Magnetically ordered sublattices in conventional superconductors
compound

$[\text{InRh}_x\text{B}_y, \text{InMg}_3\text{S}_3, \text{InMg}_5\text{Se}_8]$

Two weakly interacting systems suggest

- Mobile SCing subsystem - $\text{RhB}_x\text{B}_y, \text{Mg}_5\text{S}_3, \text{Mg}_5\text{Se}_8$ "checkers"

- Localized magnetic subsystem - In sublattice

Weak exchange interaction between local spins and

$\Rightarrow SCing, T \ll T_c$

What about $g < 0$ case in In sublattice

This gives Kondo effect $\Rightarrow$ heavy fermion behavior

$\Rightarrow$ unconventional SC that can coexist with magnetic order
Crystal structure

$\text{RRh}_4\text{B}_4$

[Diagram of a crystal structure with labeled atoms $\bullet R$, $\circ \text{Rh}$, and $\bullet \text{B}$]
Antiferromagnetic superconductors

- Compounds
  
  $\text{RM}_{6}\text{S}_{8}$  \hspace{1cm} $R = \text{Gd, Tb, Dy, Er}$  \hspace{1cm} (U. Geneva)
  
  $\text{RM}_{6}\text{Se}_{8}$  \hspace{1cm} $R = \text{Gd, Tb, Er}$  \hspace{1cm} (UCSD)
  
  $\text{RRh}_{4}\text{B}_{4}$  \hspace{1cm} $R = \text{Nd, Sm, Tm}$  \hspace{1cm} (UCSD)

- \text{SC} & \text{AFM coexist in} \ H = 0

  $<H_{ex}> = 0$ over scale of $\xi$

  $\uparrow \downarrow \uparrow \downarrow \uparrow \downarrow \uparrow \xi \gg a$

  \hspace{1cm} $\leftarrow a \rightarrow$

- \text{AFM modifies SC'ing properties}

  e.g., \(H_{c2}\) vs \(T\)

  \(H_{c2}\) increases or decreases below \(T_N\) — nonuniversal

Mechanisms

\text{Enhancement}

(1) Decrease in \(<M>\) below \(T_N\) \(\Rightarrow\) reduced pair breaking

\text{Suppression}

(1) \(\mu\) fluctuations near \(T_N\) \(\Rightarrow\) increased pair breaking

(2) AFM magnons \(\Rightarrow\) repulsive electron-electron interaction

(3) Gap in \(E(k)\) due to change in lattice periodicity \(\Rightarrow\) reduction of available phase space for virtual pair scattering
$H_{c2}(T)$ of $RRh_4B_4$ magnetic superconductors

AFM-SC's: $NdRh_4B_4$, $SmRh_4B_4$, $TmRh_4B_4$

FM-SC: $ErRh_4B_4$

*Upper critical field vs temperature plots for different compounds.*

Conventional SC: $H_{c2}$
Ferromagnetic superconductors

- Compounds
  - HoMo$_6$S$_8$ (U. Geneva)
  - ErRh$_4$B$_4$ (UCSD)
  - ErRh$_{1.1}$Sn$_{3.6}$ (AT&T, UCSD, BNL)

- Reentrant SC'ing behavior due to onset of FM order

  First-order SC'ing–normal FM transition at $T_{c2} \sim T_M < T_{c1}$

  Narrow temperature interval above $T_{c2}$:
   - Microscopic coexistence of SC and sinusoidally modulated magnetic state with $\lambda \sim 10^2$ Å (HoMo$_6$S$_8$, ErRh$_4$B$_4$)
   - Macroscopic coexistence of SC'ing regions (containing sinusoidal magnetic state) and normal FM domains (ErRh$_4$B$_4$)

  Reminiscent of "cryptoferromagnetic" state — based on exchange interaction (Anderson & Suhl '59)

  $$\lambda \sim 2(\pi^2\xi/3k_F^2)^{1/3} \lesssim 10\,\text{Å} \quad \text{(ErRh}_4\text{B}_4)$$

  Various theories based on electromagnetic interaction (~'80)

  $$\lambda \sim (4\pi^3D/C)^{1/4}\lambda_L^{1/2} \sim 10^2\,\text{Å}$$

  Spontaneous vortex lattice
Reentrant SC due to FM order: $\text{ErRh}_4\text{B}_4$ ($\text{HoM}_{2}\text{S}_3$)

$\chi_{dc}$ (arb. units)

Resistance ($\mu$O$	ext{m}$)

$T_{c2} \theta_c$

$T_{c1}$

$\text{SC}$

$\text{FM}$

SC'ing & FM regions

SC is sinusoidally-modulated magnetic state ($\lambda \sim 100\text{Å}$)

*Fertig, Johnston, DeLong, McCallum, Maple, Matthias '77

*Moncton, McWhan, Schmidt, Shirane, Thomlinson, Maple, Mackay, Woolf, Fisk, Johnston '80 (neutron scattering)
Macroscopic coexistence of SC & normal FM domains

