Non-Fermi liquid behavior near magnetic quantum critical points in U-based systems

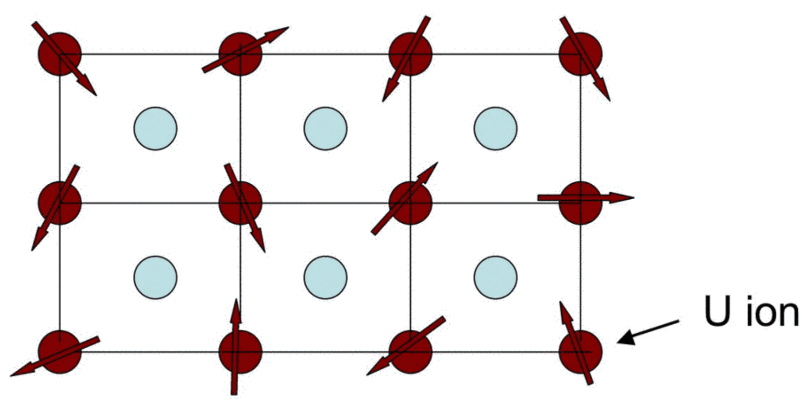
M. Brian Maple
U. California, San Diego

- Introduction to the physics of strongly correlated f-electron materials
- Recent experiments on $M_{1-x}U_xPd_3$ (M = Y,Sc) systems
  $Y_{1-x}U_xPd_3$: first f-electron system to exhibit NFL behavior
  Both systems: NFL behavior near spin glass QCP
  Interplay between Kondo effect, disorder, antiferromagnetic interactions
  $Sc_{1-x}U_xPd_3$: ω/T scaling of $\chi''(\omega,T)$
- Recent experiments on $URu_{2-x}Re_xSi_2$
  URu_2Si_2: “Hidden order?”
  $URu_{2-x}Re_xSi_2$: NFL behavior near ferromagnetic QCP

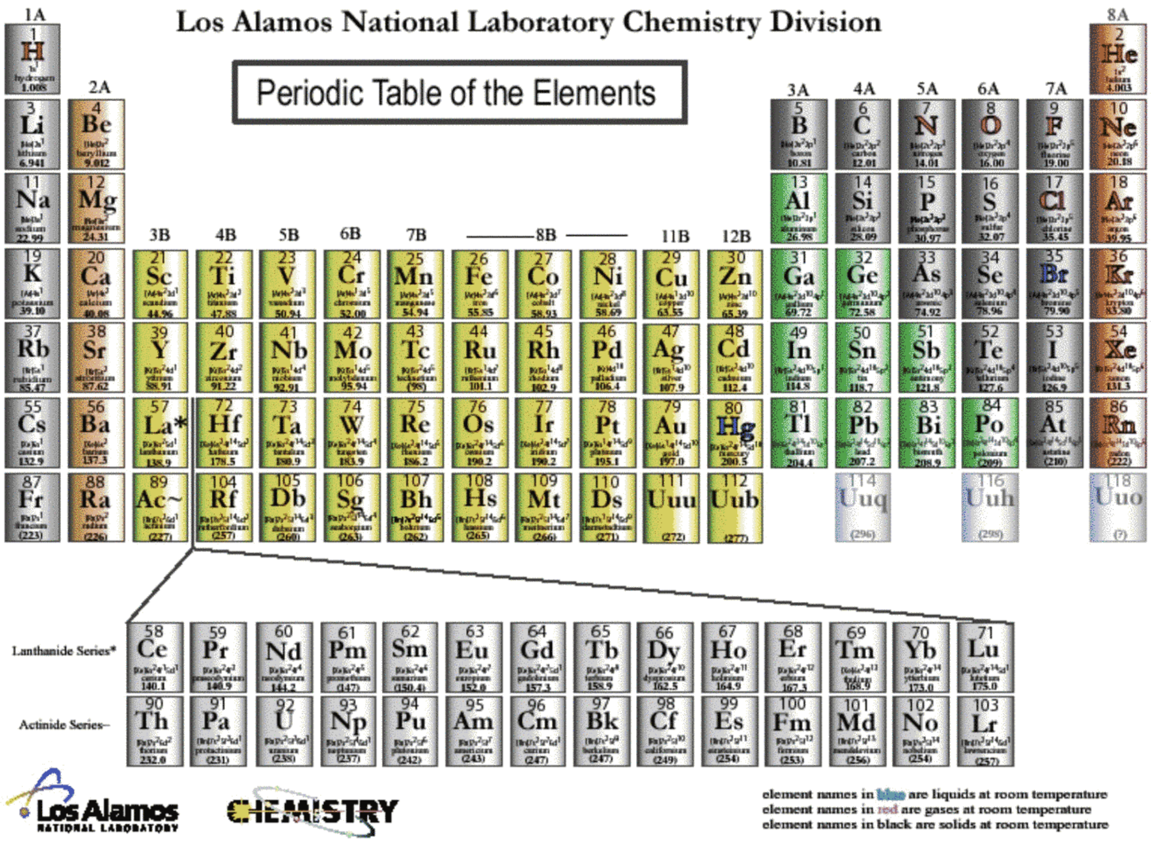
Supported by US DOE and NSF

U-based systems

- Intermetallic compounds containing sublattice of U ions



- U ion carries magnetic moment μ_{eff}
  Associated with partially-filled 5f electron shell
  Derived from Hund’s rules: S, L, J = |L - S| (<half-filled shell)
  Degeneracy = 2J+1
  $\mu_{eff} = g_J[J(J + 1)]^{1/2}\mu_B$
  $g_J = 1 + \{[J(J+1)+S(S+1)-L(L+1)]/2J(J+1)\}$



U-based systems

U: [Rn]$5f^3 6d^1 7s^2$ (atom)

- Trivalent (U^{3+}): $5f^3$

  S = 3/2, L = 6, J = 9/2, degeneracy = 10

  $\Rightarrow \mu_{eff} = 3.58\mu_B$
- Tetravalent (U^{4+}): $5f^2$

  S = 1, L = 5, J = 4, degeneracy = 9

  $\Rightarrow \mu_{eff} = 3.62\mu_B$
- CEF can lift degeneracy of Hund's rule multiplet

  e.g., U^{4+} in cubic CEF

  Γ_1 singlet, Γ_3 nonmagnetic doublet, Γ_4 & Γ_5 triplets

  Example of energy level scheme appropriate for quadrupolar Kondo effect →

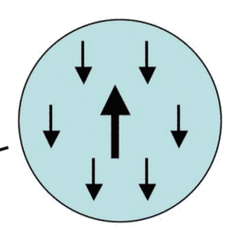
U-based systems

Hybridization of localized 5f and conduction electron states
⇒ interesting correlated electron physics!
Hybridization strength:
Weak ⇒ ionic behavior (5f occupation number <n> integral)
Moderate ⇒ Kondo behavior (<n> ~integral)
Appreciable ⇒ Valence fluctuations (<n> nonintegral)
Strong ⇒ f-bands

Heavy fermion f-electron materials

Underlying physics — significant hybridization between localized 5f- & conduction-electron states

- AFM exchange interaction
  $\mathcal{H}_{ex} = -2\mathcal{J}\mathbf{S}\cdot\mathbf{s}$ where $\mathcal{J} \sim -\langle V_{kf}^2\rangle/(E_F - F_f) < 0$
- Kondo effect (lattice of U ions)
  $T_K \sim T_F \exp(-1/N(E_F)|\mathcal{J}|) \sim T^*$ (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)

