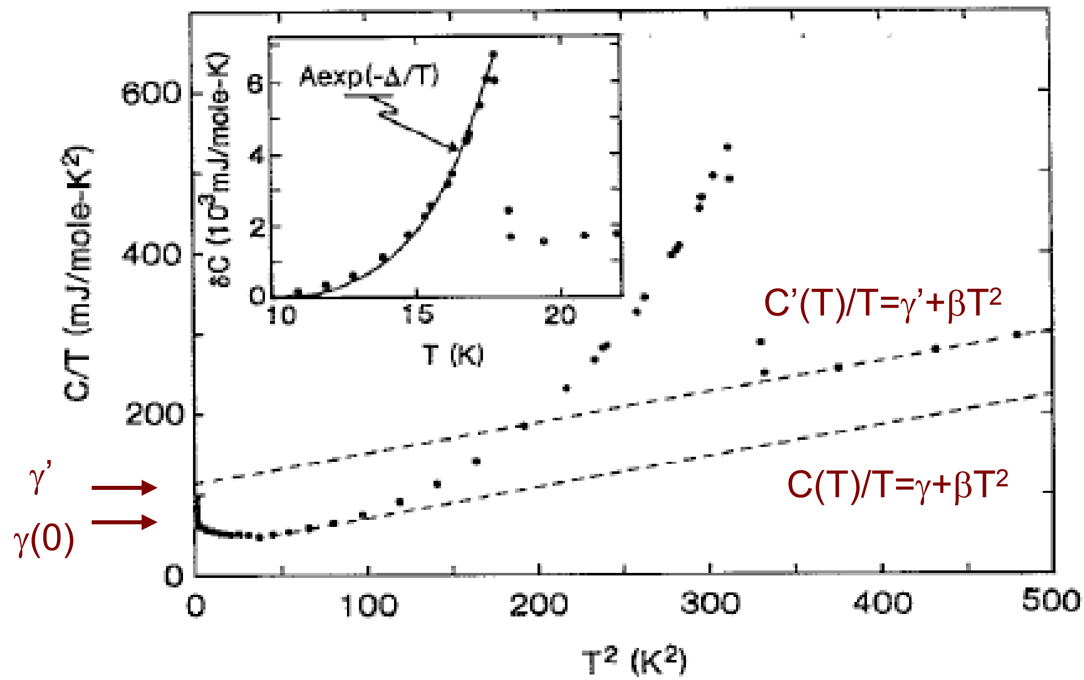
Why URu_2Si_2 is interesting



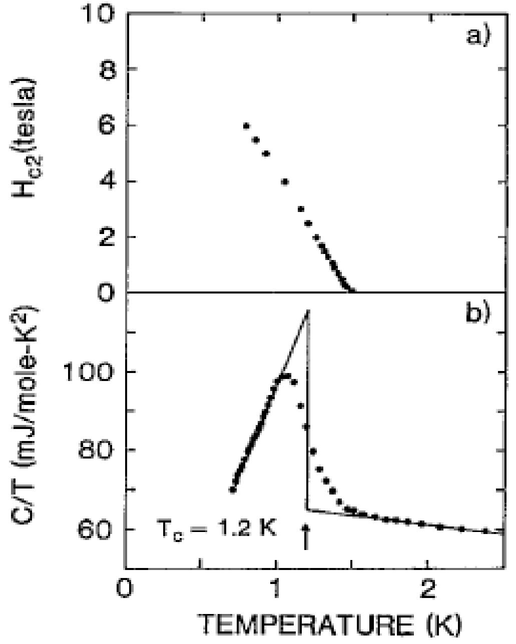
$ThCr_2Si_2$ structure
a = 4.13 Å, c = 9.58 Å

- Moderately heavy Fermi liquid ($m^* \sim 25\ m_e$)
- "Hidden order (HO)" phase ($T_0 \approx 17.5$ K)
  - BCS-like feature in C(T) at 17.5 K suggests partial gapping of Fermi surface by CDW or SDW
  - Small moment antiferromagnetism (SM-AFM): $\mu \approx 0.03\ \mu_B$/U, ||c-axis, (100) modulation
  - $\delta S \approx 0.2\ln(2)$ too large $\Rightarrow$ "HO" phase
  - Large moment antiferromagnetism (LM-AFM) observed at $P_c \sim 5 - 15$ kbar: $\mu \sim 0.4\ \mu_B$/U
  - SM-AFM phase – small volume fraction of LM-AFM phase that coexists with HO phase $\Rightarrow \mu_{av} \approx 0.03\ \mu_B$
  - Is HO/LM-AFM phase separation due to strain?
- Superconductivity (SC) ($T_c \approx 1.5$ K)
  - Coexists with HO and SM-AFM phases
- Ordered phases can be "tuned" with P, H, x:
  - Produces LM-AFM (Rh) and LM-FM (Re) phases
  - Non-Fermi liquid (NFL) behavior

Low temperature specific heat of URu_2Si_2



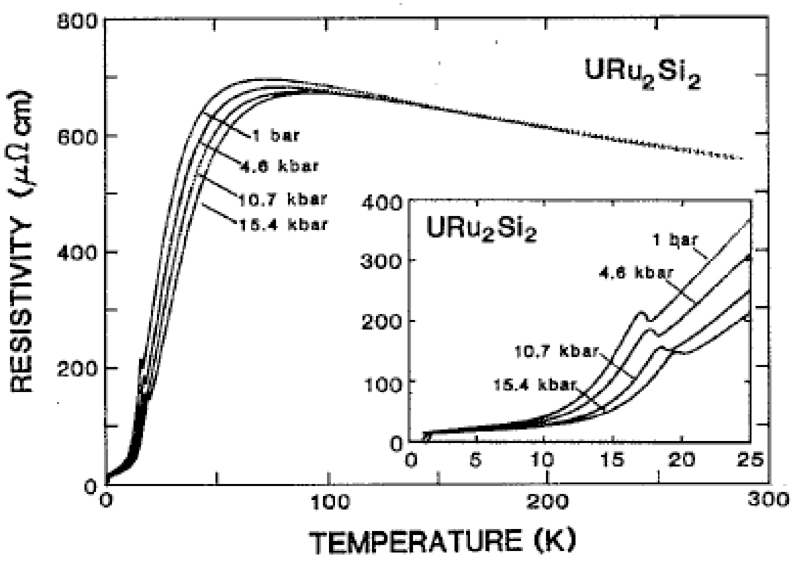
Maple, Dalichaouch, Kohara, Rossel, Torikachvili, McElfresh, Thompson, PRL (86)

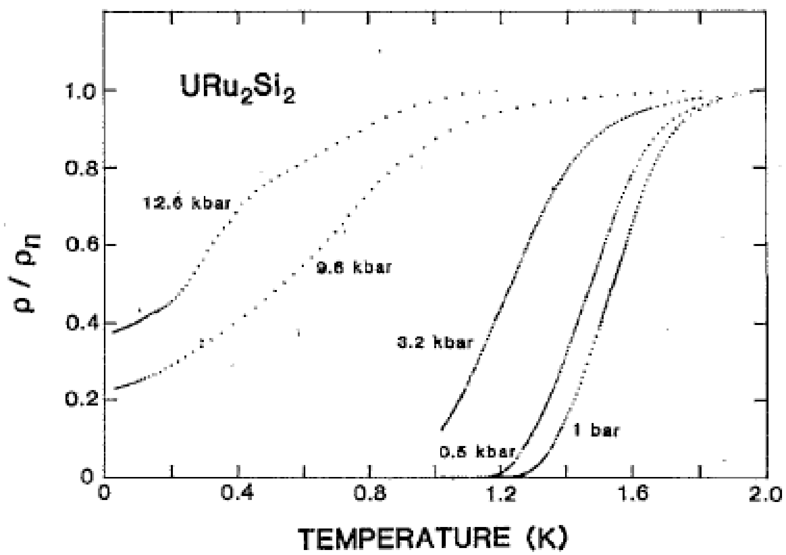


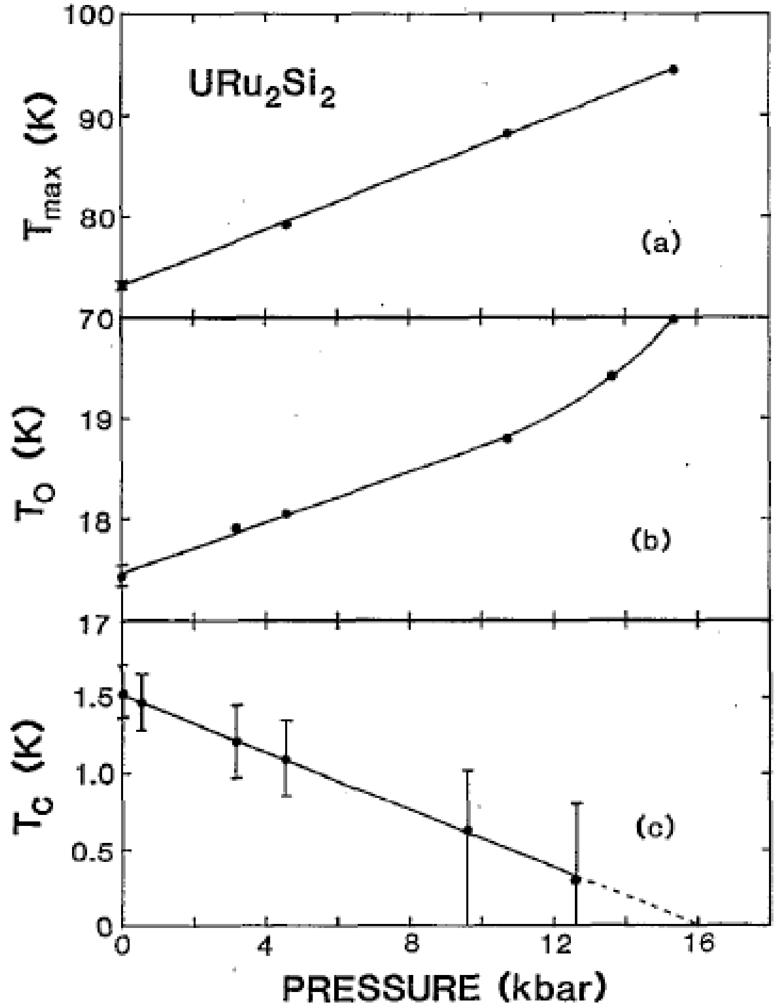
SCing transition

- BCS-type mean field transition at T_o = 17.5 K
  - $\delta C \approx A\exp(-\Delta/T)$; $\Delta \sim 10^2$ K ⇒ SDW or CDW
  - $\gamma(0)/\gamma' \approx 0.6$ ⇒ ~ 40 % Fermi surface removed by SDW or CDW
  - SC & SDW or CDW compete for Fermi surface!
- $\delta S \approx 0.2\ln(2)$ too large for AFM with small $\mu \approx 0.03\ \mu_B$ ⇒ Hidden order (HO)?
- Superconductivity below $T_c \approx 1.5$ K (onset)