ErRh$_4$B$_4$
SAMPLE II
$T_{C1} = 8.7\,\text{K}$
$T_{C2} = 0.93\,\text{K}$

Microscopic coexistence of SD and Sinusoidally-modulated magnetic state with $\lambda \sim 10^2 A$

$ErRh_4B_4, T_c \approx 0.65 K$

10^2 NEUTRON COUNTS / 2.5 min

SCATTERING ANGLE $2\theta$ (degrees)


Similar behavior - HoNi$_2$S$_2$, Lynn et al. '81
LINEARLY POLARIZED SINUSOIDALLY MODULATED MAGNETIC STATE ($\mu \perp \mathbf{c}$) – ErRh$_4$B$_4$

Anisotropy

ErRh$_4$B$_4$ single crystal
Sinha, Crabtree, Hicks & Mook '81

Theories
Anderson, Suhl '59
Suhl '78
Blount, Varma '79
Bulaevski, Rusinov, Kulik '79
Matsumoto, Umezawa, Tachiki '79
Anderson-Suhl (59) cryptoferromagnetism

\[ F_n(H) - F_s(H) = \frac{1}{2} N c E \Delta^2 - \frac{1}{2} [\mu_n(q) - \mu_s(q)] H^2 \]

For wave number \( q \gg F_n(H) - F_s(H) > 0 \)

\[ \implies \frac{N c E \Delta^2}{H^2} > \frac{\mu_n(q) - \mu_s(q)}{2} \]

Maximize \( \mu(q) \) \( \implies q = (2\pi k F / E_0)^{1/2} \implies \lambda \approx 50 \AA \)

Note! W/o ordering at finite \( q = Q \)

FM: \( S_F \sim C N(k_B T) \) \( N \)-no. atoms

SC: \( S_F \sim (k_B T / E_F) N(k_B T) \)

\( \implies c \gg k_B T / E_F \implies FM \) ground state
\[(\text{Er}_{1-x}\text{Ho}_x)\text{Rh}_4\text{B}_4\] \[\text{FM-}\mu\text{Lc vs }\mu\text{Lc}\]

**Graph:**
- **X-axis:** \(x\) (range 0.0 to 1.0)
- **Y-axis:** \(T(\text{K})\)

**Regions:**
- **Superconducting**
- **Paramagnetic**
- **Ferromagnetic (\(\mu\text{Lc}\))**
- **SC Sinusoidally Modulated**
- **Normal FM Regions**
- **Mixed**

**Notes:**
- 2nd order
- 1st order

**References:**
- D.C. Johnston, W.A. Fertig, M.B. Maple & B.T. Matthias '78
$\text{Ho(Rh}_{1-x}\text{Ir}_x)\text{B}_4$  \textit{FM vs AFM}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{chart.png}
\caption{Phase diagram of $\text{Ho(Rh}_{1-x}\text{Ir}_x)\text{B}_4$}
\end{figure}

\textbf{References:}

H.C. Ku, F.Acker & B.T. Matthias '80

K.N. Yang, S.E. Lambert, H.C. Hamaker, M.B. Maple, H.A. Mook & H.C. Ku '82

S.E. Lambert, M.B. Maple, O.A. Pringle & H.A. Mook '85

\textit{Re'To for Ru \textit{\leftrightarrow} Local moment FM
Heavy fermion compounds

- Metallic lanthanide and actinide compounds
  SC’ing, magnetic, nonmagnetic
  Nonmagnetic compounds: heavy Fermi liquids
  \[ \gamma(T) = C(T)/T \sim \chi(T) \sim \text{const.}, \quad \rho(T) \sim T^2 \quad (T \ll T_F^*) \]
  \[ \gamma \sim 1 \text{ J/mol-K}^2 \propto m^* \quad (m^* \sim 10^2-10^3 \text{ m}_e, \quad T_F^* \sim 1-10 \text{ K}) \]

- Heavy fermion superconductors \((T_c \sim 1 \text{ K})\)
  \(\text{CeCu}_2\text{Si}_2, \text{ UBe}_{13}, \text{ UPt}_3, \text{ URu}_2\text{Si}_2, \text{ UNi}_2\text{Al}_3, \text{ UPd}_2\text{Al}_3\)

- Anisotropic superconductivity
  \(\Delta(k)\) vanishes at points or lines on Fermi surface
  SC’ing properties \(\sim T^n\) for \(T \ll T_c\) [instead of \(\exp(-\Delta/T)\)]
  Electron pairing mediated by spin fluctuations

- SC coexists with weak AFM \((\mu \sim 10^{-2} \mu_B)\)

- Multiple superconducting phases
  Coupling between multicomponent superconducting order parameter and AFM order parameter (e.g., \(\text{UPt}_3\))
  Complex superconducting phase diagrams
  \(\text{UPt}_3\quad \text{H}, \text{T}, \text{P}\)
  \(\text{U}_{1-x}\text{Th}_x\text{Be}_{13}\quad \text{T}, \text{x}, \text{P}\)

