Heavy Fermion Scaling
Uranium vs. Cerium and Ytterbium Compounds

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OUTLINE

Motivation: Compare scaling in U vs Ce and Yb paramagnetic heavy Fermions (HF)
Are spin fluctuations (SF) of itinerant 5f electrons different than SF for local 4f moments?

Low-T Scaling: $\chi(0)$ and $\gamma \sim 1/T_{sf}$
where $\gamma = C_{mag}/T$ is the linear coefficient of specific heat
$\chi(0)$ is the low temperature susceptibility
$k_B T_{sf}$ is the characteristic energy for spin fluctuations
$k_B T_{sf} = E_{\text{max}}$, the maximum in the inelastic magnetic neutron scattering spectrum $\chi''(E)$

$\Rightarrow$ The (Wilson) ratio $\chi(0)/\gamma$ and the product $E_{\text{max}} \gamma$ are constants.
(but depend on the $f$-orbital degeneracy $N_J = 2J+1$)

Compare data to a rough phenomenology of low temperature scaling
(spin fluctuation with degeneracy $N_J$ at energy $k_B T_{sf}$)

and to T-dependent scaling of Kondo/Anderson impurity model (K/AIM):

Rare earth (RE) HF: Low-T scaling good; K/AIM good for for T- dependence of $\chi(T)$ and $C(T)$ and $E$-dependence of $\chi''(E)$. (Example: YbFe$_2$Zn$_{20}$)

U- HF: Low-T scaling not so good ; K/AIM poor for $\chi(T)$ and $C(T)$; badly overestimates the entropy. (Example: UCo$_2$Zn$_{20}$ and URu$_2$Zn$_{20}$)

Fix up low-T scaling for U-HF:
Subtract out estimated contribution to $\gamma$ from antiferromagnetic (AF) fluctuations
Since paramagnetic moment and associated entropy are not well-known for itinerant 5f compounds, estimate $N_J$ from measured high-T Curie constant.

How good is this? What does theory have to say?
**General scaling behavior:** $\text{UCo}_2\text{Zn}_{20}$ vs. $\text{URu}_2\text{Zn}_{20}$

\[ \frac{\chi(2K)_\text{Co}}{\chi(2K)_\text{Ru}} = 2.63 \quad \frac{\chi(2K)_\text{Co}}{\chi(2K)_\text{Ru}} = 3.01 \quad \frac{E_{\text{max}}(\text{Ru})}{E_{\text{max}}(\text{Co})} = 2.84 \]

\[ \frac{\gamma(T_{\text{max}})_\text{Co}}{\gamma(2K)_\text{Ru}} = 2.93 \]

**Approximate factor of three scaling:** $\chi$ and $\gamma$ scale as $1/k_B T_{\text{sf}} = 1/E_{\text{max}}$. 
Poor but happy phenomenology of scaling

\[ N_J = 2J + 1 \text{ degenerate magnetic excited state at } E = k_B T_{sf} \]
(the characteristic energy for spin fluctuations)

Nonmagnetic singlet ground state

Damped oscillator spin fluctuations
(Fermionic, with \( \chi'' \sim E \) at small \( E \))
\[
\chi''(E) = \chi(0) E (\Gamma/\pi) \frac{(E-E_0)^2 + \Gamma^2}{(E-E_0)^2 + \Gamma^2}
\]
\( E_0 = k_B T_{sf}; \Gamma = \text{SF damping rate} \)

Van Vleck-like susceptibility
\[
\chi(0) \sim C_J / T_{sf} \text{ at low } T; \chi(T) \sim C_J / (T + T_{sf}) \text{ at high } T
\]
\[
C_J = g^2 J(J+1) \mu_B^2 / 3k_B
\]

\( C_{sh} \): like Schottky
but Fermionic, so \( C \sim \gamma T \) at low \( T \)
\( (C_K \): Kondo approximation)

“Rough Approximation”:  
“Half the spin entropy by \( T_m^C \)”:
\[
\gamma T_m^C = \frac{1}{2} R \ln(2J+1)
\]
Together with \( T_m^C \sim 1/3 T_{sf} \) (as for Schottky)
\[
\Rightarrow \gamma T_{sf} = \frac{3}{2} R \ln(2J+1)
\]
Scaling in Kondo/Anderson Impurity Model:

- Low energy Kondo resonance on a scale $k_B T_0$
- Universal behavior of $C_m(T)$, $\chi(T)$ and $\chi''(\Delta E)$.
- Scaling dependent on orbital degeneracy $N_J = 2J + 1$.