Effect of pressure on competing electronic states in URu_2Si_2



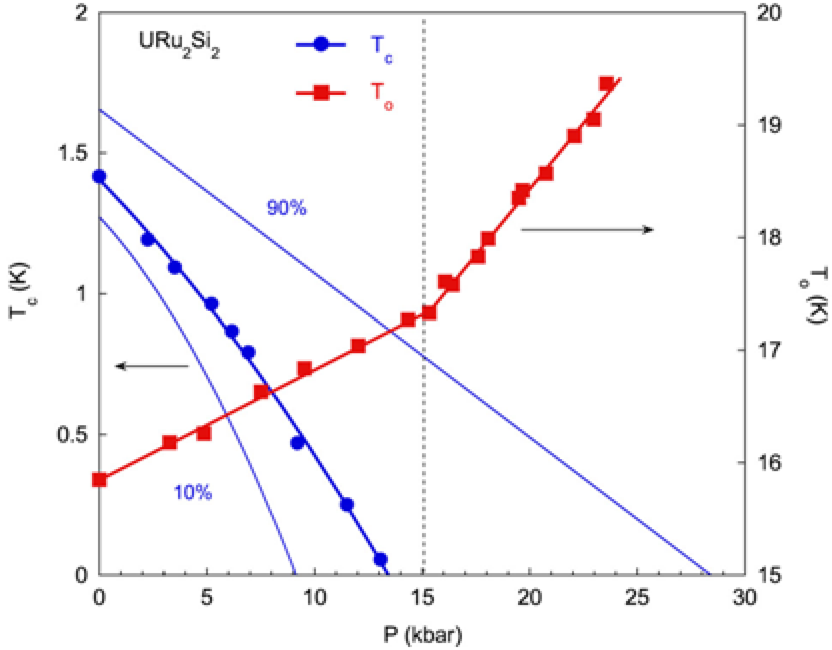


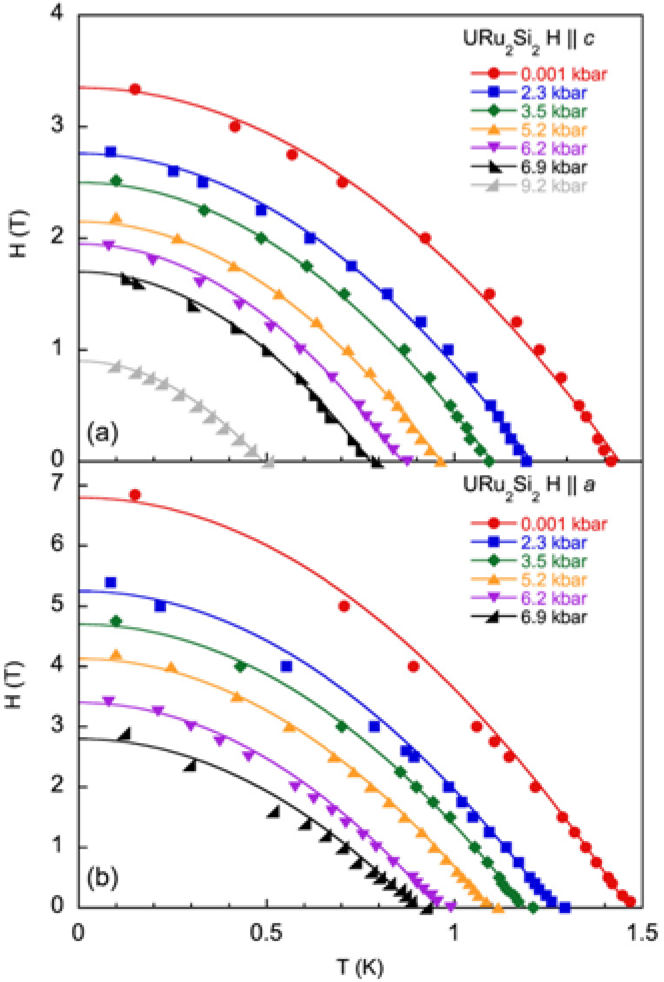


McElfresh, Thompson, Willis, Maple, Kohara, Torikachvili, PRB (87)

Superconductivity under pressure – $T_c(P)$ & $H_{c2}(T,P)$

URu_2Si_2 single crystal



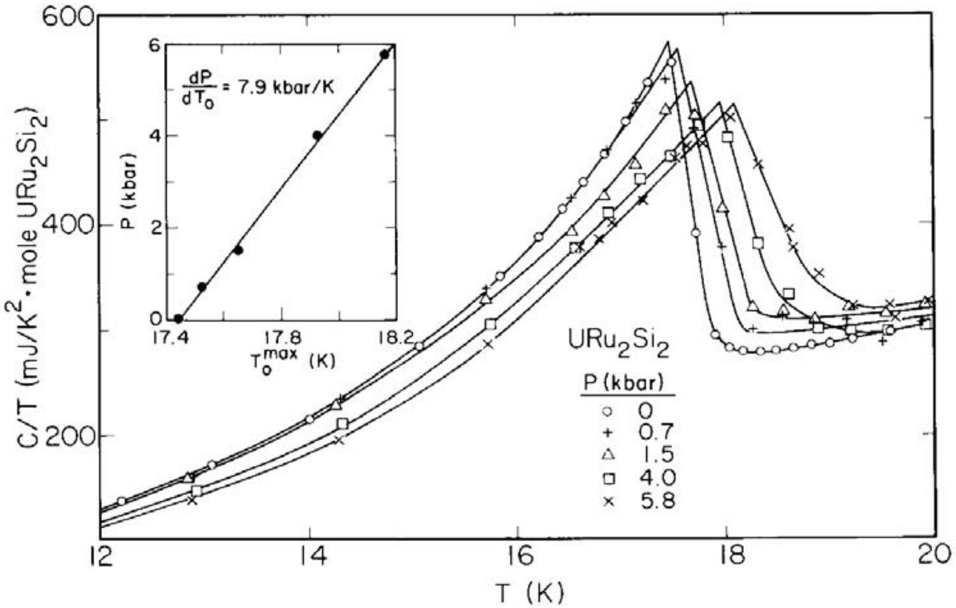


Phenomenological fits to $H_{c2}(T)$ data:
$H_{c2}(T) = H_{c2}(0)[1 - A(T/T_c)^2]$

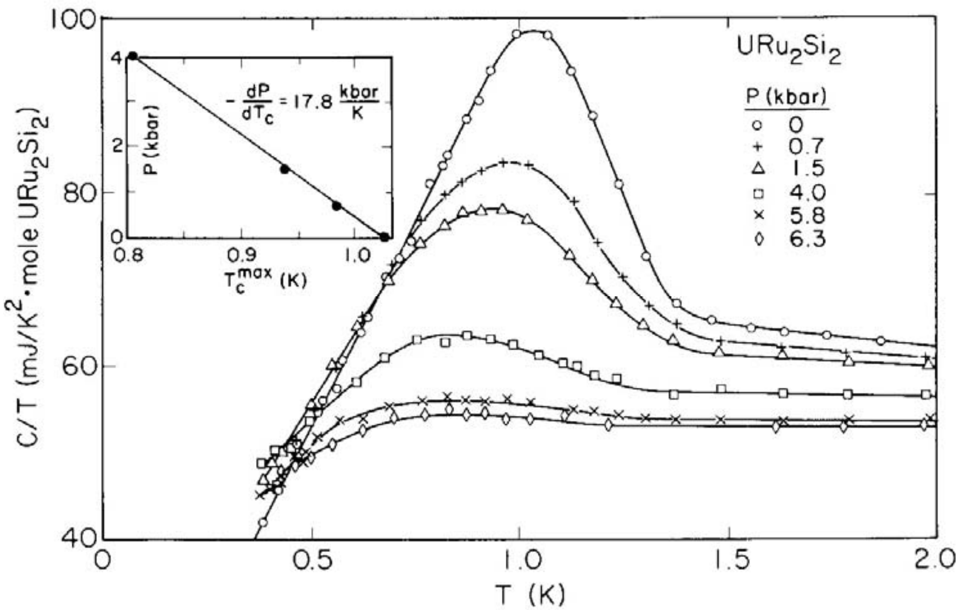
No features in $T_c(P)$ and $H_{c2}(T,P)$ curves to ~15 kbar ⇒
no qualitative change in SC due to onset of LM-AFM phase near 5 kbar!

J. R. Jeffries, N. P. Butch, B. T. Yukich, M. B. Maple, PRL (07)

Low temperature specific heat of URu_2Si_2 under pressure



R. A. Fisher, S. Kim. Y. Wu,
N. E. Phillips, M. W. McElfresh,
M. S. Torikachvili, M. B. Maple (90)



NOTE: Rapid reduction of specific heat jump ΔC at T_c with P

Two possible interpretations:

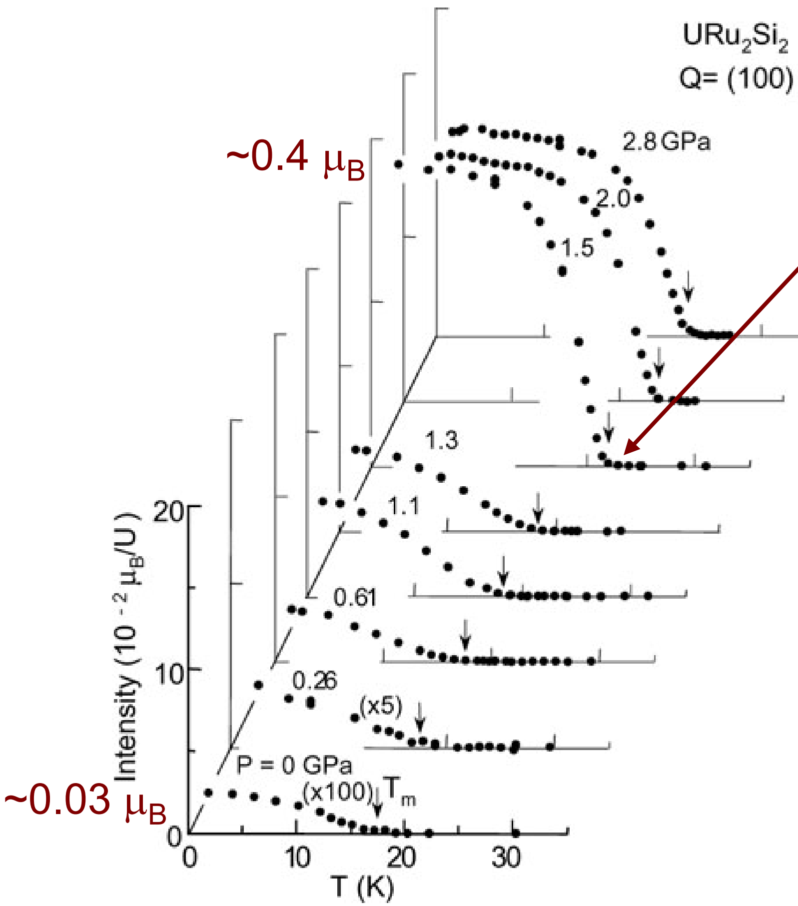
(1) Suppression of SCing volume fraction to immeasurably small value by ~ 5 kbar;

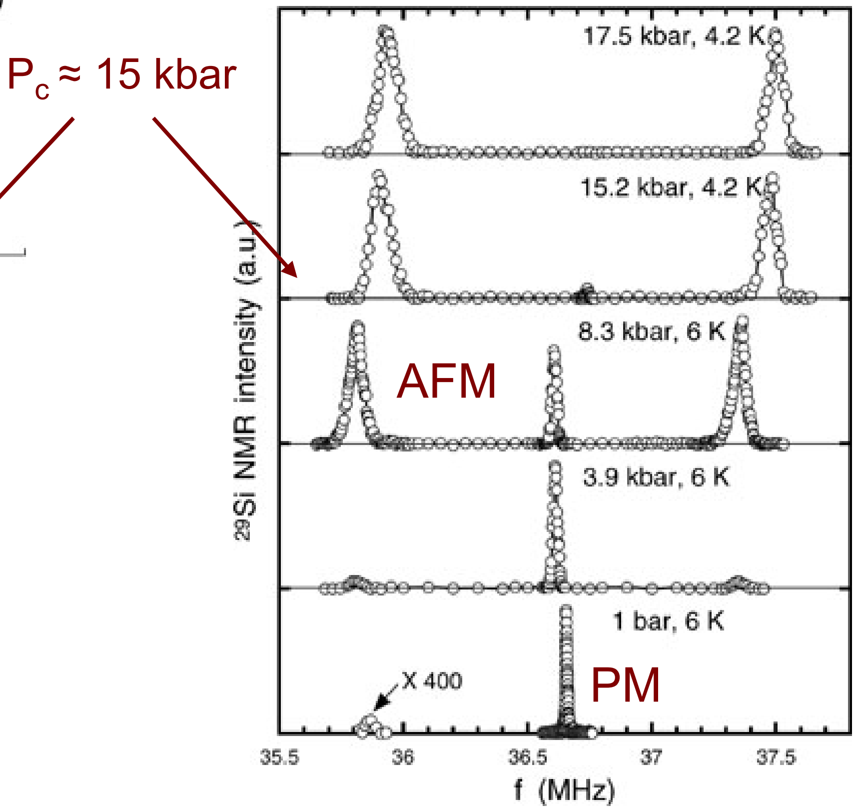
(2) Manifestation of unconventional nature of SC

URu_2Si_2: HO – LM-AFM phase transition under P

Neutron diffraction: AFM μ increases with P

^{29}Si NMR: phase separation – AFM volume increases with P (HO volume decreases with P)