- Chemical substitution
  Suppresses SC and weak AFM
  Induces local moment AFM or FM \((\mu \sim 1 \mu_B)\)
  \(\text{UPt}_3\) — Th for U; Pd, Au for Pt \(\Rightarrow\) local moment AFM
  \(\text{URu}_2\text{Si}_2\) — Rh for Ru \(\Rightarrow\) local moment AFM
  Re, Tc for Ru \(\Rightarrow\) local moment FM
Specific heat/temperature (joules/mole-K^2)

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

\( \Rightarrow m^* \sim 10^3 m_e \)

Origin: Kondo effect

CeCu2Si2, Steglich et al. '79

UBe13, Ott et al. '83
Magnetic susceptibility of selected heavy fermion compounds

\[ \chi^{-1} \text{ (mole/emu)} \]

\[ \chi \text{ (emu/mole)} \]

\[ T \text{ (K)} \]

After Fisk, Ott, Rice & Smith 86
Electrical resistivity of selected heavy fermion compounds

After Fisk, Ott, Rice & Smith 86
Fermi liquid aspects of heavy fermion metals

Wilson-Sommerfeld ratio $R$

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

$R \approx 1$ for free electrons

Fermi liquid (Landau)

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

$\chi \sim m^*/(1 + F_0)$

$A T^2 \sim (T/T^*)^2 \sim \gamma^2 T^2$

$\Delta \rho = A T^2$
Electron-electron scattering

Heavy quasiparticles

\[ \rho_{el-el}(T) \sim (T/T_F)^2 \sim AT^2 \sim \gamma^2 T^2 \]

\[ \Rightarrow A \sim \gamma^2, \ln A \propto 2\ln \gamma \]

Kadowaki-Woods plot '86
Heavy fermion superconductors

<table>
<thead>
<tr>
<th>Material</th>
<th>$T_c$ (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CeCoIn$_5$</td>
<td>2.3</td>
</tr>
<tr>
<td>* CeCu$_2$Si$_2$</td>
<td>0.49</td>
</tr>
<tr>
<td>CeIrIn$_5$</td>
<td>0.4</td>
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<tr>
<td>U$_6$Fe</td>
<td>3.7</td>
</tr>
<tr>
<td>* UPd$_2$Al$_3$</td>
<td>2.0</td>
</tr>
<tr>
<td>* URu$_2$Si$_2$</td>
<td>1.5</td>
</tr>
<tr>
<td>* UNi$_2$Al$_3$</td>
<td>1.0</td>
</tr>
<tr>
<td>UBe$_{13}$</td>
<td>0.85</td>
</tr>
<tr>
<td>* UPt$_3$</td>
<td>0.55</td>
</tr>
<tr>
<td>* URhGe</td>
<td>0.4</td>
</tr>
<tr>
<td>PrOs$<em>4$Sb$</em>{12}$</td>
<td>1.8</td>
</tr>
<tr>
<td>PuCoGa$_5$</td>
<td>18</td>
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</table>

Superconducting under pressure:

<table>
<thead>
<tr>
<th>Material</th>
<th>$T_c$ (K)</th>
<th>$P$ (kbar)</th>
</tr>
</thead>
<tbody>
<tr>
<td>* CeRhIn$_5$</td>
<td>2.2</td>
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<tr>
<td>* Ce$_2$RhIn$_8$</td>
<td>2</td>
<td>23</td>
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<tr>
<td>* CeCu$_2$Ge$_2$</td>
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<td>* CePd$_2$Si$_2$</td>
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<td>* CeRh$_2$Si$_2$</td>
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<td>CeNi$_2$Ge$_2$</td>
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<tr>
<td>* CeIn$_3$</td>
<td>0.17</td>
<td>25</td>
</tr>
<tr>
<td>* UGe$_2$</td>
<td>0.7</td>
<td>10</td>
</tr>
</tbody>
</table>

* Magnetic order
Maple, Chen, Lambert, Fisk, Smith, Ott, Brooks, Naughton '85
Anisotropic SC: $\Delta(k)$ vanishes at points or lines on FS.
SC'ing properties $\sim T^\nu$
Isotropic SC (BCS): $\Delta(k) \sim \text{constant}$
SC'ing properties $\sim \exp(-\Delta/T)$
UBe$_{13}$: $\lambda \sim T^2$ (odd parity SC)

Gross, Chandrasekhar, Einzel, Andres, Hirschfeld, Ott, Bauer, Fisk, Smith '86
$T_c(K)$

$X(\%)$

$U_{1-x}Th_xBe_{13}$

$A$

$B$

$B'$

$C$

MAGNETIC

R.H. Heffner et al. '90
Two distinct SC'ing transitions (sensitive to $H$ & $P$)

Coupling between multicomponent SC'ing order parameter & AFM order parameter

AFM: $T_s \approx 5K$, $\mu \approx 0.02\mu_B/4$ (basal plane)