- Rajan’s (Coqblin-Schrieffer) results for low-T scaling [1]:
  \[ \gamma_0 = \frac{\pi J R}{3T_0} \]
  \[ \chi_0 = \frac{(2J + 1)C_J}{2 \pi T_0} \]

- Cox’s results (NCA) for the neutron scattering cross section:
  \[ E_{\text{max}} = 1.36 T_0 \text{Cox} = 1.36 \left( \frac{T_0}{1.15} \right) = 1.18 T_0 \]
  Depends on $J$, 4f occupation $n_f$ (degree of intermediate valence).
  We assume $E_{\text{max}} = T_0$ is valid to 20% 

**Low T scaling laws:**

1) \(E_{\text{max}}\gamma\)

Rough: \((3/2)R \ln(2J+1)\)

Kondo: \((\pi/3)RJ\)

<table>
<thead>
<tr>
<th>J</th>
<th>Rough</th>
<th>Kondo</th>
</tr>
</thead>
<tbody>
<tr>
<td>1/2</td>
<td>8.6</td>
<td>4.35</td>
</tr>
<tr>
<td>5/2</td>
<td>22.3</td>
<td>21.75</td>
</tr>
<tr>
<td>7/2</td>
<td>25.9</td>
<td>30.45</td>
</tr>
<tr>
<td>9/2</td>
<td>28.7</td>
<td>39.15</td>
</tr>
</tbody>
</table>

2) \(W = (\pi^{2}R/3C_{J}) \chi(0)/\gamma\)

Rough: \((2\pi^{2}/9)/\ln(2J+1)\)

Kondo: \((1+(1/(2J)))\)

<table>
<thead>
<tr>
<th>J</th>
<th>Rough</th>
<th>Kondo</th>
</tr>
</thead>
<tbody>
<tr>
<td>1/2</td>
<td>3.15</td>
<td>2.00</td>
</tr>
<tr>
<td>5/2</td>
<td>1.22</td>
<td>1.20</td>
</tr>
<tr>
<td>7/2</td>
<td>1.05</td>
<td>1.14</td>
</tr>
<tr>
<td>9/2</td>
<td>0.95</td>
<td>1.11</td>
</tr>
</tbody>
</table>

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**Low T Scaling in Ce, Yb and U compounds**

<table>
<thead>
<tr>
<th>Compound</th>
<th>(\gamma(J/\text{mol-K}^{2}))</th>
<th>(E_{0}(\text{K}))</th>
<th>(\gamma E_{0}(J/\text{mol-K}))</th>
<th>(\chi(0)/\text{(emu/mol)})</th>
<th>Wilson</th>
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<tbody>
<tr>
<td>CeRu₂Si₂</td>
<td>0.35</td>
<td>23.2</td>
<td>8.12</td>
<td>0.016</td>
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<tr>
<td>Ce₃In</td>
<td>0.7</td>
<td>17</td>
<td>11.9</td>
<td>0.06</td>
<td></td>
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<tr>
<td>CeCu₂Si₂</td>
<td>1.1</td>
<td>8.7</td>
<td>9.57</td>
<td>0.008</td>
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<tr>
<td>CeCu₅</td>
<td>1.6</td>
<td>3.5</td>
<td>5.6</td>
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<tr>
<td>CeAl₃</td>
<td>1.62</td>
<td>7.66</td>
<td>12.4092</td>
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<tr>
<td>CePd₃</td>
<td>0.03</td>
<td>700</td>
<td>21</td>
<td>0.002</td>
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<tr>
<td>CeSn₃</td>
<td>0.042</td>
<td>491</td>
<td>20.622</td>
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<td>YbAl₃</td>
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<td>634</td>
<td>25.36</td>
<td>0.005</td>
<td>1.32</td>
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<td>YbInCu₄</td>
<td>0.041</td>
<td>507</td>
<td>20.787</td>
<td>0.006</td>
<td>1.54</td>
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<tr>
<td>YbMgCu₄</td>
<td>0.053</td>
<td>545</td>
<td>28.885</td>
<td>0.004</td>
<td>0.8</td>
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<tr>
<td>YbAgCu₄</td>
<td>0.199</td>
<td>139</td>
<td>27.661</td>
<td>0.017</td>
<td>0.9</td>
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<tr>
<td>YbFe₂Zn₂₀</td>
<td>0.52</td>
<td>70</td>
<td>36.4</td>
<td>0.05</td>
<td>1.02</td>
</tr>
<tr>
<td>UA₁₂</td>
<td>0.14</td>
<td>243</td>
<td>34.02</td>
<td>0.004</td>
<td>0.48</td>
</tr>
<tr>
<td>USn₃</td>
<td>0.17</td>
<td>60</td>
<td>10.2</td>
<td>0.01</td>
<td>0.99</td>
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<tr>
<td>URu₂Zn₂₀</td>
<td>0.188</td>
<td>191</td>
<td>35.908</td>
<td>0.012</td>
<td>1.08</td>
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<tr>
<td>UPt₃</td>
<td>0.45</td>
<td>70</td>
<td>31.5</td>
<td>0.009</td>
<td>0.34</td>
</tr>
<tr>
<td>UC₁₂Zn₂₀</td>
<td>0.558</td>
<td>70</td>
<td>39.06</td>
<td>0.047</td>
<td>1.42</td>
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<tr>
<td>UBe₁₃</td>
<td>1.1</td>
<td>150</td>
<td>165</td>
<td>0.015</td>
<td>0.23</td>
</tr>
</tbody>
</table>