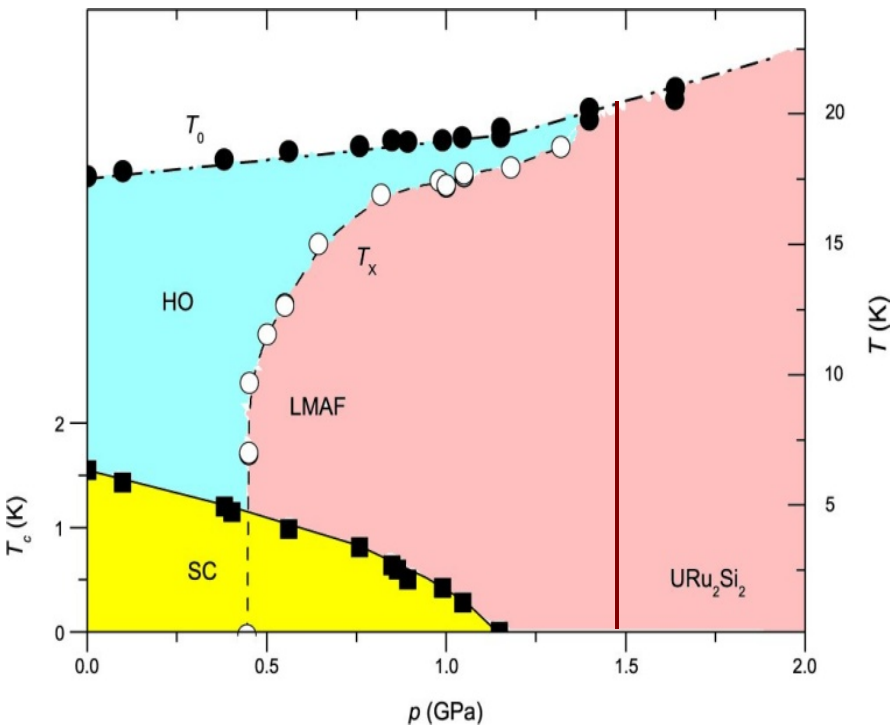




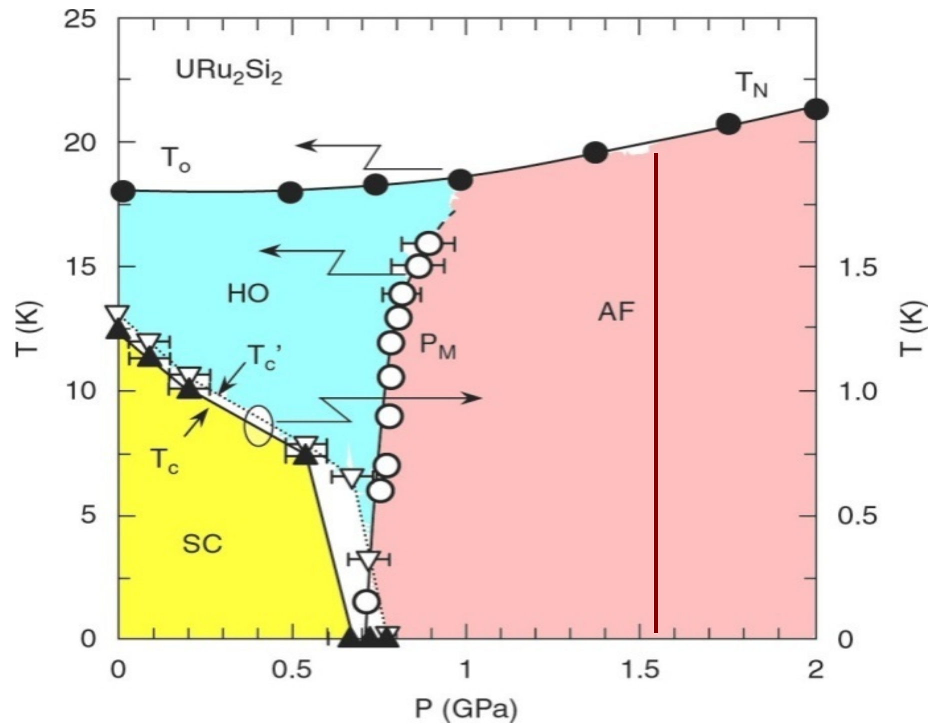
H. Amitsuka et al., PRL (99)

K. Matsuda et al., JP:CM (03)

URu_2Si_2: Possible T-P phase diagrams



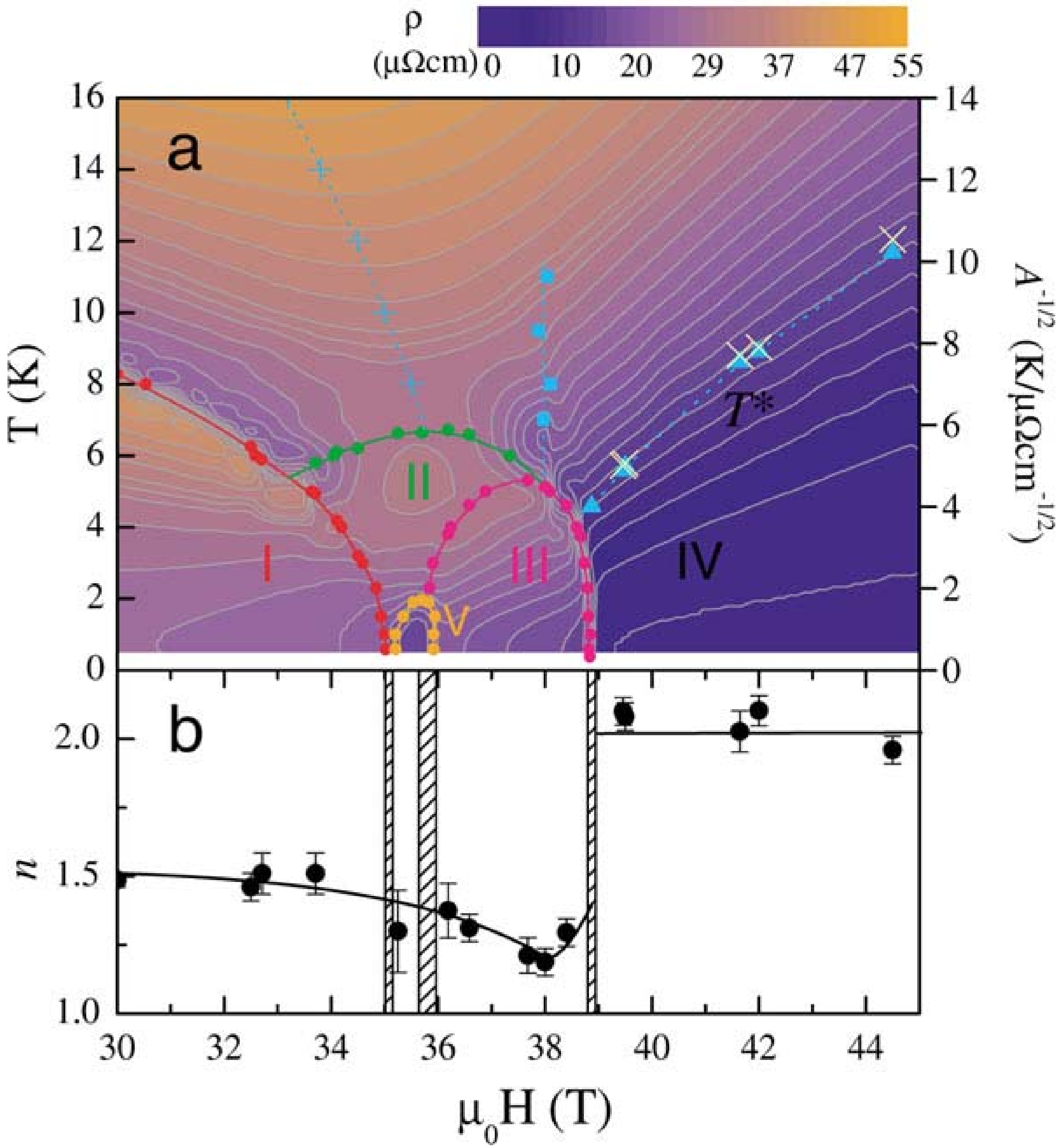
Knebel et al., JMMM (07)



Amitsuka et al., JMMM (07)

- Left hand T – P phase diagram favored by continuous behavior of T_c(P) and H_{c2}(T,P)
- Right hand T – P phase diagram could be favored if rapid diminution of ΔC at T_c near 6 kbar signals loss of bulk SC
- There appears to be a steep (vertical?) phase boundary at $P_c \approx 15$ kbar (nearly independent of T - red line in figures)
- HO – LM-AFM phase boundary or another phase boundary?