Aeppli et al. '88
B-phase:
(1) Point contact spectroscopy $\rightarrow$ gap-like feature
Goll et al.'93
(2) Zero field $\mu$SR $\rightarrow$ increase in internal magnetic field
Luke et al.'93
Odd-parity, spin-triplet SC'ing state $\rightarrow$ Sauls '94
URu$_2$Si$_2$: heavy electron AFM–SC
Schl&uuml;bitz et al. ’86 polycrystalline material
Palstra et al. ’85 single crystals

[Graph showing the dependence of $C/T$ on $T^2$ and $\Delta = 129$ K]

$C/T = A \exp(-\Delta/T)$

$\gamma(0)/\gamma' \approx 0.6 \Rightarrow \sim 40\%$ Fermi surface removed by SDW

BCS-type mean field transition at $T_N = 17.5$ K

$\delta C \approx A \exp(-\Delta/T); \Delta \sim 10^2$ K $\sim 10$ meV

AFM ($\mu \approx 0.02 \mu_B/U$) coexists with SC

Maple, Dalichaouch, Kohara, Rossel, Torikachvili, McElfresh, Thompson ’86
Electron tunneling $\text{UPd}_2\text{Al}_3 - \text{Pb}$

\begin{align*}
T &= 0.3 \text{K} \\
\mu_0 H &= 0.3 \text{T}
\end{align*}

![Graphs showing tunneling behavior with $V$ vs. $dI/dV$](image)

$\text{UPd}_2\text{Al}_3$, Geibel et al., '91

Moderately heavy electron

$\gamma = 140 \text{mJ/} \text{mol} \cdot \text{K}^2$

$T_N = 14.6 \text{K} \left( \mu = 0.85 \mu_B \right)$

$T_c \approx 2 \text{K}$

d-wave pairing, line nodes

gapped dispersive spin excitations with $\Delta E \approx 1.5 \text{meV}$ at magnetic Bragg point

$Q = (0, 0, \frac{1}{2})$, Sato et al., '97

Jourdan, Huth, Adrian, '98
Non-Fermi liquid behavior in f-electron materials

- **Materials** – Ce, Yb & U intermetallic compounds
  
  Ce, Yb, U – magnetic dipole, electric quadrupole moments

  *Chemically substituted materials*
  
  $La_{1-x}Ce_xCu_2Si_2$, $Y_{1-x}U_xPd_3$, $CeCu_{6-x}Au_x$, $UCu_{5-x}Pd_x$, …

  *Stoichiometric compounds*
  
  $CeNi_2Ge_2$, $YbRh_2Si_2$, $UBe_{13}$ (P = 0)
  
  $CeIn_3$, $CePd_2Si_2$ (high P)

- **Physical properties at low $T \ll T_0$**

  Non-Fermi liquid (NFL) behavior: characterized by weak power law, logarithmic divergences in $T$

  $T \ll T_0$:

  - $\rho(T) \approx \rho(0)[1 - a(T/T_0)^n]$ \hspace{1cm} (1 \leq n \leq 1.5; \hspace{0.2cm} a > 0 \text{ or } a < 0, \hspace{0.2cm} |a| \sim 1)
  
  - $C(T)/T \approx -(1/T_0)ln(T/T_0)$ \hspace{0.5cm} $S(0) \sim (k_B/2)ln(2)$
  
  - $\chi(T) \approx \chi(0)[1 - c(T/T_0)^n]$ \hspace{1cm} (n \sim 0.5; \hspace{0.2cm} c \sim 1)

  
  $\chi(T) \approx -(1/T_0)ln(T/T_0)$

  $\chi(T) \approx C/(T^\alpha + \theta)$

- $\chi''(\omega, T)$: $\omega/T$ scaling

  T-dependence below $T_0$:

  * Appreciable $\Rightarrow$ lower energy scale than Fermi liquid (FL)
  * Scales with $T_0$
• Magnetic or charge degrees of freedom

• Two scenarios

  Single ion

  Unconventional Kondo effect (multichannel?)

  Inter-ionic interactions

  Fluctuations of OP in vicinity of $x_c$ or $P_c$ where magnetic or quadrupolar phase transition vanishes

• Atomic disorder (distribution of $T_K$'s; Griffiths' phase)

• Stoichiometric f-electron compounds under pressure

  SC observed in narrow range of $P$ in vicinity of $P_c$ where $T_m \to 0$ K in single crystal specimens with $l >> \xi_0$

  **AFM:** CePd$_2$Si$_2$, CeIn$_3$   Cambridge

  CeCu$_2$Ge$_2$                   Geneva

  CeNi$_2$Ge$_2$                  Dresden, Cambridge

  **FM:** UGe$_2$                  Cambridge, Grenoble

  ZrZn$_2$                        Karlsruhe, Cambridge
Models of NFL behavior in f-electron materials

**Single ion**

- **Multichannel Kondo effect** — Nozières & Blandin '80; ...