Heavy fermion f-electron materials

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 $T << T_c$
  - 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, $U_{1-x}Th_xBe_{13}$, $PrOs_4Sb_{12}$
- 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 mediated by spin fluctuations

Recent experiments on $M_{1-x}U_xPd_3$ (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 $0 < x \le 0.2$

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 ⇒ 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*)

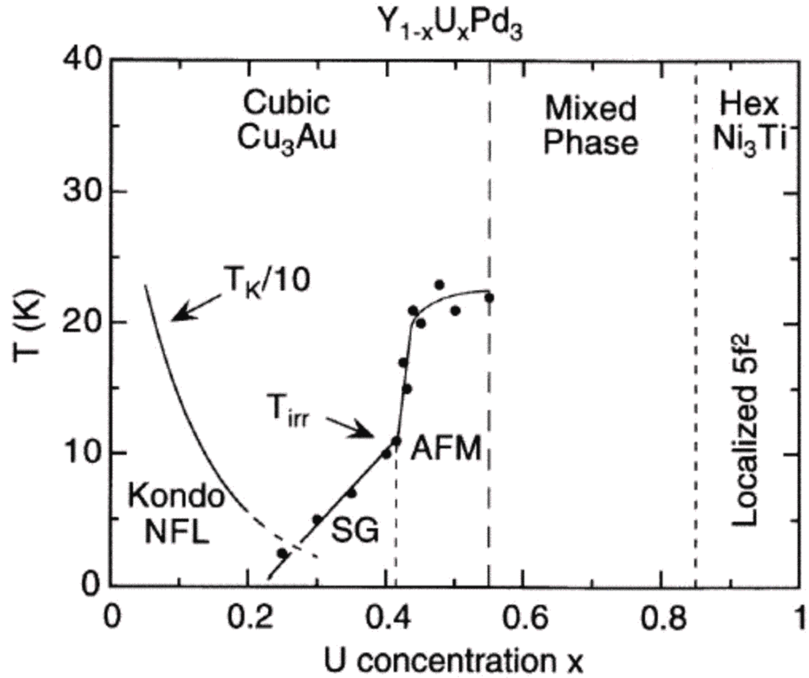
$Sc_{1-x}U_xPd_3$: NFL behavior similar to that observed in $Y_{1-x}U_xPd_3$ system

More homogeneous distribution of U

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

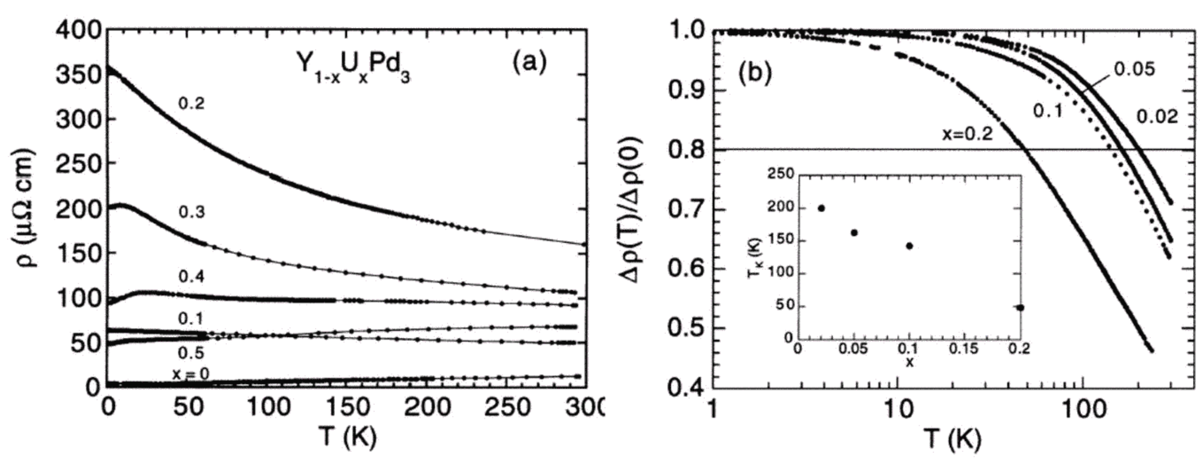
Interplay between Kondo effect, disorder, AFM interactions

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



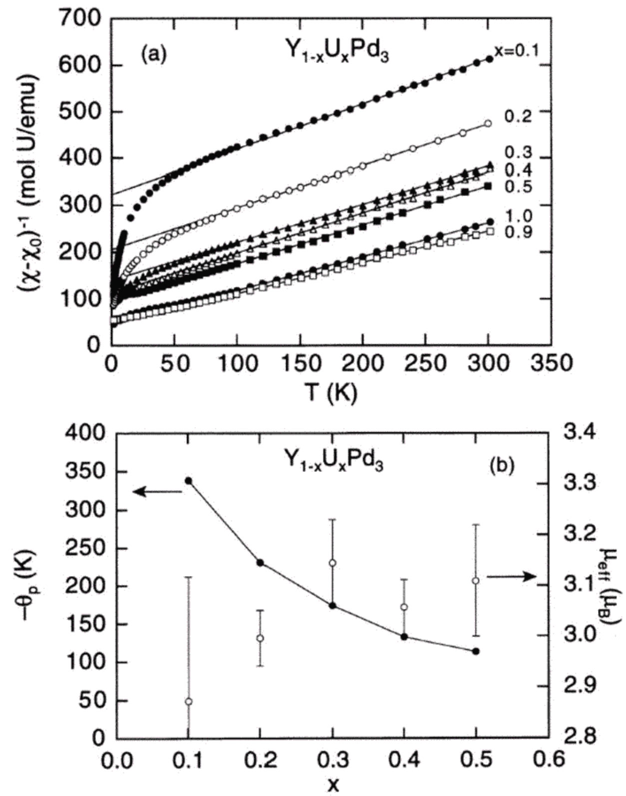
*C. L. Seaman et al., PRL **67**, 2882 '91*
*D. A. Gajewski, N. R. Dilley, R. Chau, M. B. Maple, J. Phys.: Condens. Matter **8**, 9793 '96*

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*

Magnetic susceptibility χ vs T for the $Y_{1-x}U_xPd_3$ system (high T)



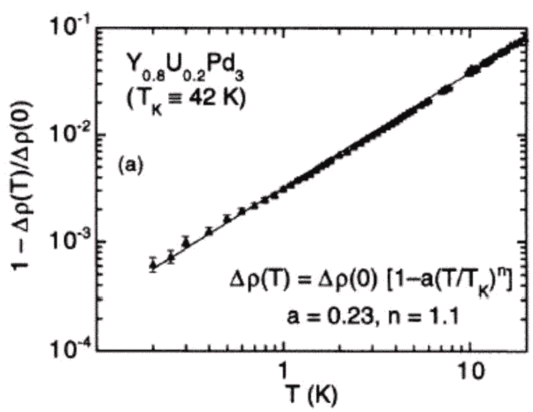
For Kondo systems:

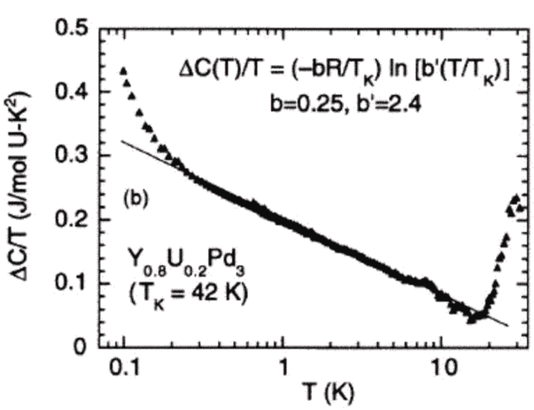
$\chi(T) = N\mu_{eff}^2/3k_B(T-\theta_p)$

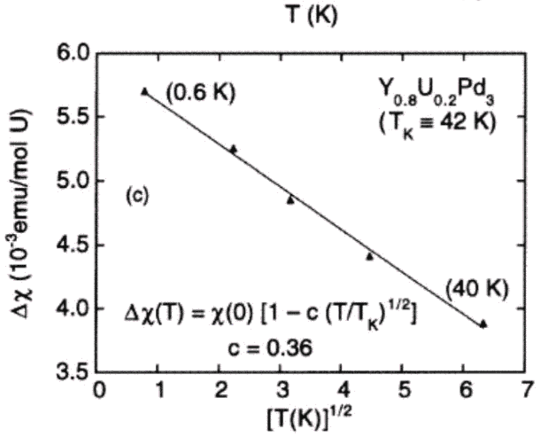
$\theta_p \approx -3T_K$

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

Low-T NFL behavior in $\rho(T)$, $C(T)$, $\chi(T)$ for $Y_{1-x}U_xPd_3$







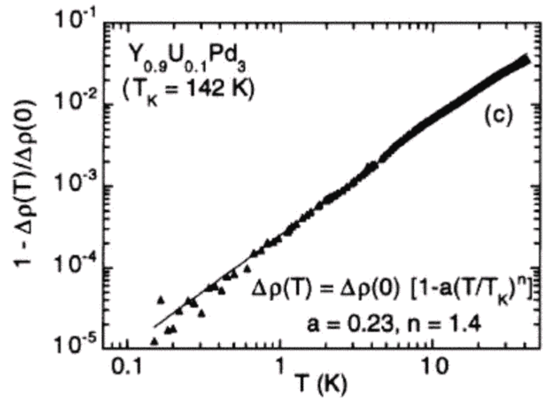
($T << T_K$):

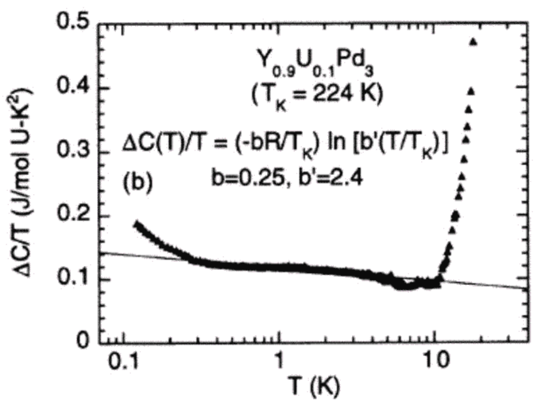
$\rho(T)$, $C(T)$, $\chi(T)$

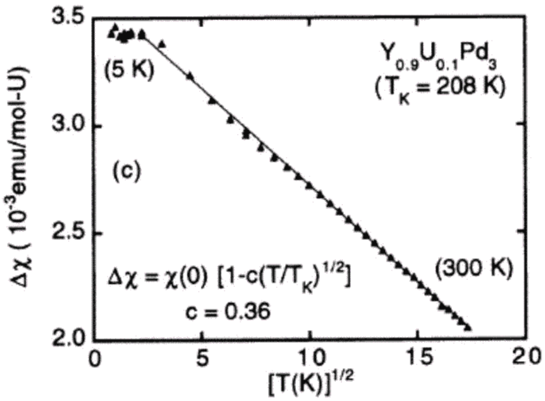
scale with T_K

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

Low-T NFL behavior in $\rho(T)$, $C(T)$, $\chi(T)$ for $Y_{1-x}U_xPd_3$



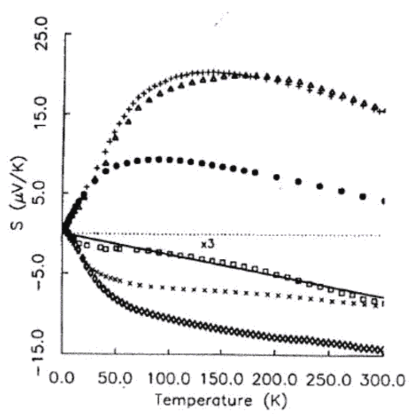


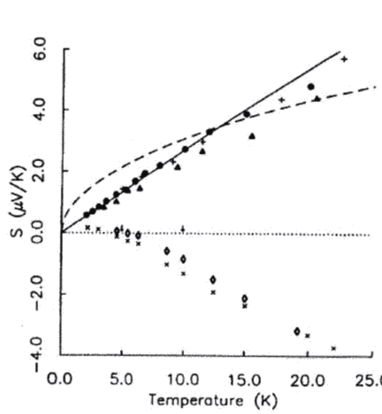


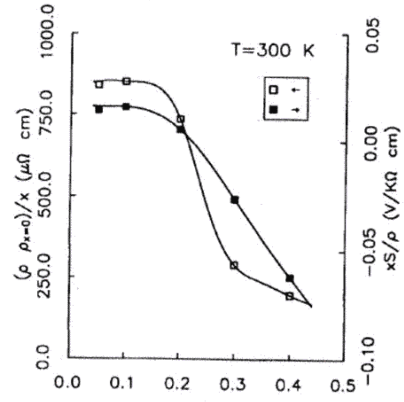
($T << T_K$):
$\rho(T)$, $C(T)$, $\chi(T)$
scale with T_K

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

Thermoelectric power S vs T for the $Y_{1-x}U_xPd_3$ system



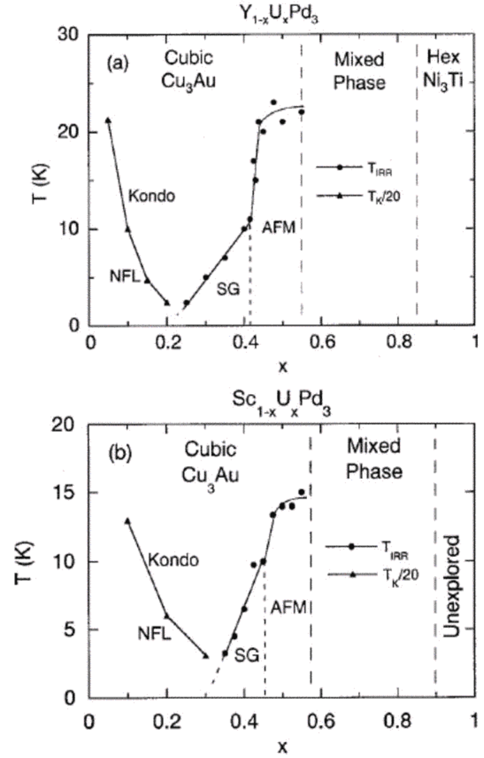




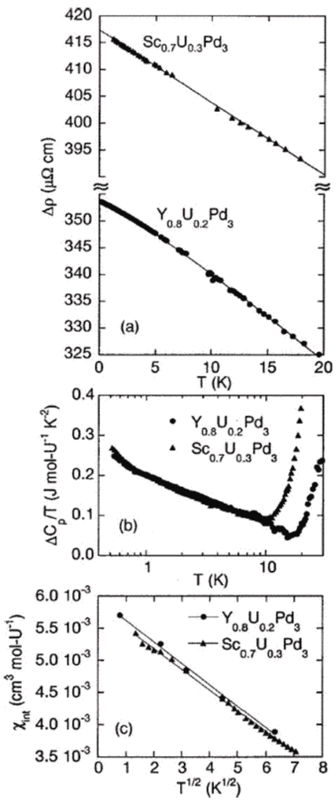
Symbols:
X=0 (star)
X=0.03 (square)
X=0.1 (plus)
X=0.2 (triangle)
X=0.3 (x)
X=0.4 (diamond)