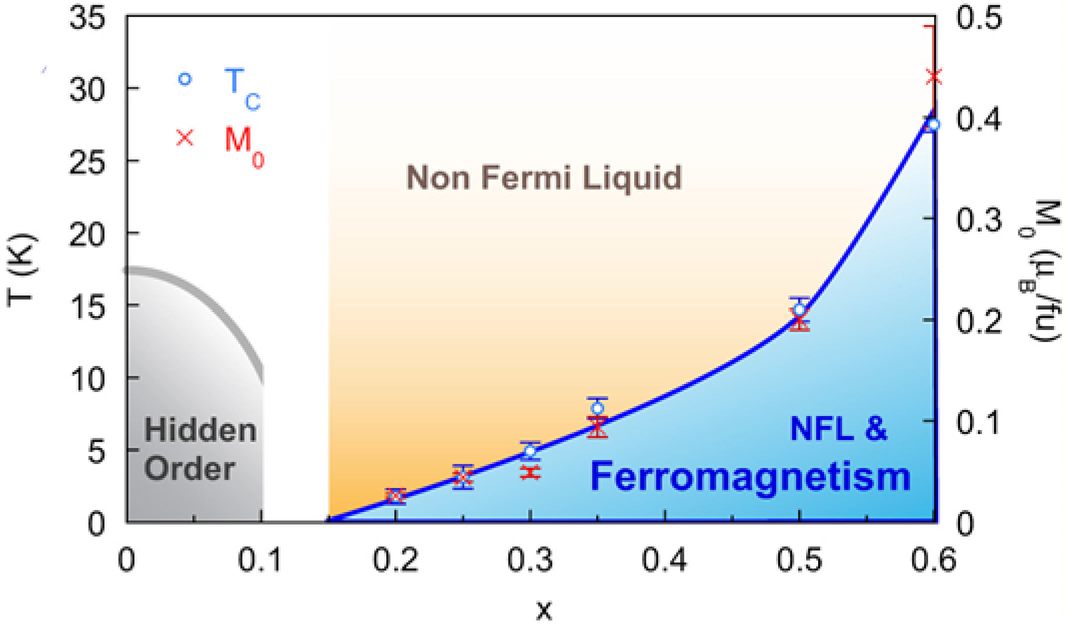
Field-induced phases in URu_2Si_2

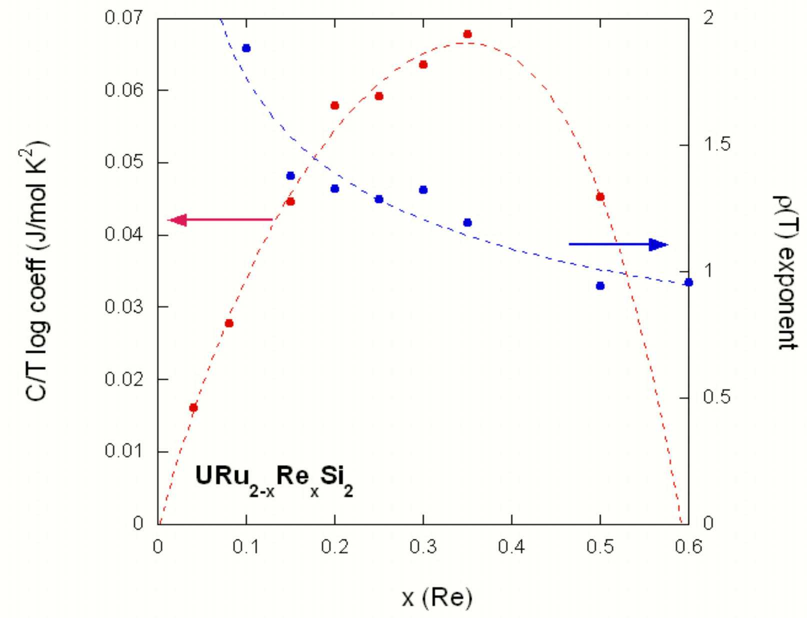


Electrical resistivity
$\rho = \rho_o + AT^n$
n = 2: FL
n ≈ 1 - 1.5: NFL

*K. H. Kim et al., PRL **91** '03*

$URu_{2-x}Re_xSi_2$: FM and NFL behavior

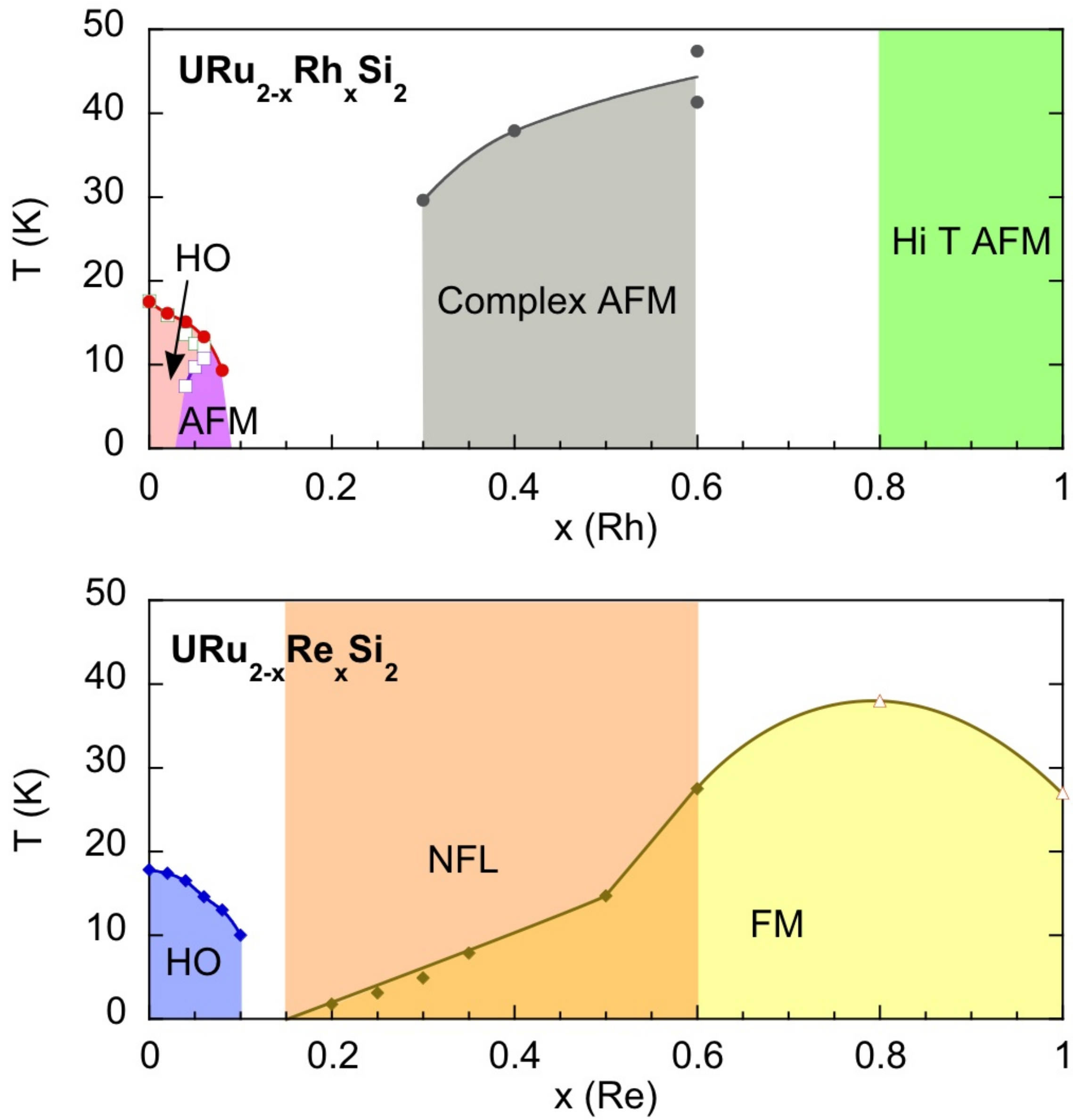




Relation to NFL?

- NFL features in ρ(T), C(T) not clearly peaked at x = 0.15
- Extend deep into FM state
- 3D itinerant FM QCP:
  - $\rho(T) \sim T^{5/3}$
  - $C(T)/T \sim -\ln T$
  - $\chi(T) \sim T^{3/4}$
  - $T_C \sim (x - x_c)^{3/4}$
- Quantum Griffith's phase scenario:
  - $\chi(T) \sim C(T)/T \sim T^{\lambda-1}$

Comparison of $URu_{2-x}M_xSi_2$ (M = Rh, Re) phase diagrams



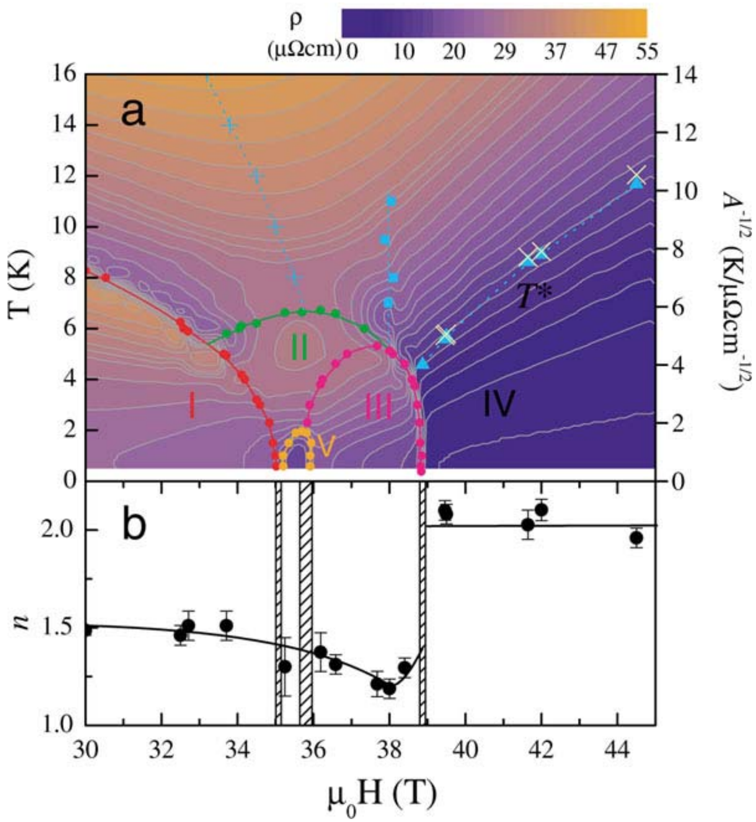
Kawarazaki et al., JPSJ (94)
Yokoyama et al., JPSJ (04)

This work

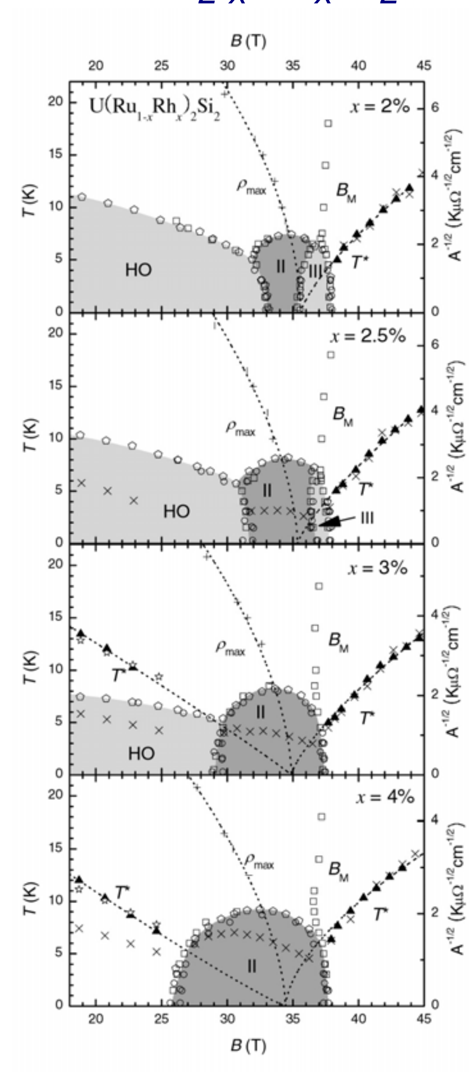
Superconductivity in pure elemental solids

Field-induced phases in pure and Rh substituted URu_2Si_2

URu_2Si_2



$URu_{2-x}Rh_xSi_2$



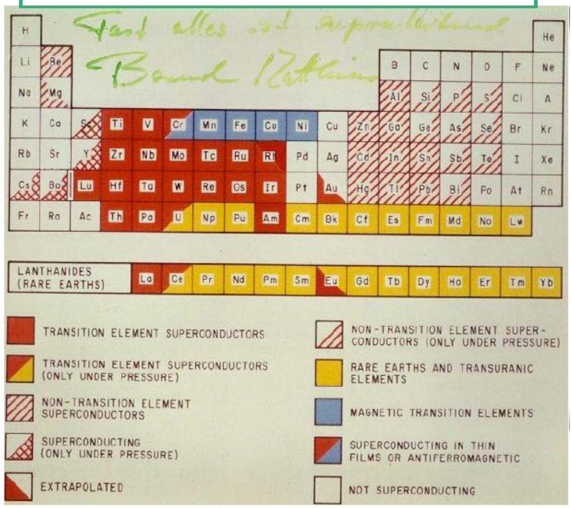
K. H. Kim et al., PRL (03)
K. H. Kim et al., PRL (04)

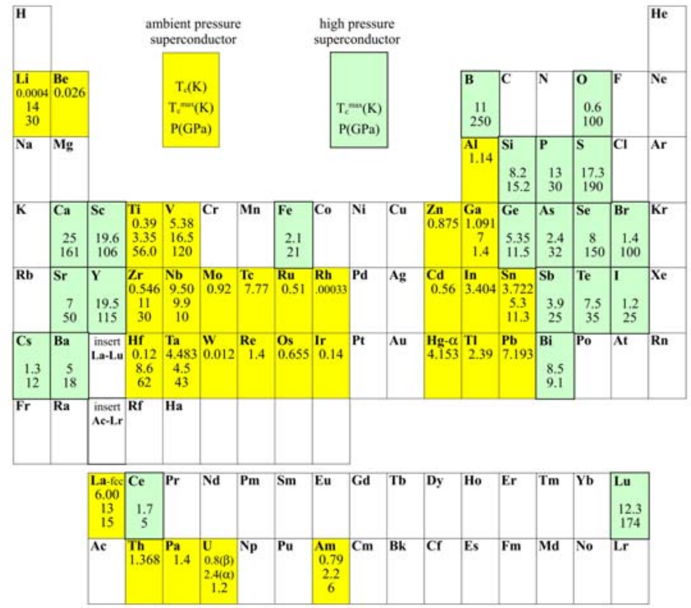
$URu_{2-x}Re_xSi_2$
S. Francoul et al., (09); this conference

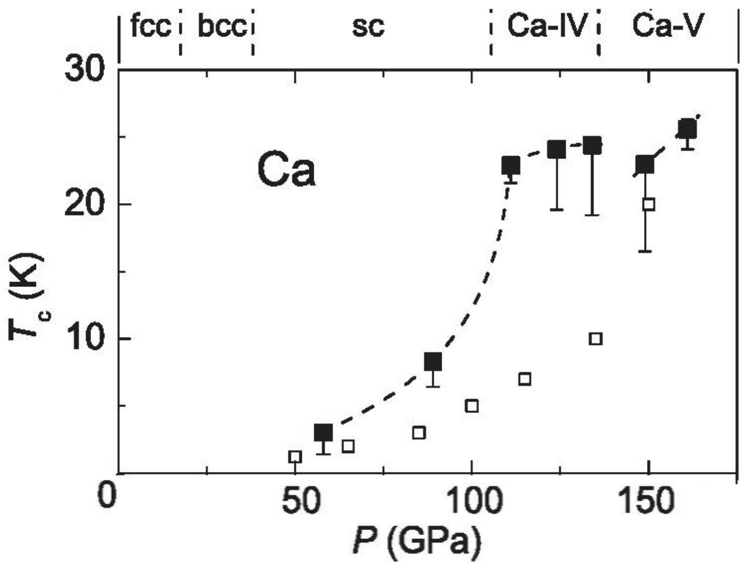
Electrical resistivity
$\rho = \rho_o + AT^n$
n = 2: FL
n ≈ 1 - 1.5: NFL