  Two-channel, spin-1/2 Kondo effect: two channels of conduction electrons "overscreen" spin-1/2 impurity ion

  ⇒ Residual spin at \( T = 0 \) K ⇒ Local NFL (single ion QCP)

  Quadrupolar Kondo effect (\( U^{4+}, \Gamma_3 \) g.s. ) — Cox '87

- **Kondo disorder** — Bernal et al. '95; Miranda et al. '96

  Local disorder ⇒ Distribution of coupling constants \( N(E_F) \)

  ⇒ Distribution of values of \( T_K \sim T_F \exp(-1/N(E_F)/\Delta) \)

  Impurities with \( T_K < T \) remain magnetic ⇒ NFL behavior

**Inter–ionic interactions**

- **Fluctuations of OP near 2\textsuperscript{nd} order phase transition at \( T = 0 \) K** — Hertz '76, Moriya '85; Millis '93; Continentino '93; ...

  QCP — groundstate changes from ordered to disordered as control parameter \((x, P)\) changed

  Quantum fluctuations in OP near QCP ⇒ NFL behavior

- **Griffith's phase** — Castro Neto, Castilla, Jones '97

  Competition between Kondo effect & RKKY interaction + disorder ⇒ Inhomogeneous system: paramagnetic FL phase (Kondo effect) & magnetic clusters (RKKY) ⇒ NFL behavior

**Other**

- **Electronic polarons plus disorder** — Liu '97

- **Proximity to disorder–induced metal–insulator transition** — Süßow et al. '00
The $M_{1-x}U_xPd_3$ ($M = Sc$, $Y$) systems

$Y_{1-x}U_xPd_3$

- **NFL behavior for $0 < x < \approx 0.2$**
  
  $\rho(T,H)$, $C(T)$, $M(T,H)$ for $0 \leq x \leq 0.55$

  Quadrupolar Kondo model

  Seaman, Maple, Lee, Ghamaty, Torikachvili, Kang, Liu, Allen, Cox '91

- **NFL behavior for $x = 0.2$**
  
  $\rho(T,H)$, $C(T,H)$, $M(T,H)$ for $x = 0.2$

  Second order magnetic phase transition at $T = 0$ K

  Andraka, Tsvelik '91

- **NFL behavior scales with $T_K$ and $x$ ($0 \leq x \leq 0.2$)**

  Single ion effect

  $\Delta\rho(T) = \Delta\rho(0)[1 - a(T/T_K)]$

  QKE: no

  $\Delta\rho(T) = \Delta\rho(0)[1 - a(T/T_K)^{1/2}]$

  $\Delta C(T)/T = -(bR/T_K)\ln[b'(T/T_K)]$

  QKE: yes

  $S(0) \approx (R/2)\ln(2)$

  QKE: yes

  $\Delta\chi(T) = \Delta\chi(0)[1 - c(T/T_K)^{1/2}]$

  QKE: yes

  Small magnetoresistance

  QKE: yes

  QKE — not applicable or needs to be extended
High-$T$ electrical resistivity

$Y_{1-x}U_xPd_3$

$\rho$ (µΩ⋅cm)

- 0.2
- 0.3
- 0.4
- 0.1
- 0.5
- $x=0$

$T$ (K)

$\Delta\rho(T)/\Delta\rho(0)$

- 0.05
- 0.1
- 0.02
- $x=0.2$

$T$ (K)

$T_x$ (K)

$0.5$ $0.6$ $0.7$ $0.8$ $0.9$ $1.0$

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$0.5$ $0.6$ $0.7$ $0.8$ $0.9$ $1.0$
\[ \chi - \chi_0 = \frac{C}{T - \Theta_{cw}}; \quad C = N \mu_{eff}^2 / 13k_b, \quad -\Theta_{cw} \approx 3 - 4 \Theta_K \]

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Graphs showing the behavior of \( Y_{1-x}U_xPd_3 \) with varying parameters. The graphs illustrate the temperature dependence of magnetic susceptibility and effective magnetic field. The upper graph depicts \( (\chi - \chi_0)^{-1} \) vs. temperature, while the lower graph shows \( -\Theta_{cw} \) vs. \( x \).
Y_{1-x}U_xPd_3 — Fermi Level Tuning (FLT) of T_K

- PES/BIS measurements (UM/UCSD — Kang et al. '89)
  \( \Rightarrow \epsilon_{5f} = (E_F - E_{5f}) \text{ increases with } x \) (by \( \sim 1 \) eV as \( x = 0 \rightarrow 1 \))

\[
\begin{align*}
E_F & \quad \epsilon_{5f} = (E_F - E_{5f}) \sim \epsilon_0 + \epsilon_1 x \quad (\epsilon_1 \approx 1 \text{ eV}) \\
E_{5f} & \quad \text{U 5f state}
\end{align*}
\]

- Substitution of U^{4+} for Y^{3+}
  \( \Rightarrow n_e, E_F, \) and \( \epsilon_{5f} = (E_F - E_{5f}) \text{ increase with } x \)