J. McCarten, S. E. Brown, C. L. Seaman, M. B. Maple, PRB ***49****, 6400 '94*

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

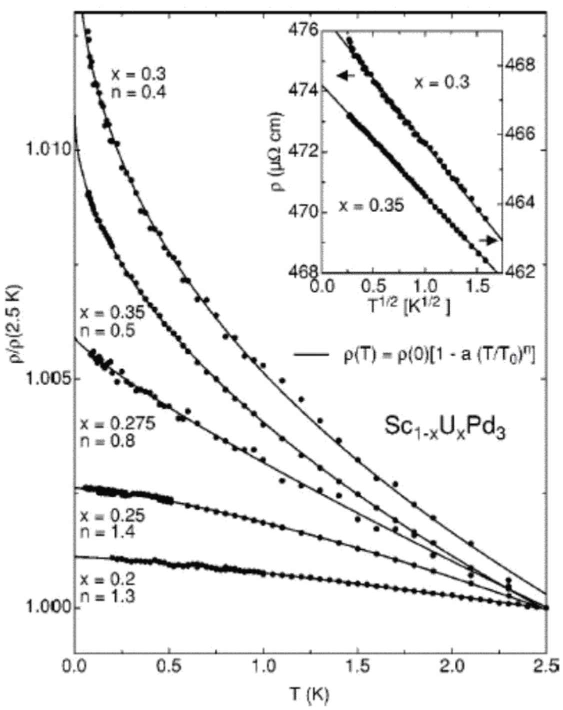


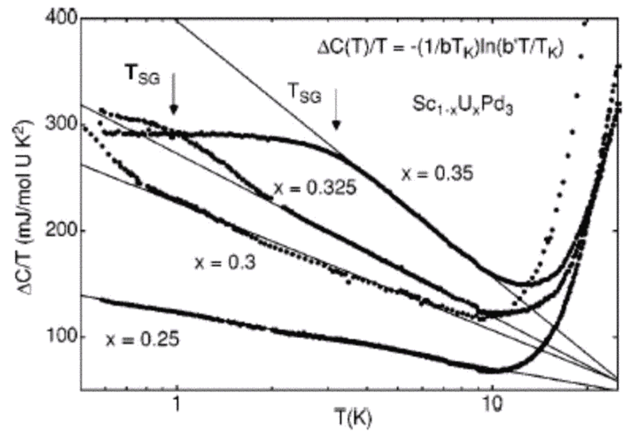
- Similar T-x phase diagrams & low T NFL characteristics in ρ(T), C(T)/T, χ(T)
- T_K decreases with x
- AFM & SG phases
- SG QCP
  M=Y: $x_c \approx 0.2$
  M=Sc: $x_c \approx 0.3$
- NFL behavior associated with SG QCP



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

ρ(T) and C(T)/T of the $Sc_{1-x}U_xPd_3$ system near SG QCP

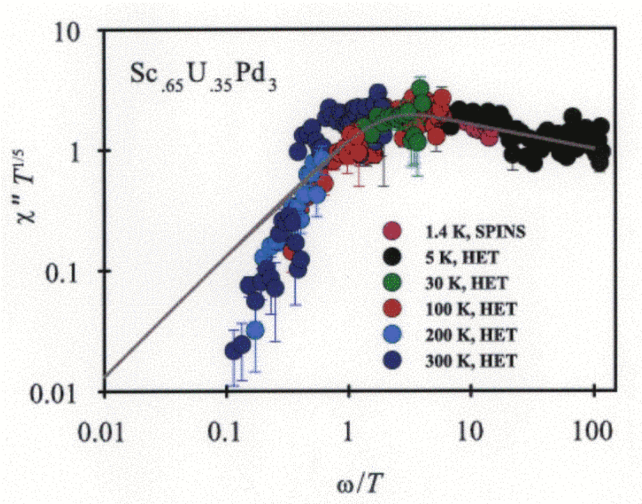




Near SG QCP ($x_c \approx 0.3$)
$\rho(T) \approx \rho(0)[1 - (T/T_o)^{1/2}]$
$C(T)/T \approx -\ln(T/T_o)$

*R. P. Dickey, V. S. Zapf, P.-C. Ho, E. J. Freeman, N. A. Frederick, M. B. Maple, PRB **68**, 104404 ‘03*

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 ⇒ 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); α = 1/3

Near AFM QCP

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

$CeCu_{5.9}Au_{0.1}$; α = 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

The $URu_{2-x}Re_xSi_2$ system

NFL behavior in the vicinity of FM QCP

- Substitution of other elements for Ru (i.e., $URu_{2-x}M_xSi_2$) depresses T_N amd T_c *Dalichaouch et al. '90*
- FM instability observed in $URu_{2-x}M_xSi_2$ for M=Tc, Re at higher x *Dalichaouch, Torikachvili, Maple, Giorgi '89*
  - Similar behavior for Tc, Re — FM extends over range of x values, maxima in θ_c, μ_s, γ at nearly same x
  - No anomalies in C(T) & ρ(T) due to FM near θ_c
- FM determined from M(H,T) measurements & analyzed by means of modified Arrott plots
  $M^{1/\beta}$ vs $(H/M)^{1/\gamma}$
  - Standard Arrott plots: $\beta = 1/2$, $\gamma = 1$ (MFT)
  - Critical exponents β, γ, δ defined by
    $M(t,H=0) \propto t^{\beta}$; $M(t=0,H) \propto H^{1/\gamma}$; $\gamma=\beta(\delta-1)$
  - Analysis based on Arrott-Noakes equation
    $(H/M)^{1/\gamma} = (T-\theta_c)/T_1+(M/M_1)^{1/\beta}$
    where T_1 & M_1 are normalization factors

Parent compound URu_2Si_2

Moderately heavy Fermi liquid: $m^* \approx 25\ m_e$

Superconductivity: $T_c = 1.5$ K, $\Delta_s \approx 0.1$ meV

Hidden order (HO) phase: $T_o = 17.5$ K, $\Delta \approx 11$ meV

AFM (U 5f): $\mu \approx 0.02\text{-}0.04\ \mu_B$/U, (||c-axis), (100) modulation

Transport, thermal, magnetic properties:

Single crystal specimens

Palstra, Menovsky, van den Berg, Dirkmaat, Kes, Nieuwenhuys, Mydosh '85

Polycrystalline specimens

Schlabitz, Baumann, Pollit, Rauchschwalbe, Mayer, Alheim, Bredl ZP '86

Maple, Dalichaouch, Kohara, Rossel, Torikachvili, McElfresh, Thompson PRL '86

Neutron scattering

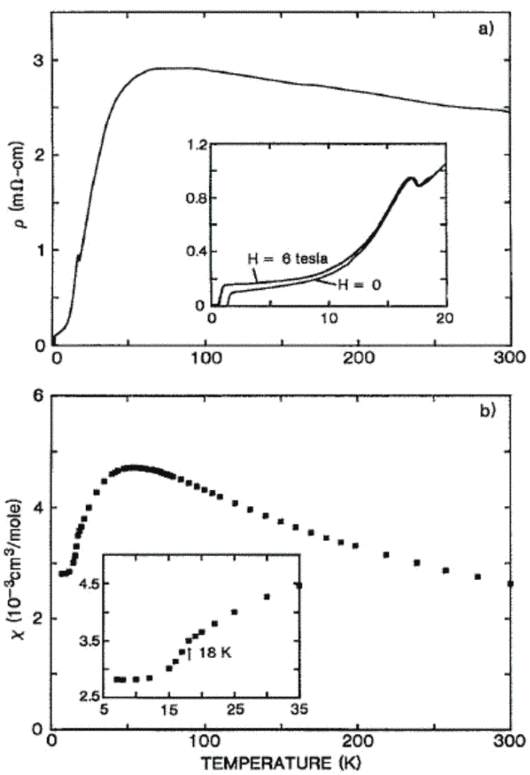
Single crystal specimens

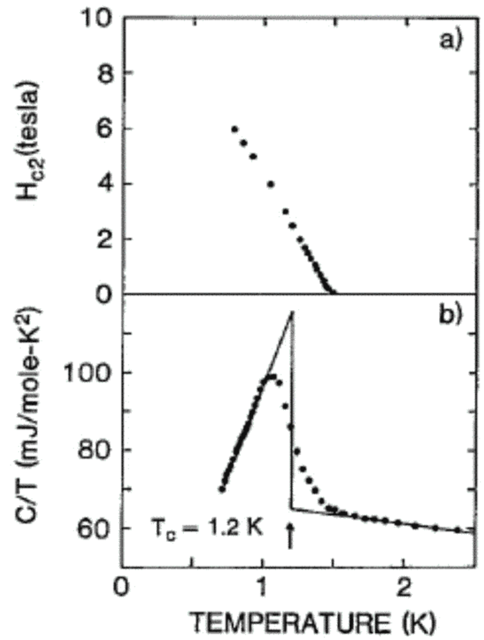
Broholm, Kjems, Buyers, Matthews, Palstra, Menovsky, Mydosh '86

Polycrystals (INS)

Walter, Loong, Loewenhaupt, Schlabitz '86

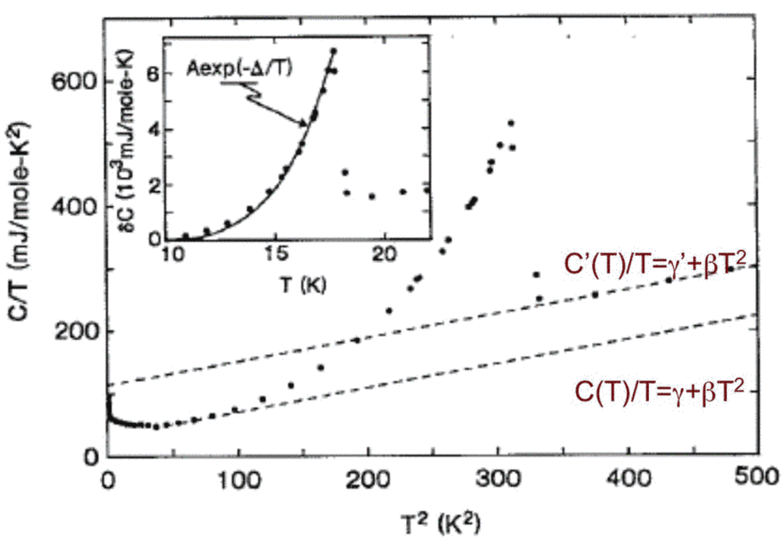
Normal and superconducting state properties of URu_2Si_2





Maple, Dalichaouch, Kohara, Rossel, Torikachvili, McElfresh, Thompson, PRL '86

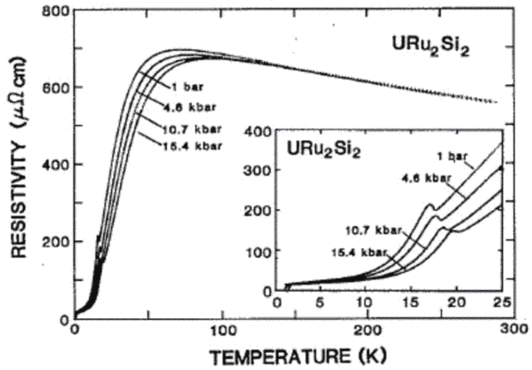
Specific heat of URu_2Si_2

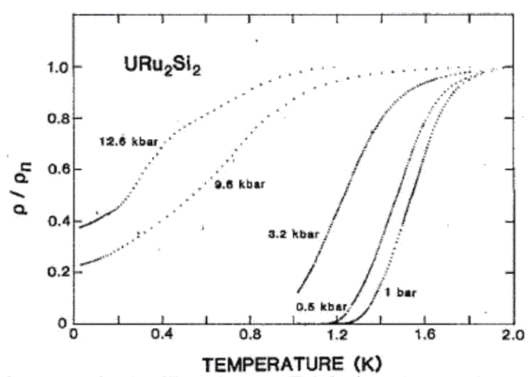


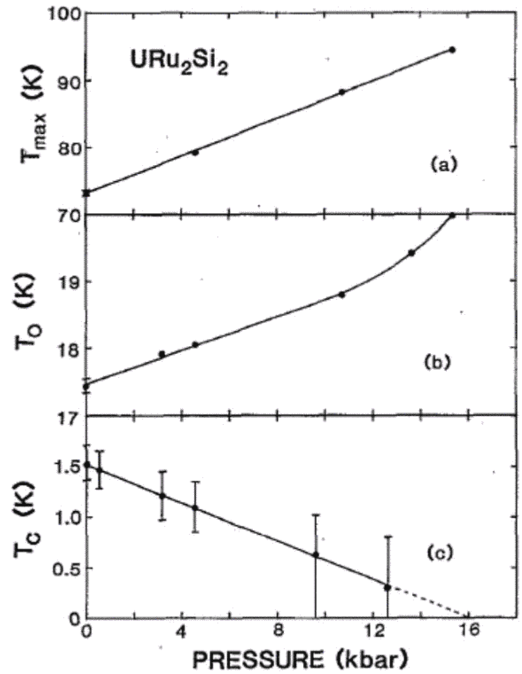
BCS-type mean field transition at T_o = 17.5 K
$\delta C \approx A\exp(-\Delta/T)$; $\Delta \sim 10^2$ K ~ 10 meV
AFM ($\mu \approx 0.02\ \mu_B$/U) coexists with SC
$\gamma(0)/\gamma' \approx 0.6 \Rightarrow$ ~ 40 % Fermi surface removed by SDW or CDW
SC & SDW or CDW compete for Fermi surface!