Superconductivity in pure elemental solids

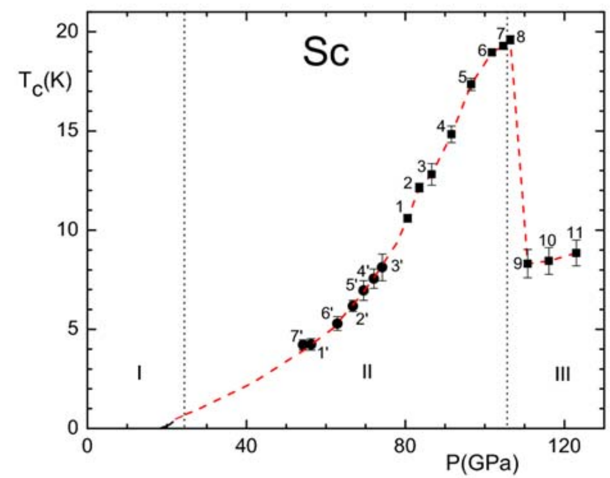
Periodic Table of Superconductivity in the 1970's
"Fast alles ist supraleitend"
Bernd Matthias







T. Yabuuchi, T. Matsuoka, Y. Nakamoto, K. Shimizu
*J. Phys. Soc. Japan, **75**, 083703 (2006)*



M. Debessai, J. J. Hamlin, J. S. Schilling
arXiv:0806.3407 (submitted to PRB)

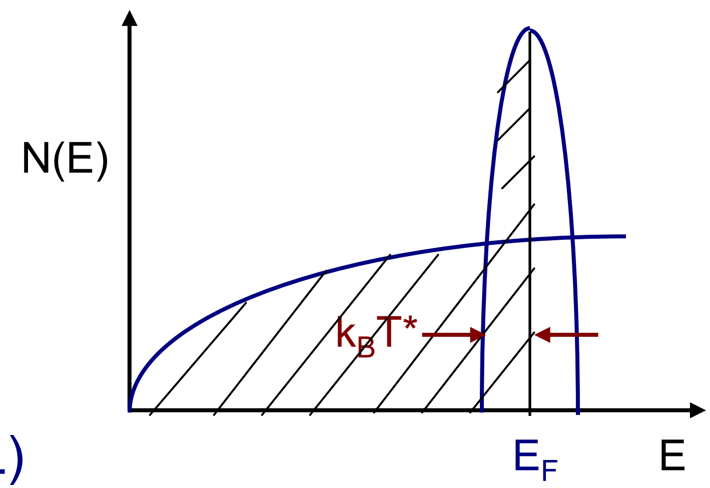
END

Heavy fermion f-electron materials

- Metallic compounds containing Ln & A ions with partially-filled f-electron shells & unstable valence (no. of f-electrons)
- Lanthanides – Ce, Pr, Yb; actinides – U, Pu
- Hybridization between localized f- & conduction-electron states
- Narrow resonance near the E_F
- Effective Fermi temperature – T*
- T >> T*: local moment behavior

$$\chi(T) \sim N\mu_{eff}^2/3k_B(T+T^*)$$

$$\rho(T) \sim -\ln T$$



- T << T*: nonmagnetic heavy Fermi liquid (FL)

$$\chi(T) \to \chi_o \propto m^* \propto 1/T^*$$

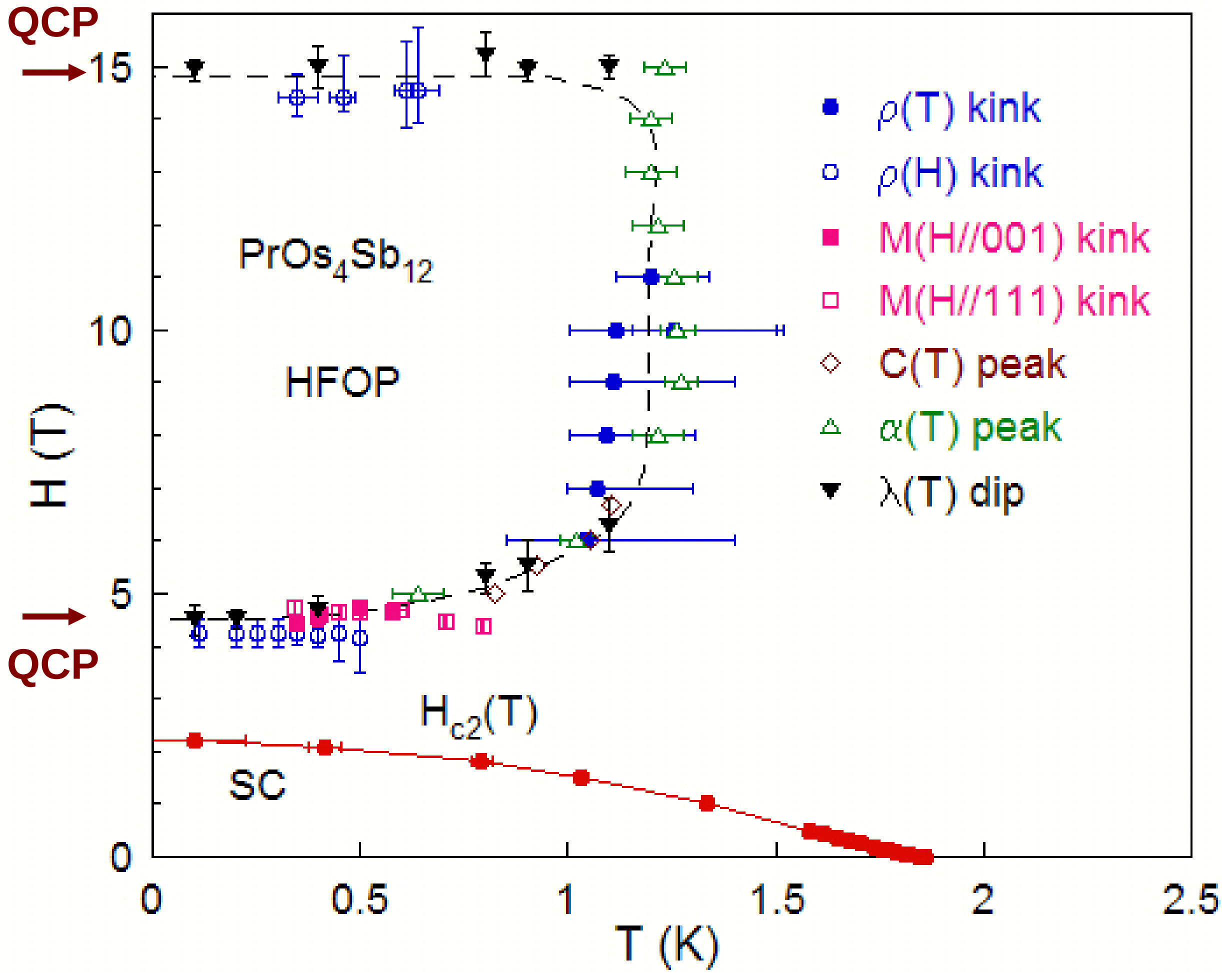
$$\gamma(T) = C_e(T)/T \to \gamma_o \propto m^* \propto 1/T^* \sim \text{several J/mol K}^2!$$

$$R = (\chi_o/\mu_{eff}^2)/(\gamma_o/\pi^2 k_B^2) \approx 1 \text{ (Wilson-Sommerfeld ratio)}$$

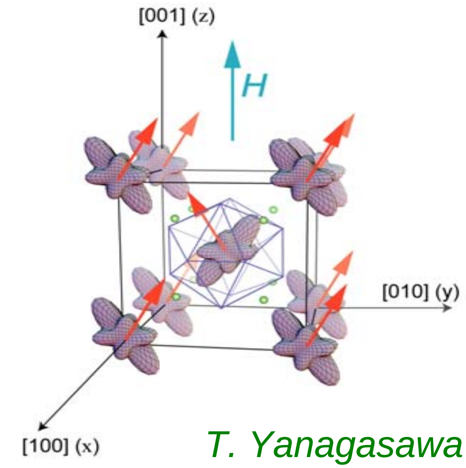
$$\rho(T) \propto \rho_{e-e}(T) \sim AT^2 \text{ with } A \sim \gamma_o^2$$

- Heavy FL unstable to unconventional SC & magnetic order (RKKY)

H–T phase diagram of $PrOs_4Sb_{12}$



Ho et al., PRB (03)



T. Yanagasawa (06)

HFOP

- Related to crossover of CEF energy levels
- Identified with antiferro-quadrupolar order: neutron diffraction *Kohgi et al., JPSJ (03)*
- Anisotropic phase boundary: M(H,T) *Tayama et al., JPSJ (03)*
- SC in vicinity of antiferro-quadrupolar QCP!

Why $PrOs_4Sb_{12}$ is interesting

- 1^{st} Pr-based heavy fermion superconductor (T_c = 1.85 K)
  Bauer, Frederick, Ho, Zapf, Maple, PRB (02)
  (All others based on Ce, U, & Pu)
- Nonmagnetic heavy Fermi liquid ($m^* \approx 50\ m_e$)
- Unconventional strong coupling superconductivity:
  - May consist of several distinct SCing phases
  - Evidence for nodes in energy gap – *Chia et al., PRB (03)*
  - Breaks time reversal symmetry – *Aoki et al., PRL (03)*
  - Triplet spin superconductivity?
- High field ordered phase (HFOP): ρ(H,T) – *Maple et al., JPSJ (02)*;
  Ho et al., PRB (03); C(H,T) – *Aoki et al., JPSJ (02); Vollmer et al., PRL (03)*;…
- HFOP identified with antiferroquadrupolar order, based on neutron diffraction at high H: *Kohgi et al., JPSJ (03)*
- SC appears to be near quadrupolar quantum critical point (QCP)
- Formation of heavy Fermi liquid &/or unconventional SCing state may involve <u>electric</u> <u>quadrupole</u>, rather than <u>magnetic</u> <u>dipole</u>, fluctuations
- Off-center rattling & tunneling – *Goto et al., PRB (04)*
- Multiband superconductivity – *Seyfarth et al., PRL (05)*

Heavy fermion superconductivity in $PrOs_4Sb_{12}$

Normal state:

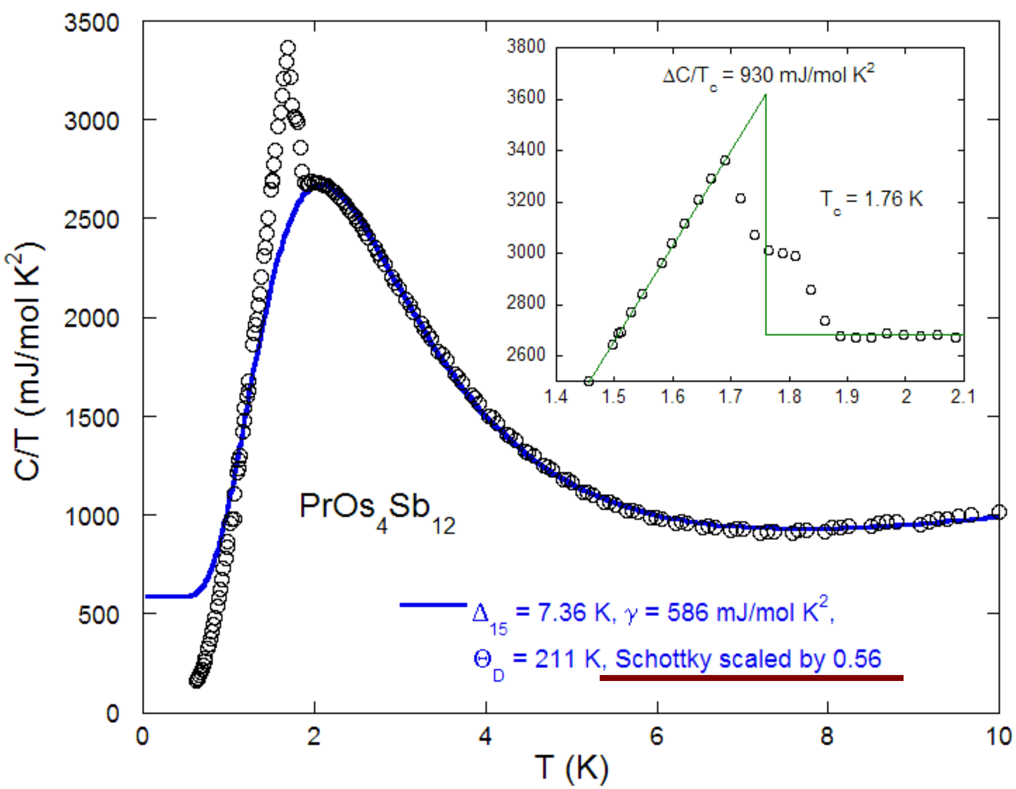
$C(T) = \gamma T + \beta T^3 + C_{Sch}(T)$

$\gamma \approx 500$ mJ/mol K^2, $\theta_D \approx 200$ K

Γ_1 singlet g.s., Γ_5 triplet 1st e.s., $\Delta \approx 7$ K

$C_{Sch}(T)$ scaled by 0.56

$\Rightarrow$ ~50% entropy $\rightarrow$ conduction electrons



Superconducting state:

- Analyses of ΔC & slope of $H_{c2}(T)$ at T_c confirm large value of γ derived from C(T) in normal state
- Structure in C(T) near T_c:

  Two SCing phases?