- \( J \sim -\langle V_{ki}^2 \rangle/\epsilon_{5f} < 0 \) where \( H_{ex} = -2J S \cdot \sigma(0) \)
  \( \epsilon_{5f} \text{ increases with } x \)
  \( \Rightarrow |J| \text{ decreases with } x \)
  \( \Rightarrow T_K \sim T_F \exp[-1/N(E_F)|J|] \text{ decreases with } x \)

i.e., \( T_K \sim T_F \exp[-\epsilon_{5f}/N(E_F)\langle V_{ki}^2 \rangle]
\sim T_F \exp[-(\epsilon_0 + \epsilon_1 x)/N(E_F)\langle V_{ki}^2 \rangle]
\sim T_F \exp[-\epsilon_0/N(E_F)\langle V_{ki}^2 \rangle] \exp[-\epsilon_1 x/N(E_F)\langle V_{ki}^2 \rangle]
\sim (T_K)_0 \exp[-\alpha x] \)
\[ \frac{\Delta C(T)}{T} = \frac{-bR}{T_K} \ln \left[ \frac{b'}{(T/T_K)} \right] + \gamma \]

Same form as two-channel spin-1/2 Kondo effect where
\( b = 0.251, b' = 2.44 \) and residual entropy \( S(0) = (R/2) \ln(2) \)

*Tsvelik '85; Sacramento & Schlottmann '89*

Upturn in \( \Delta C(T)/T \) below \( \sim 0.6 \) K \( \Rightarrow \) removal of residual entropy

<table>
<thead>
<tr>
<th>Compound</th>
<th>( T_K (K) )</th>
<th>( \gamma ) (mJ/mol U-K(^2))</th>
</tr>
</thead>
<tbody>
<tr>
<td>( Y_{0.9}U_{0.1}Pd_3 )</td>
<td>220</td>
<td>11.5</td>
</tr>
<tr>
<td>( Y_{0.8}U_{0.2}Pd_3 )</td>
<td>42</td>
<td>12.2</td>
</tr>
<tr>
<td>( Y_{0.8}Th_{0.1}U_{0.1}Pd_3 )</td>
<td>30</td>
<td>12.8</td>
</tr>
</tbody>
</table>
The $U_{1-x}M_xPd_2Al_3$ ($M = \text{Th, Y, La}$) systems

- **Parent compound**
  
  $UPd_2Al_3$ *C. Geibel et al. ‘91*
  
  Moderately heavy electron AFM-SC
  
  $\gamma = 140$ mJ/mol-K$^2$, $T_N = 14.6$ K, $T_c \approx 2$ K
  
  Hexagonal PrNi$_2$Al$_3$ structure

- **Neutron & X-ray scattering**
  
  AFM stacking along c-axis of FM sheets in basal plane  
  
  ($\mu \approx 0.85 \mu_B$)  
  
  *Krimmel et al. ‘92*  
  
  *Kita et al. ‘94*  
  
  *Paolasini et al. ‘94*
Superconductivity near AFM QCP accessed by application of pressure (high purity single crystal specimens)

CePd$_2$Si$_2$

- $P \approx 28$ kbar
- $\rho \approx \rho_0 + AT^{1.2}$
- $T_c \leq T \leq 40$ K

CeIn$_3$

- $P \approx 28$ kbar
- $\rho \approx \rho_0 + AT^{1.5}$
- $T_c \leq T \leq 4$ K

S. R. Julian, G. G. Lonzarich et al. '98
Superconducting ferromagnet UGe$_2$

- First pressure-induced superconducting ferromagnet (Saxena et al. '00)
- Itinerant electron FM with $T_c = 53$ K ($P = 0$)
- $\gamma \approx 35$ mJ/mol K$^2$
- $m^* \approx 20$ $m_e$ (Onuki et al. '93)
- Curie temperature suppressed at $P_c \approx 16$ kbar (Oomi et al. '98)
Polycrystalline UGe$_2$

1. $C(P,T)$: Vollmer, Pfleiderer, v. Löhneysen, Bauer, Maple '01

$\Delta C/\gamma T_c \sim 0.2$ at 15 kbar $\Rightarrow$ bulk SC ($\sim 20$ vol$\%$ at 15 kbar)

SC'ing volume increases with P

$\gamma$ increases with P; $\gamma(15\text{ kbar})/\gamma(0) \sim 3$

(similar results for single crystal; Tateiwa et al. '01)

2. $l \sim \xi_0 \sim 10^2$ Å for polycrystalline UGe$_2$ with $\rho_0 \sim 4 \mu\Omega$cm

p-wave SC? $l \sim \xi_0 \Rightarrow T_c \rightarrow 0$ K - Sr$_2$RuO$_4$ MacKenzie et al. '98

Inhomogeneous SC, FM filamentary structure?

ErRh$_4$B$_4$ – narrow range of $T$ between $T_{c2}$ & $\theta_c$
Kadowaki-Woods relation (’86)

\[ \rho = \rho_0 + AT^2 \]

\[ A = 6 \times 10^{-5} \mu \Omega \text{cm} \text{ (mol-K/mT)}^2 \chi^2 \propto \chi^2 \]
THEORY - Coexistence of SC & FM

S-wave.