Maple, Dalichaouch, Kohara, Rossel, Torikachvili, McElfresh, Thompson, PRL '86

Effect of pressure on competing electronic states in URu_2Si_2

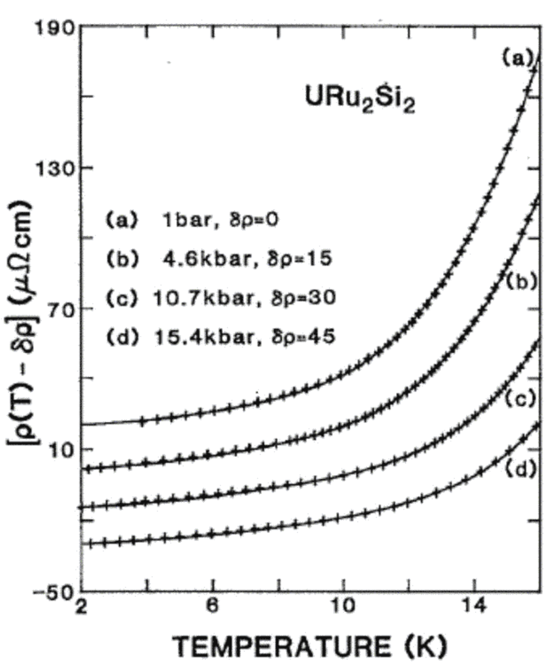


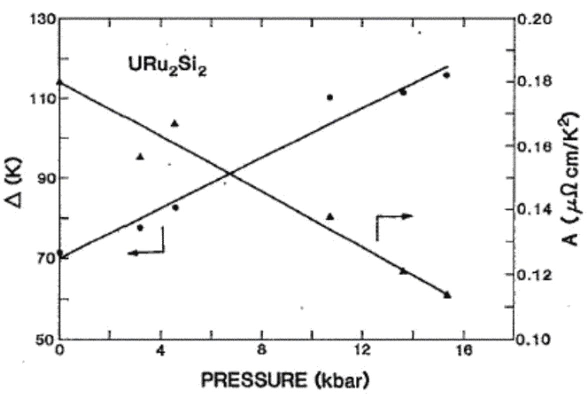




McElfresh, Thompson, Willis, Maple, Kohara, Torikachvili '87

Electrical resistivity $\rho(T)$ of URu_2Si_2 under pressure





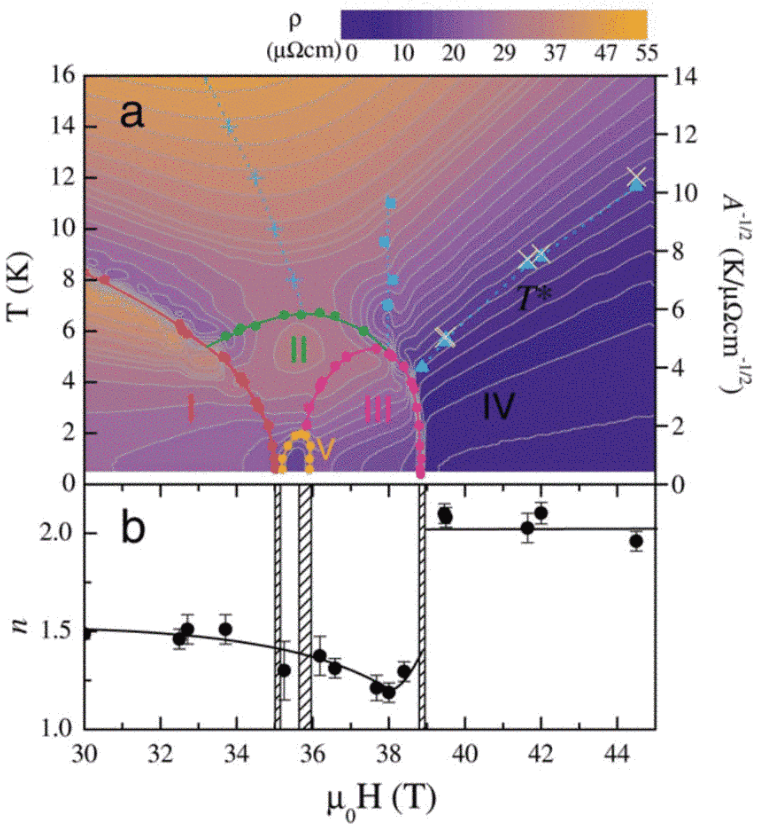
$\rho(T)=\rho_o+AT^2-B(T/\Delta)(1+2T/\Delta)\exp(-\Delta/T)$
Hessel Anderson '80

Palstra, Menovsky, Mydosh '86
$\rho(T)$: URu_2Si_2 (P=0)

$A \propto T_K^{-2} \propto T_{max}^{-2}$

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

Field-induced phases in URu_2Si_2

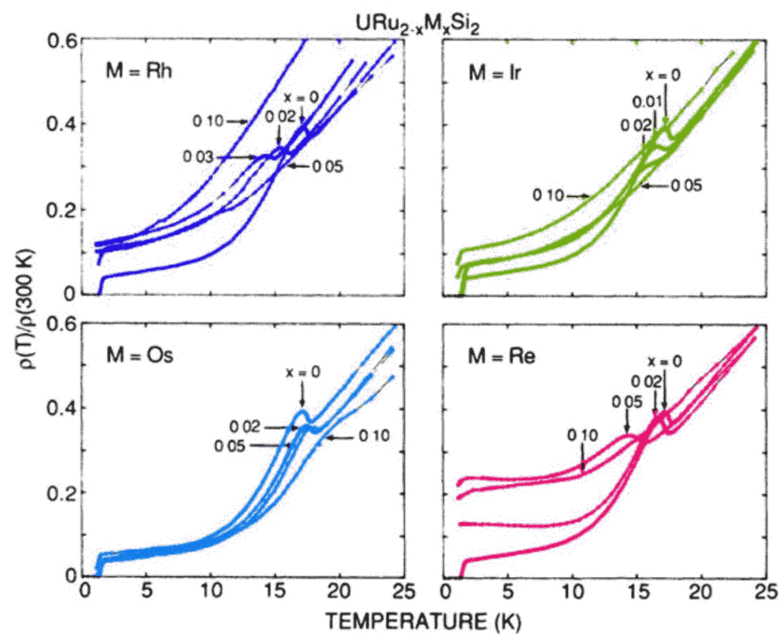


Electrical resistivity
$\rho = \rho_o + AT^n$
n = 2: FL
n ≈ 1 - 1.5: NFL

*K. H. Kim et al., PRL **91** ‘03*

Transition metal substitutions for Ru in URu_2Si_2

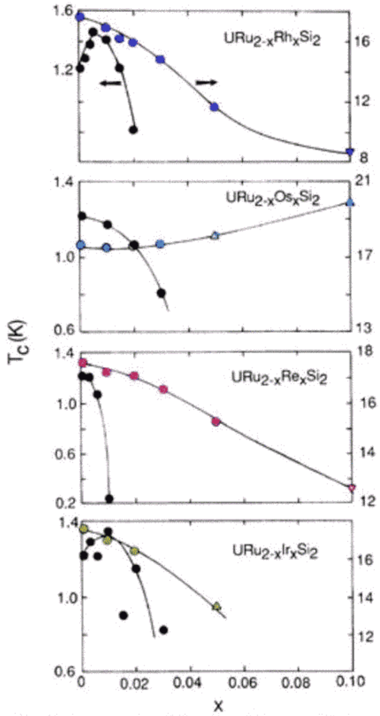
$URu_{2-x}M_xSi_2$ (M = Rh, Os, Re, Ir)