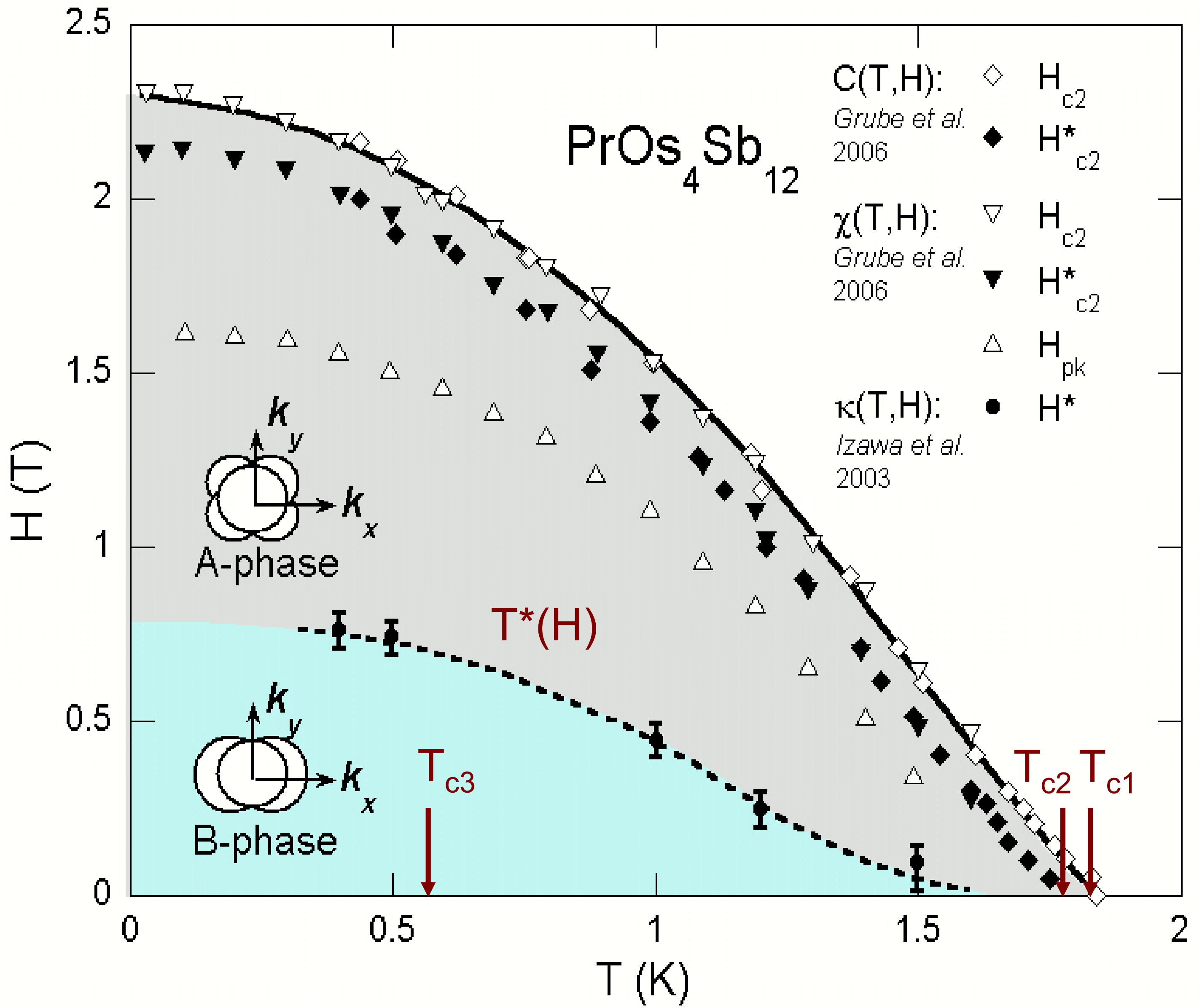
  $T_{c1} \approx 1.85$ K, $T_{c2} \approx 1.70$ K

  Intrinsic effect?

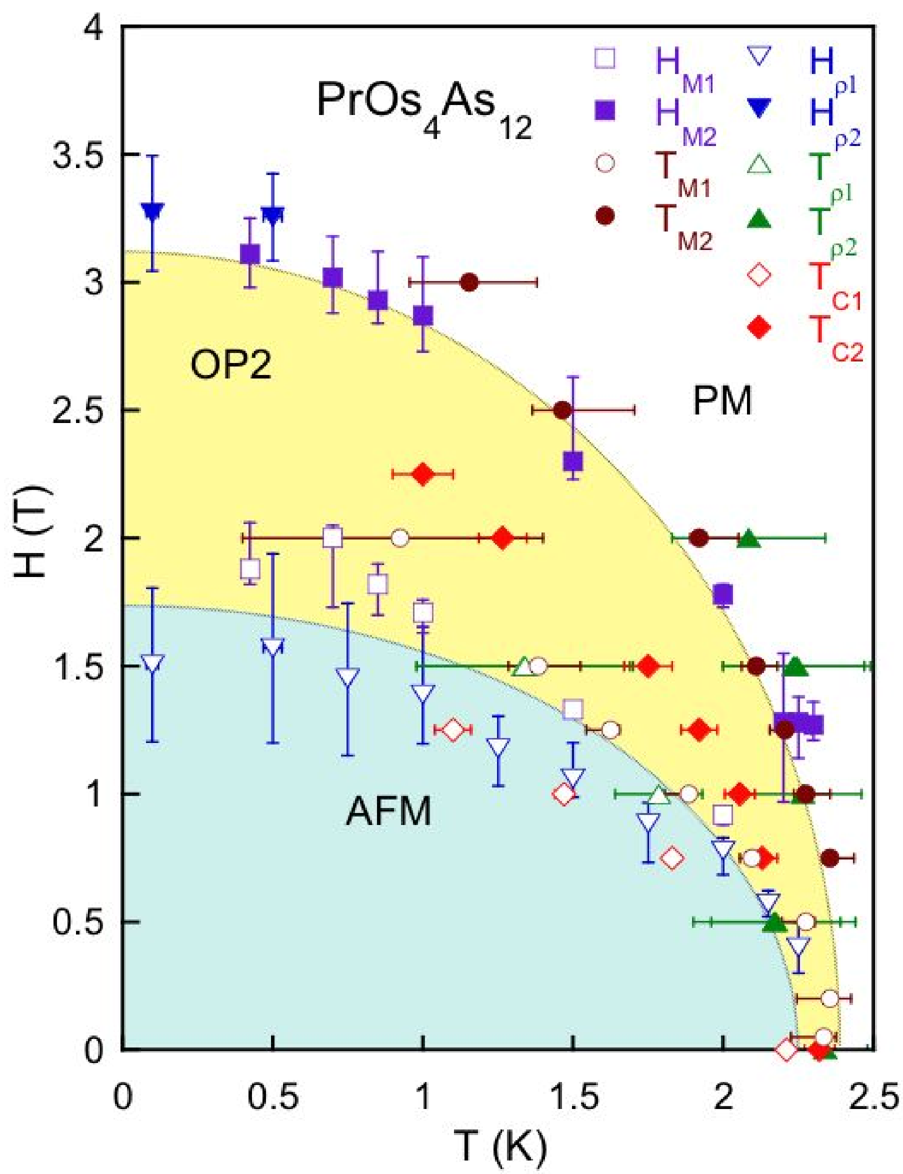
  Measson et al., PRB (05)

Bauer, Frederick, Ho, Zapf, Maple, PRB (02); Maple et al., JPSJ (02)

$PrOs_4Sb_{12}$: Possible superconducting phase diagram



$PrOs_4As_{12}$: H-T phase diagram

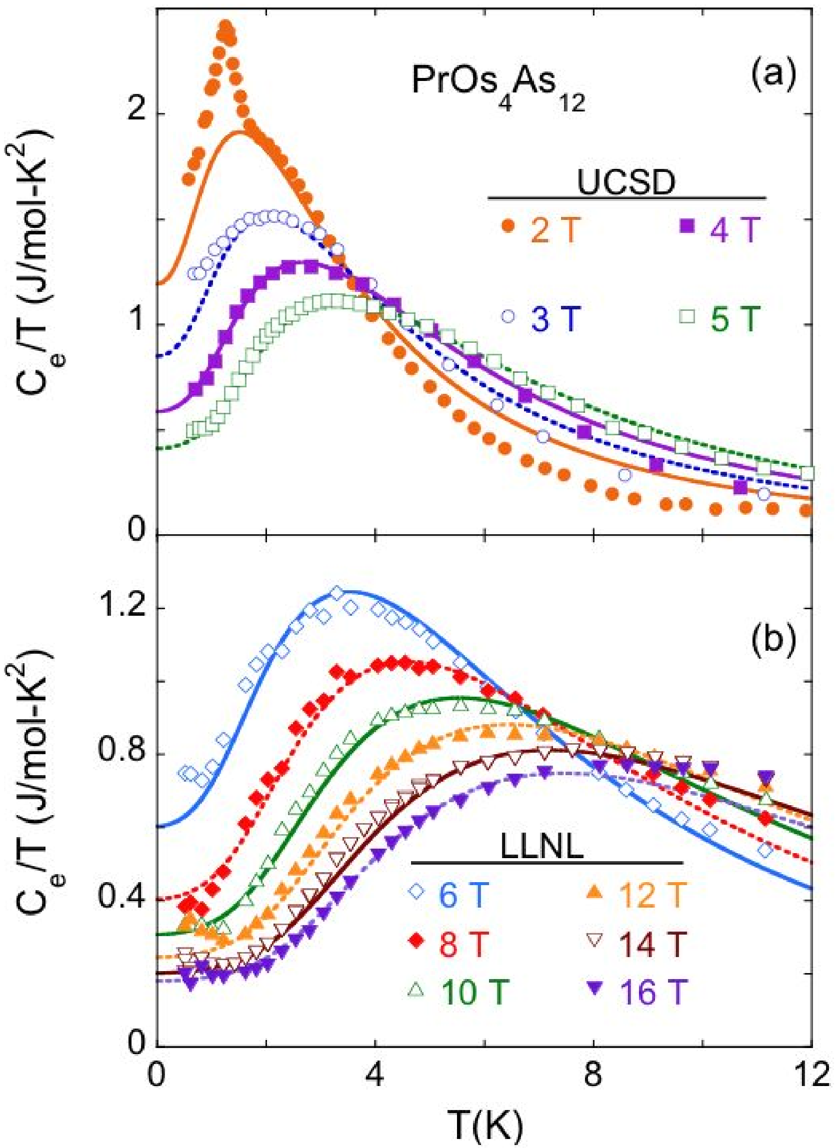


- Single crystals: *Z.Henkie*
- Kondo lattice:
  T_K ~ 1 K
- Heavy fermion behavior:
  γ ~ 1 J/mol-K^2
- Two ordered phases:
  AFM: Low H
  Unknown: High H

<u>AFM structure</u>: Alternating (100) planes of FMically ordered Pr μ's, oppositely oriented with one another

Yuhasz et al., PRB (06); Maple et al., PNAS (06)

$PrOs_4As_{12}$: C_e/T vs T in magnetic fields



- $C_e(T)$ data:

  UCSD ($0 \le H \le 5$ T)

  LLNL ($6\ T \le H \le 16$ T)

- $C_e(T)$ fits:

  Resonance level model

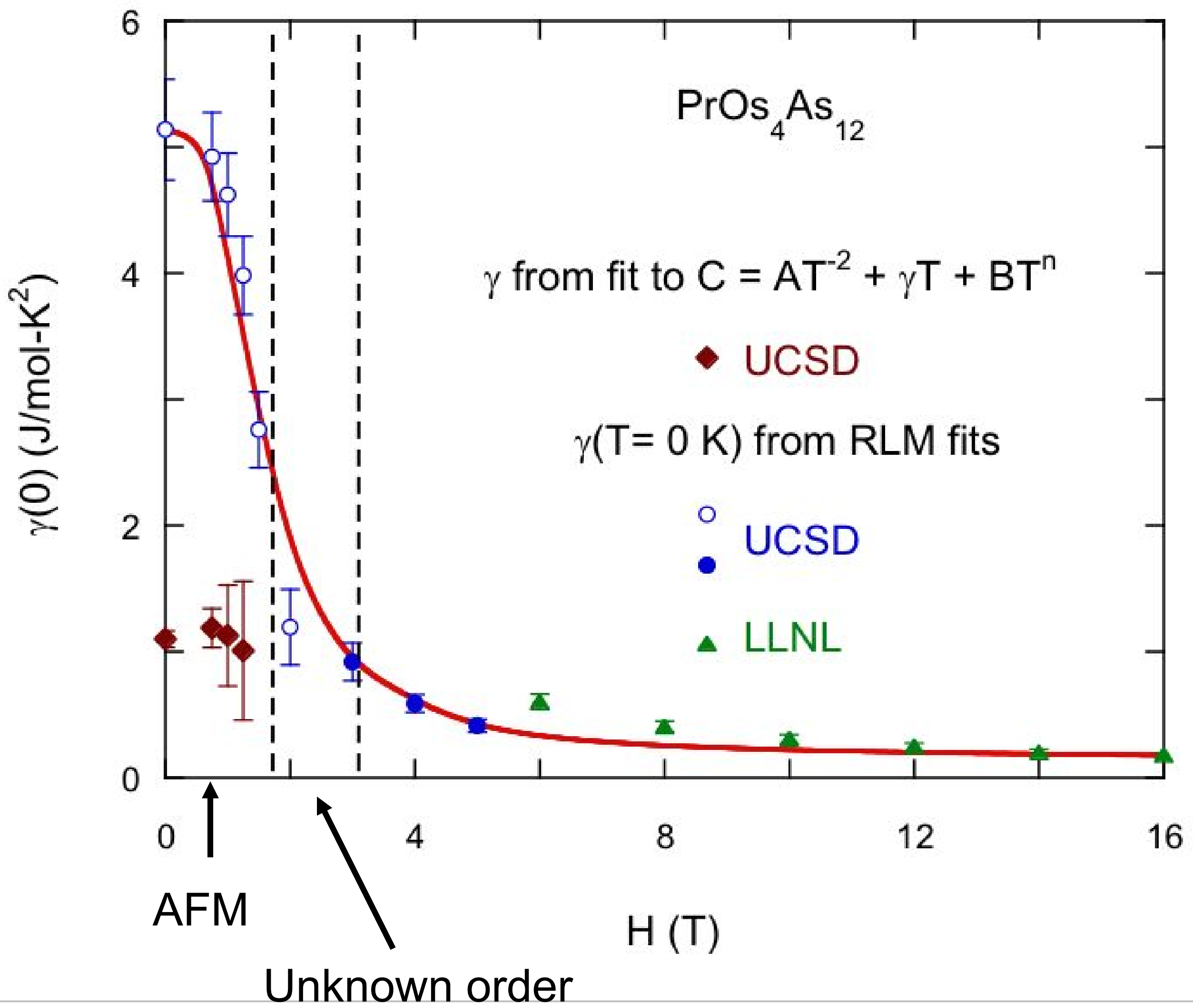
  K. D. Schotte, U. Schotte, Phys. Lett. (75)

  - Spin: S = 1 (triplet CEF g.s.)
  - Level width: $\Delta = k_B T_K \approx 3.5$ K
  - Zeeman splitting of resonances: $g\mu_B H$

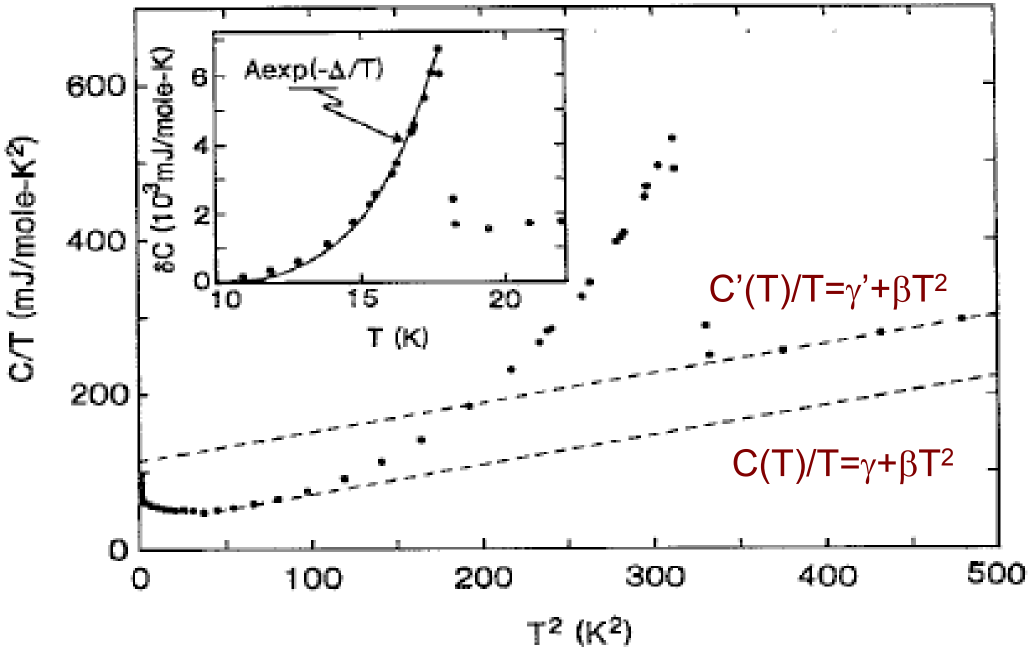
- Similar to behavior of $PrFe_4P_{12}$

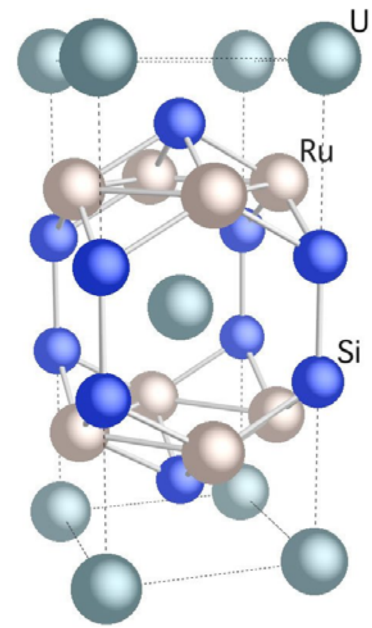
  Aoki et al., PRB '02

$PrOs_4As_{12}$: $\gamma(0)$ vs H



Interplay of hidden, magnetic, and superconducting order in URu_2Si_2

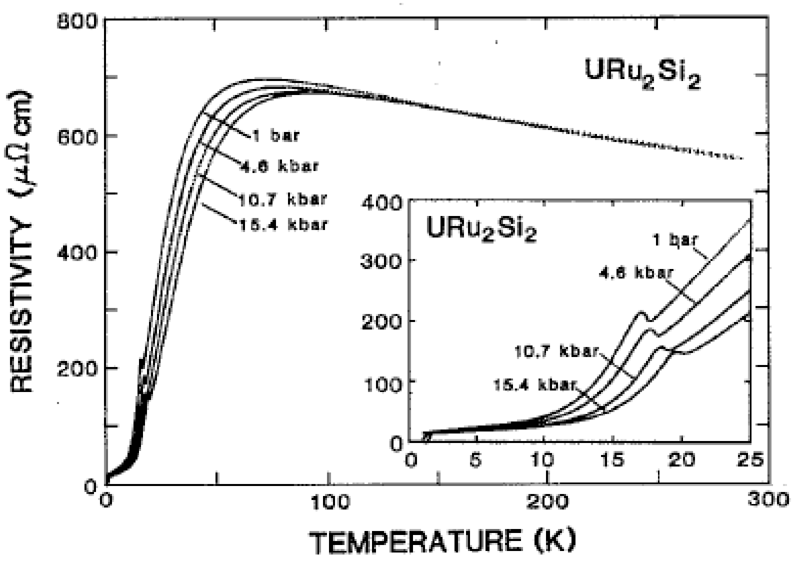


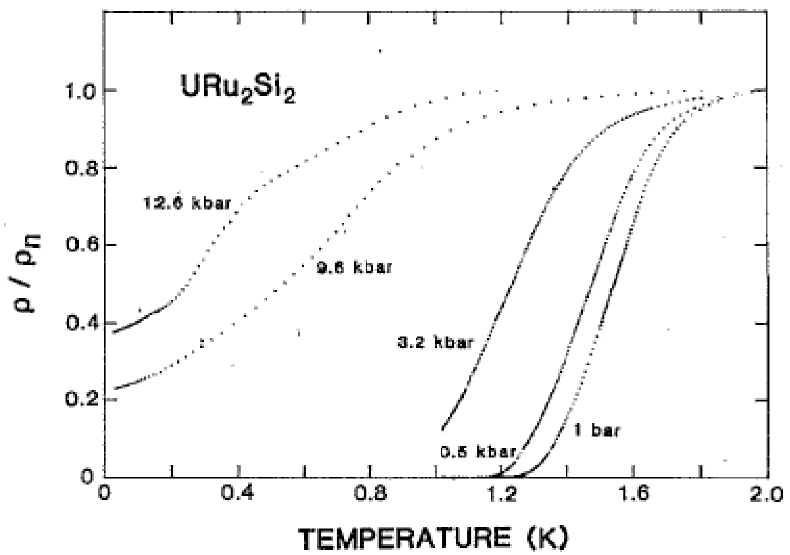


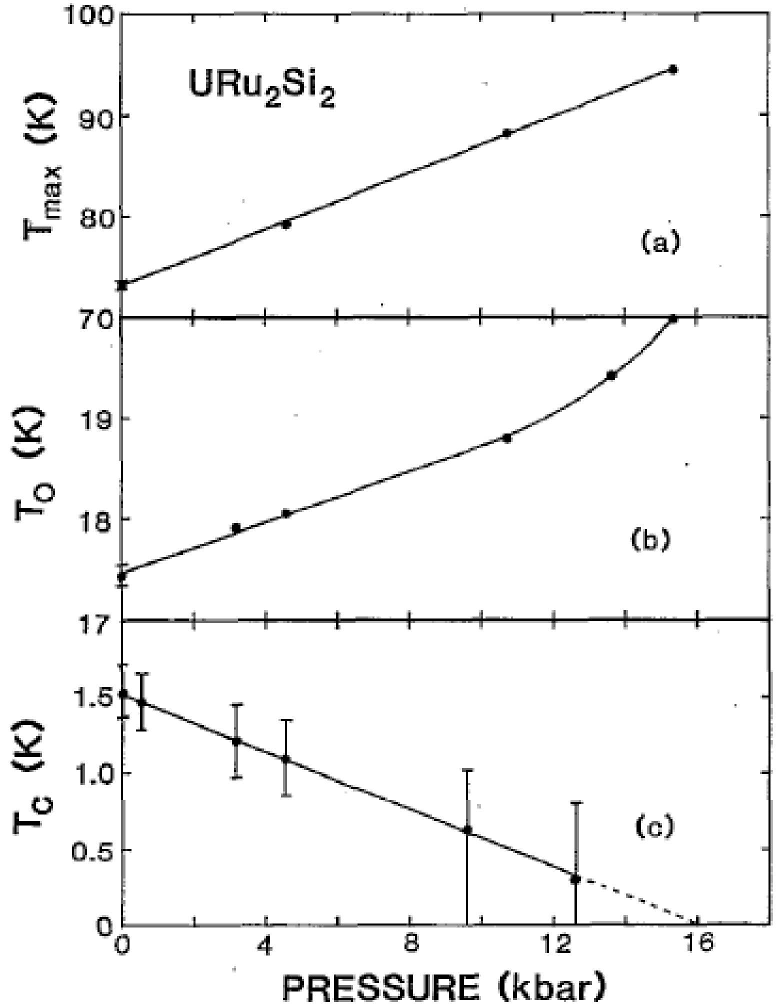
Maple, Dalichaouch, Kohara, Rossel, Torikachvili, McElfresh, Thompson, PRL (86)

- BCS-type mean field transition at T_o = 17.5 K ⇒ SDW or CDW
  - $\Delta \sim 10^2$ K, $\delta C \approx A\exp(-\Delta/T)$
  - ~ 40 % of FS removed by SDW or CDW ($\gamma(0)/\gamma' \approx 0.6$)
  - Small μ AFM ($\mu \approx 0.03\ \mu_B$/U, ||c-axis, (100) modulation)
  - Hidden order (HO) phase ($\delta S \approx 0.2\ln(2)$ too large for AFM with small μ)
  - SC below $T_c \approx 1.5$ K coexists with HO and AFM
- Inhomogeneous mixture of HO and large μ AFM phases; AFM fraction increases with P
- Identity of HO phase? Inhomogeneous HO/AFM mixture intrinsic? T vs P phase diagram?