K.B. Blagoev, J.R. Engelbrecht, K.S. Bedell '98, '99 T-P phase diag
H. Suhl '01
A.A. Abrikosov '01

* N.I. Karchov, K.B. Blagoev, K.S. Bedell, P.B. Littlewood '01

P-wave.

K. Levin, O. Valls '78
D. Fay, J. Appel '80
K. Machida, T. Ohmi '01
AR. Schick, W.F. Pickett '01
T.R. Kirkpatrick, D. Belitz, T. Vojta, R. Narayanan '01
Why is PrOs$_4$Sb$_{12}$ interesting?
And, what does it have to do with quantum criticality?

- Nonmagnetic heavy Fermi liquid ($\gamma \approx 500$ mJ/mol-K$^2$; $m^* \approx 50$ m$_e$)
- Unconventional superconductivity (different than that of Ce, U-based compounds)
- PrOs$_4$Sb$_{12}$: first Pr-based heavy fermion superconductor (all others: Ce, U-based)
- Formation of heavy Fermi liquid (and, possibly, superconductivity) may involve electric quadrupole fluctuations, rather than magnetic dipole fluctuations
- Pr$^{3+}$ energy level scheme in cubic CEF:
  In cubic CEF, Pr$^{3+}$ J = 4 Hund’s rule multiplet
  $\Rightarrow$ $\Gamma_1$ singlet, $\Gamma_3$ nonmagnetic doublet (quadrupole moment), $\Gamma_4$ & $\Gamma_5$ triplets
  Analysis of $\chi(T)$:
  - Ground state: $\Gamma_1$ singlet or $\Gamma_3$ doublet
  - 1st excited state: $\Gamma_5$ triplet ($\Delta \approx 10$ K)
  - 2nd & 3rd excited states: $\Gamma_4$, $\Gamma_1$ or $\Gamma_3$ ($\Delta > \sim 10^2$ K)
- Our experiments $\Rightarrow$ $\Gamma_3$ ground state (other experiments $\Rightarrow$ $\Gamma_1$ ground state)
- Hybridization between Pr$^{3+}$ localized 4f states & conduction electron states $\Rightarrow$ stage set for quadrupolar Kondo effect (2-channel, spin-1/2 Kondo effect with NFL behavior) $\Rightarrow$ quadrupolar Kondo lattice $\Rightarrow$ heavy Fermi liquid? $\Rightarrow$ SC?
- High field ordered phase (HFOP) – quadrupolar order?
- Near quadrupolar quantum critical point (QCP)?
- Analogous to occurrence of SC in heavy fermion compounds in vicinity of AFM QCP, accessed by pressure; e.g., CeIn$_3$, CePd$_2$Si$_2$
Crystal structure of the filled skutterudites $MT_4X_{12}$

Filled skutterudites: derived from binary skutterudites $TX_3$
($T = \text{Co, Rh, Ir}; X = \text{P, As, Sb}$)

Prototype $\text{CoAs}_3$: discovered in Skutterud, Norway

$M$ cations — bcc sublattice

(fill atomic cages in structure)

$T$ cations — sc sublattice

$X$ anions — distorted corner sharing octahedra centered by $T$ cation

bcc structure ($I\bar{m}-3$)

$a = 9.3068 \, \text{Å}$

W. Jeitschko &

D. J. Braun ‘77
Evidence for heavy fermion superconductivity in PrOs$_4$Sb$_{12}$

$C(T)$ of PrOs$_4$Sb$_{12}$ pressed pellet (0.6 K – 10 K)

**Normal state:**
- $\gamma = 607$ mJ/mol K$^2$
- $\theta_D = 203$ K
- $C_{\text{Sch}}(T)$: two level system
- $\Gamma_3$ doublet ground state
- $\Gamma_5$ triplet excited state
- $\Delta_{2,3} = 7.15$ K

**Superconducting state:**
- $T_c = 1.77$ K
- $\Delta C/T_c = 632$ mJ/mol K$^2$
- BCS: $\Delta C/\gamma T_c = 1.43$
- $\gamma \approx 440$ mJ/mol K$^2$

Two distinct SC’ing phases?
- $T_{c1} \approx 1.85$ K
- $T_{c2} \approx 1.70$ K
Evidence for heavy fermion superconductivity in PrOs$_4$Sb$_{12}$

Upper critical field $H_{c2}(T)$

Large initial slope.