Dalichaouch, Maple, Chen, Kohara, Rossel, Torikachvili, Giorgi ‘90

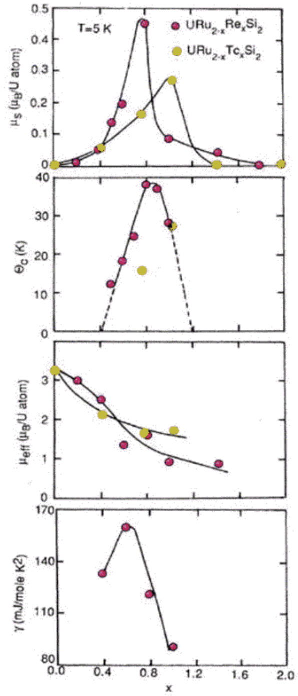
Transition metal substitutions for Ru in URu_2Si_2

$URu_{2-x}M_xSi_2$ (M = Rh, Os, Re, Ir)



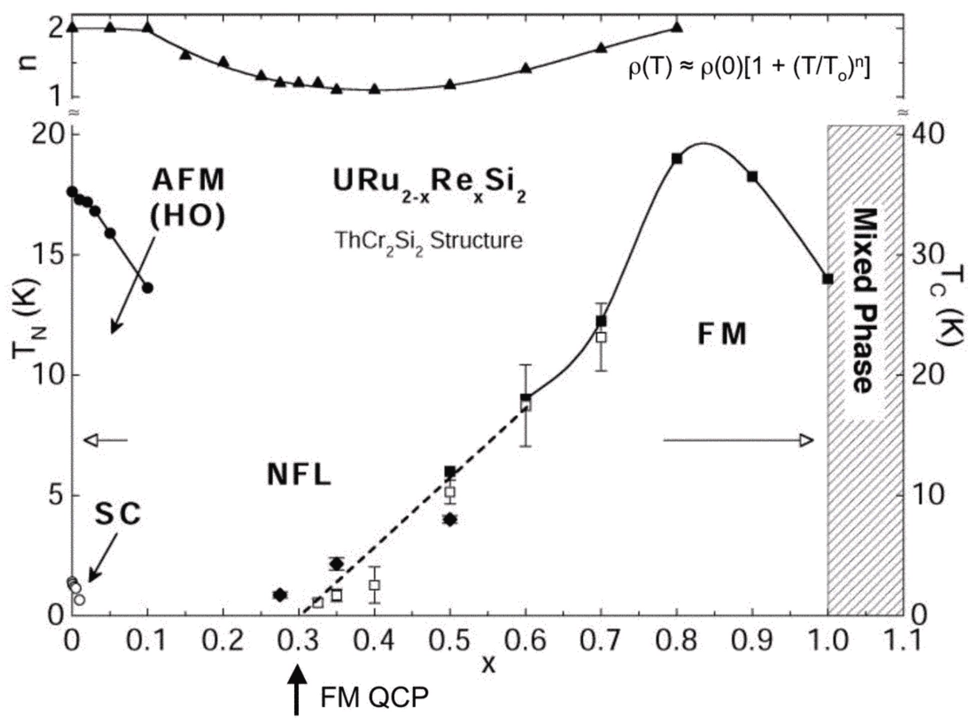
Dalichaouch, Maple, Chen, Kohara, Rossel, Torikachvili, Giorgi '90

$URu_{2-x}M_xSi_2$ (M=Re,Tc) ⇒ FM!



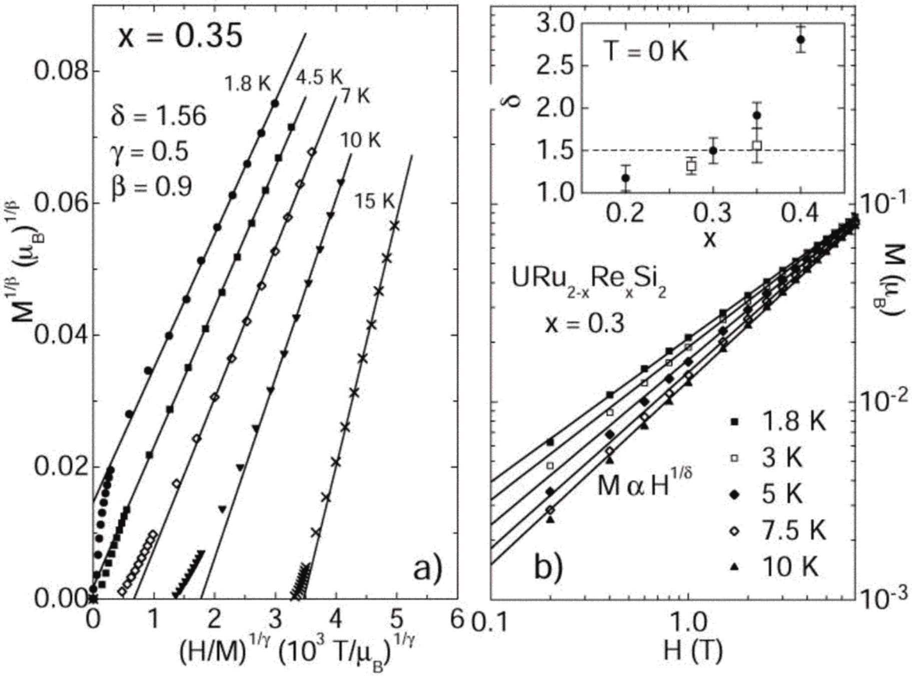
Dalichaouch, Torikachvili, Maple, Giorgi '89

T-x phase diagram of the $URu_{2-x}Re_xSi_2$ system



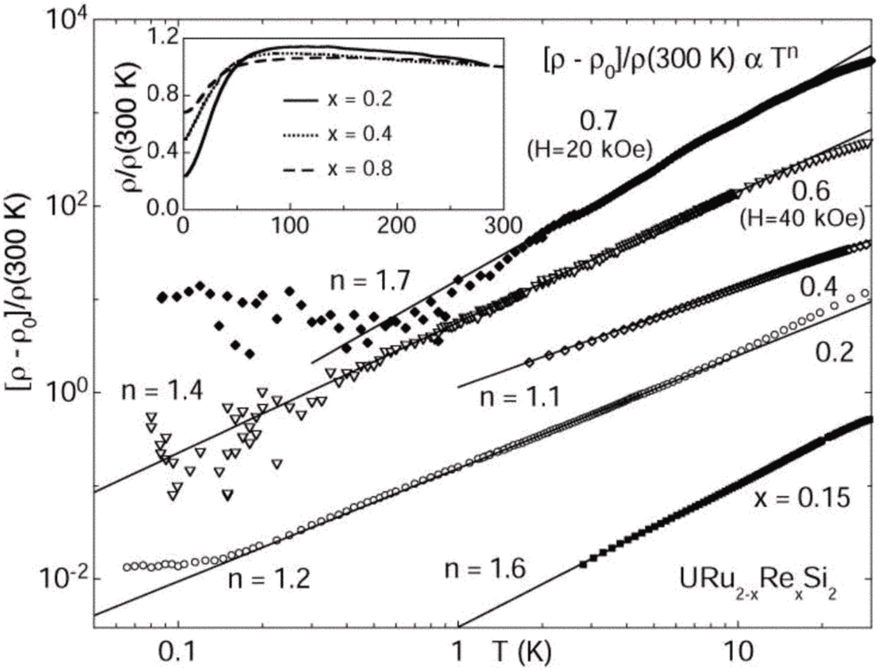
E. D. Bauer, V. S. Zapf, P.-C. Ho, E. J. Freeman, C. Sirvent, M. B. Maple, PRL '05 (in press)