Effect of pressure on competing electronic states in URu_2Si_2

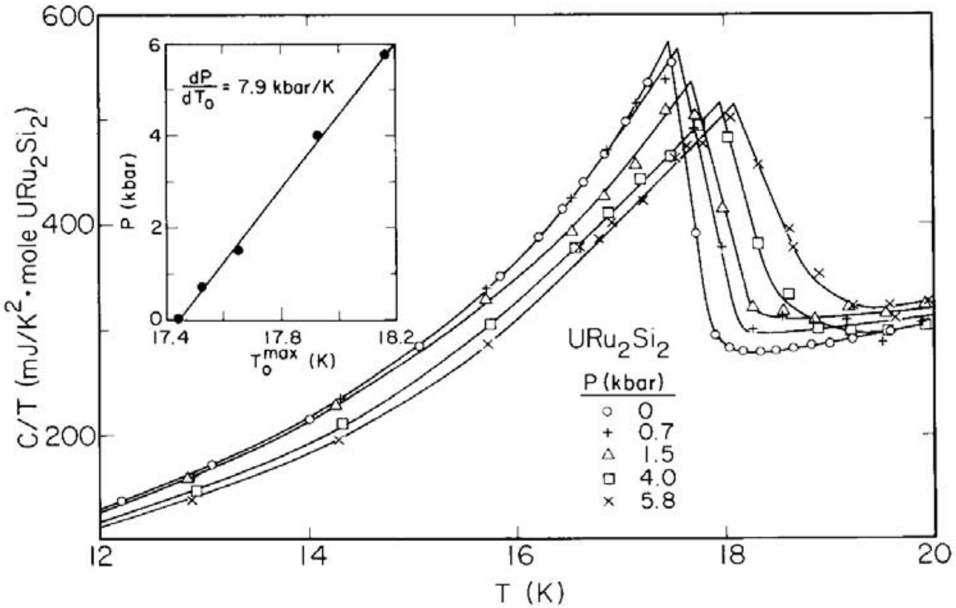




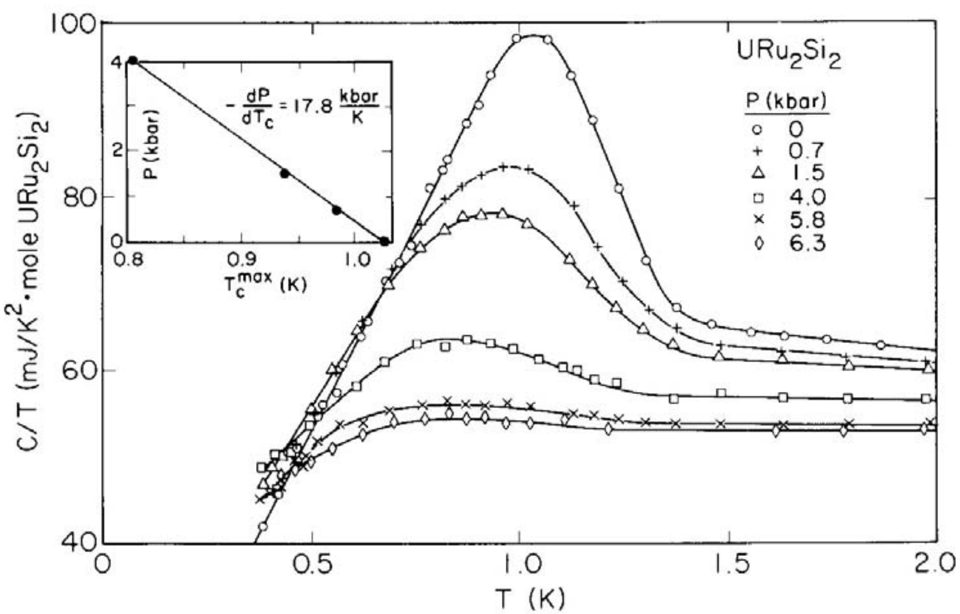


McElfresh, Thompson, Willis, Maple, Kohara, Torikachvili '87

Low temperature specific heat of URu_2Si_2 under pressure



R. A. Fisher, S. Kim. Y. Wu,
N. E. Phillips, M. W. McElfresh,
M. S. Torikachvili, M. B. Maple, 90



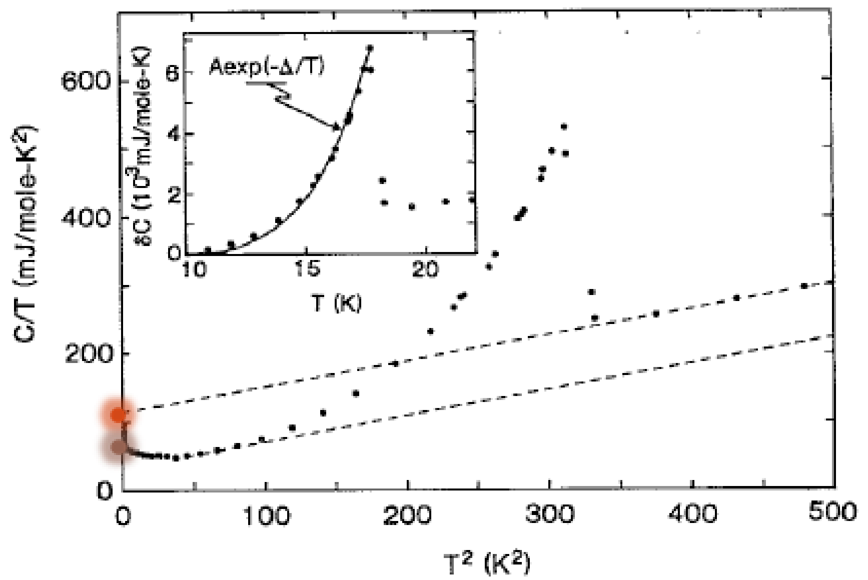
NOTE: Rapid suppression of specific heat jump ΔC at T_c with P

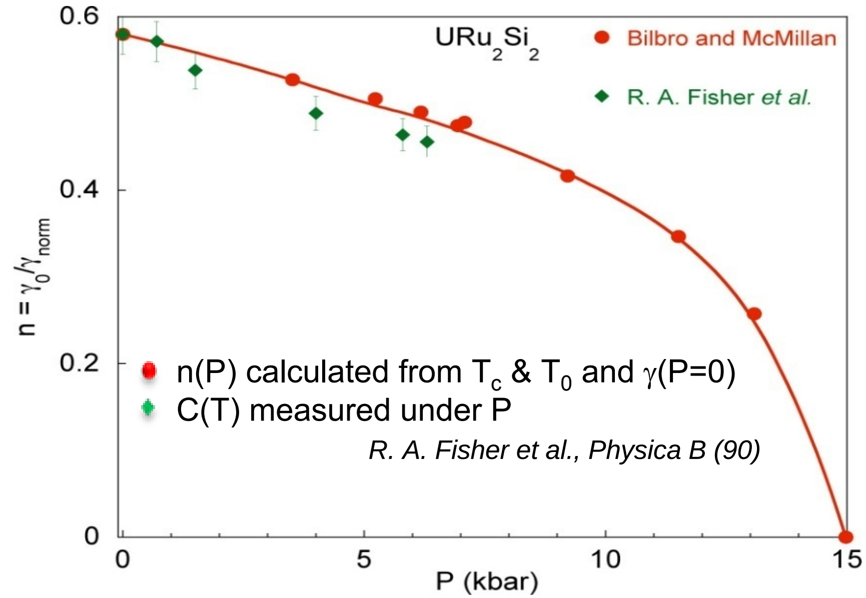
URu_2Si_2: Fermi surface competition

- *Bilbro & McMillan PRB (76)*
  - Theory – CDW/SDW competes with SCing order to gap a simple FS
  - HO/SMAFM and SCing phases compete for electrons
- $n = \gamma_0/\gamma_{norm}$
  - Amount of FS not gapped by CDW/SDW
  - C(T): n(0) = 0.58, T_{c0} = 3.9 K

$$T_{c0} = T_c(P)^{n(P)} T_0(P)^{1-n(P)}$$

M. B. Maple et al., PRL (86); J. R. Jeffries, N. P. Butch, B. T. Yukich, M. B. Maple, PRL (07)

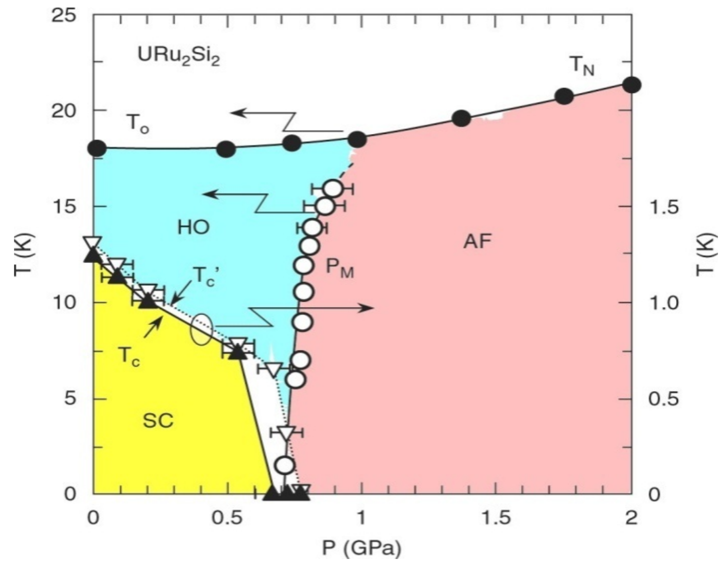


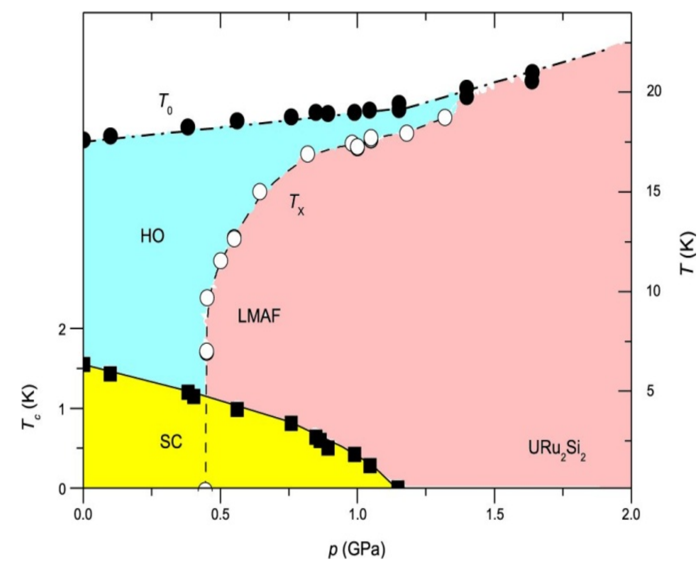


URu_2Si_2 under pressure

- T_0 increases
- T_c decreases
- μ_{AFM} increases
  - $0.03\mu_B \rightarrow 0.4\mu_B$
  - Neutron scattering, NMR, μSR
  - (In)homogeneous HO/AFM distribution?
  - HO → AFM 1st order?
  - HO/AFM coupled?

Amitsuka et al., JMMM (07)





Knebel et al., JMMM (07)