$(-dH_{c2}/dT)_{T_c} = 19$ kOe/K

$H_{c2}(0) = 0.693(-dH_{c2}/dT)_{T_c} T_c$

$T_c = 2.2 \xi_0^2$

$\phi_0 = \frac{\pi}{2} \xi_0^2$

$\phi_0^* = 0.18 \frac{\hbar v_F}{k_B T_c}$

$\phi_0^* = 50$ meV

$\gamma = 350$ mJ/mK$^2$
Analysis of $\chi(T)$ — Pr$^{3+}$ ion in cubic CEF (LLW theory)

1. $\Gamma_3$ ground state
2. $\Gamma_1$ ground state

Pr$^{3+}$ J=4 multiplet

- $\chi(T)$ consistent with $\Gamma_1$ or $\Gamma_3$ ground state and $\Gamma_5$ excited state separated by $\delta \approx 10$ K
- Better fit with $\Gamma_3$ ground state and low-lying $\Gamma_5$ excited state

PrOs$_4$Sb$_{12}$

Temperature (K) vs. susceptibility ($\chi$ cm$^3$/mol)
H-T phase diagram of PrOs$_4$Sb$_{12}$
Neutron scattering studies of single crystal of PrOs$_4$Sb$_2$ in magnetic field along [001]
M. Khalili et al. '03

Small AFM $\mu_{\parallel}$ [010] in high field ordered phase (HFOP)
Analysis based on $T_1$, singlet ground state $\Rightarrow$
AFQ ordering in HFOP
μSR measurements on PrOs$_4$Sb$_{12}$

PrOs$_4$Sb$_{12}$, $H = 200$ Oe

Relaxation Rate $\sigma_g(T)$ (µs$^{-1}$)

Temperature $T$ (K)

$\lambda(T) = \lambda(0)[1+(\pi\Delta/2T)]^{1/2}\exp(-\Delta/T)$

$\Delta/T_c = 2.1$ (BCS: $\Delta/T_c = 1.76$) $\Rightarrow$ isotropic $\Delta(k)$

D. E. MacLaughlin et al. '02
Evidence for unconventional strong-coupling superconductivity in PrOs$_4$Sb$_{12}$ by means of Sb NQR measurements.

LaOs$_4$Sb$_{12}$:
- Coherence peak
- $T_1^{-1} \sim \exp(-\Delta/k_B T)$
- $2\Delta/k_B T_c \approx 3.2$
- Weak coupling BCS SC

PrOs$_4$Sb$_{12}$:
- No coherence peak
- $T_1^{-1} \sim \exp(-\Delta/k_B T)$ ($T < 1.3 T_c$)
- $2\Delta/k_B T_c \approx 5.3$
- Strong coupling unconventional SC

H. Kotegawa et al. 02
Thermal transport studies of superconducting gap structure

Thermal transport measurements in magnetic fields rotated relative to the crystal axes

K. Izawa et al. 02
Thermal transport studies of superconducting gap structure

Thermal transport measurements made in magnetic fields rotated relative to the crystal axes

\[ T = 0.52 \text{ K}, \quad \kappa_{zz}(H_\phi) = \kappa_0 + C_2\phi \cos 2\phi + C_4\phi \cos 4\phi \]

Two Scing phases in H-T plane
A - 6 point nodes, B - 2 point nodes

K. Izawa et al. 02
Time-reversal symmetry breaking detected by μSR measurements

- \( \Lambda \): width of internal field distribution
- \( \Lambda \): relaxation rate in zero field (ZF) and in 0.01 T

Y. Aoki et al. 03
Electrical resistivity: atypical HF behavior

- Exhibits metallic behavior not typical of most HF materials
- $T^2$ coefficient $A < \frac{1}{10}$ than that expected from Kadowaki-Woods relation
  \[ A = 1 \times 10^{-5} \left( \mu \Omega \text{cm-mol}^2 \text{-K}^2 / \text{mJ}^2 \right) \gamma^2 \approx 1.2 \mu \Omega \text{cm/K}^2 \text{ for } \gamma = 350 \text{ mJ/mol K}^2
- Behavior similar to that of PrInAg$_2$ ($\gamma \approx 7 \text{ J/mol K}^2$) $\Gamma_3$ ground state
Comparison of electrical resistivity of PrOs$_4$Sb$_{12}$ to calculation based on CEF

PrOs$_4$Sb$_{12}$

\[ \rho(T) = \rho_{\text{imp}} + A \rho_{\text{La}} + \rho_0 [x \rho_{\text{M}} + (1-x)\rho_{\text{A}}] \]

- $\rho_{\text{imp}} = 1.67 \, \mu\Omega \, \text{cm}$, $A = 0.208$
- $\rho_0 = 0.385 \, \mu\Omega \, \text{cm}$, $x = 0.46$
- $\Delta_{35} = 5 \, \text{K}$, $\Delta_{34} = 70 \, \text{K}$, $\Delta_{31} = 170 \, \text{K}$
High field ordered phase: \((\rho vs T)_{H}, (\rho vs H)_{T}\) for \(\text{PrOs}_4\text{Sb}_{12}\)
Specific heat $C(T,H)$ of PrOs$_4$Sb$_{12}$

$C$ (J/molK)

$T$ (K)

$B = 0$
$B = 1.5$ T ($T < 3$ K)
$B = 2$ T ($T > 3$ K)
$B = 4$ T
$B = 4.5$ T
$B = 5$ T
$B = 5.5$ T
$B = 6$ T
$B = 6.65$ T
$B = 8$ T
$B = 14$ T

R. Vollmer et al. ‘02
Similar work: Aoki et al. ‘02