$URu_{2-x}Re_xSi_2$: scaled Arrott plots



E. D. Bauer, V. S. Zapf, P.-C. Ho, E. J. Freeman, C. Sirvent, M. B. Maple, PRL '05 (in press)

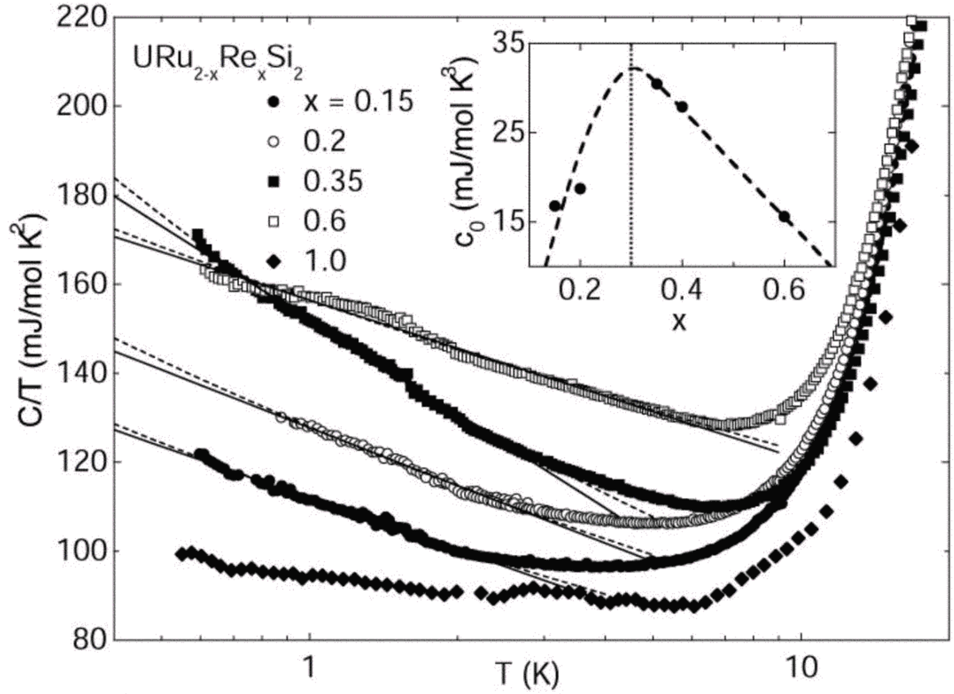
$URu_{2-x}Re_xSi_2$: ρ(T) at low T



NOTE: Each curve shifted by 1 decade for clarity

E. D. Bauer, V. S. Zapf, P.-C. Ho, E. J. Freeman, C. Sirvent, M. B. Maple, PRL '05 (in press)

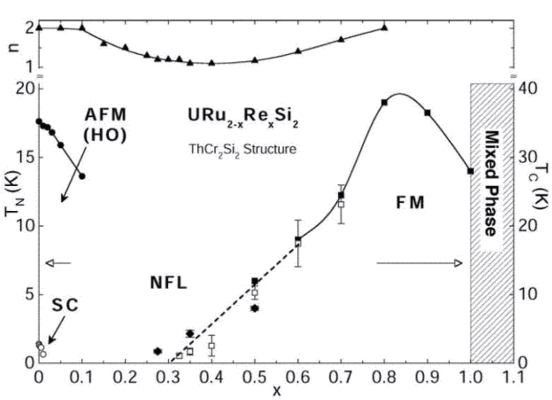
$URu_{2-x}Re_xSi_2$: C(T) at low T

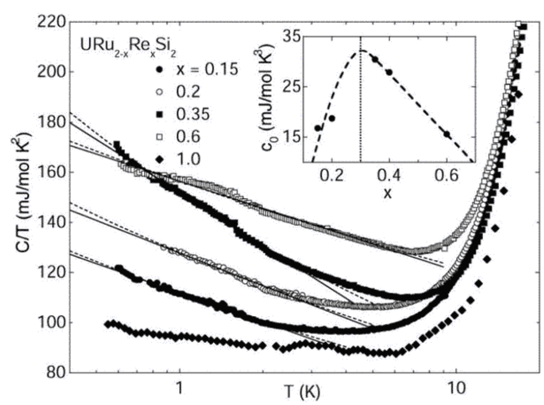


$C(T)/T = \gamma_o - c_o \ln T$
(or T^{-n} where $n \approx 0.1 - 0.2$)

E. D. Bauer, V. S. Zapf, P.-C. Ho, E. J. Freeman, C. Sirvent, M. B. Maple, PRL '05 (in press)

T-x phase diagram and C(T) of the $URu_{2-x}Re_xSi_2$ system





NFL behavior:
~0.15 < x < ~0.6 (extends into FM phase ~0.3 < x < ~1.0)
$C(T)/T = \gamma_o - c_o \ln T$ (or T^{-n} where $n \approx 0.1 - 0.2$)
$\rho(T) \propto T^n$ ($n \approx 1 - 1.5$)
Deviation from FL behavior greatest at $x_c \approx 0.3$ (FM QCP) where c_o maximum & n minimum ($n \approx 1$)
$\delta(0)$ varies with x; $\delta(0) = 1.5$ at $x_c \approx 0.3$
$\chi(T) \propto T^{-n}$ in PM region (~0.15 < x < ~0.3)
($n \approx 0.2$ for x = 0.1, $n \approx 0.4$ for x = 0.3)

Possible scenarios for NFL behavior in $URu_{2-x}Re_xSi_2$

- Griffiths-McCoy phase — *Castro Neto, Castilla, Jones '98*
  Competition between Kondo effect & RKKY interaction + disorder ⇒ magnetic clusters embedded in a nonmagnetic FL ⇒ NFL characteristics

  $C(T)/T \propto \chi(T) \propto T^{-n}$ $(n \approx 0.2 - 0.3)$

  Could account for NFL behavior in PM & FM phases in vicinity of FM QCP
- Spin fluctuation models — *Millis '93; Moriya, Takamoto '95*

  2D: $\rho(T) \propto T^{5/3}$
  $C(T)/T \propto -\ln T$
  $\chi(T) \propto T^{-4/3}$
  $\theta_c \propto |\delta - \delta_c|^{3/4}$

  3D: $\rho(T) \propto T^{4/3}$
  $C(T)/T \propto T^{-1/3}$
  $\chi(T) \propto T^{-1}$ or $1/T\ln T$
  $\theta_c \propto |\delta - \delta_c|$

- Quantum critical behavior of itinerant FMs incorporating effects of nonmagnetic disorder — *Belitz, Kirkpatrick '01*

  Near x_c: $C(T)/T \propto -\ln T$
  $\beta = 2, \gamma = 1, \delta = 1.5$