Note: Rough Approximation gives values close to Kondo for \(E_{\text{max}}\gamma\) and \(W\).
Temperature-dependent scaling in Kondo model

Temperature dependence of $\chi$ and $C_m$ for different $J$ [Rajan]

Energy dependence of the dynamic susceptibility at different temperatures. $\Gamma_{\text{neutron}}$ is the peak position $E_{\text{max}}$ [Cox].
The validity of the AIM for the rare earth 4f compound YbFe\textsubscript{2}Zn\textsubscript{20}

Susceptibility and specific heat:

- Symbols: Data for $C_{\text{mag}}(T)$ and $\chi(T)$ (M. S. Torikachvili et al., PNAS, 104, 9960 (2007)).
- Red line: Kondo (Rajan) for J=7/2.

Neutron scattering (Christianson):

- Low T peak in $\chi''(E)$
- $E_{\text{max}} = 1.18$ $T_0 = 82K=7$ meV.
- Red line: $\chi''(\Delta E)/\chi''(E_{\text{max}})$ was as determined from Fig. 4 of Cox

- Only one adjustable parameter ($T_0$) ($T_0 = 69.2$ K.)
- WOW!
**YbFe\(_2\)Zn\(_{20}\) compared to UCo\(_2\)Zn\(_{20}\)**

Susceptibility, specific heat coefficient are very similar in magnitude and temperature dependence; neutron spectra have very similar \(E_{\text{max}}\).

![Graphs showing T(K), \(\chi(\text{emu/mol})\), \(C/T(\text{J/mol-K})\), \(S_{\text{mag}}(\text{arb})\), and \(\Delta E(\text{meV})\) for YbFe\(_2\)Zn\(_{20}\) and UCo\(_2\)Zn\(_{20}\).]

**Since Kondo works so well for YbFe\(_2\)Zn\(_{20}\), we next examine how it works for UCo\(_2\)Zn\(_{20}\)**

Note: URu\(_2\)Zn\(_{20}\) and YbFe\(_2\)Zn\(_{20}\) are **good candidates for studying the K/AIM** in periodic \(f\) compounds. The \(f\)-atom content is less than 5% of the total number of atoms, the shortest \(f\)-atom/\(f\)-atom spacing is \(\sim 6\)Å, and the \(f\)-atom is surrounded by a nearly-spherical cage of Zn atoms (in cubic site symmetry) which yields small crystal field splitting.
K/AIM Scaling for UCo$_2$Zn$_{20}$ and URu$_2$Zn$_{20}$:

Use Rajan and measured value of $\gamma$

- For each $J$, estimate $T_0$ (called $T_K$ in the table) from $\gamma = \pi JR / 3 T_0$.
- For $J=5/2$ and $9/2$, use the free ion value Curie constant for $C_J$.
  (For $J = \frac{1}{2}$, use the measured low temperature Curie constant.)
- Determine $\chi_0 = (2J + 1)C_J / 2 \pi T_0$ and $E_{max} = 1.18 T_0$ – given in table for each $J$

<table>
<thead>
<tr>
<th></th>
<th>$T_K$(K)</th>
<th>$T_{C_{max}}$(K)</th>
<th>$\chi_0$($\text{emu/mole}$)</th>
<th>$T_{\chi_{max}}$(K)</th>
<th>$E_{max}$(meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Ru</td>
<td>Co</td>
<td>Ru</td>
<td>Co</td>
<td>Ru</td>
</tr>
<tr>
<td>Experiment</td>
<td></td>
<td></td>
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<tr>
<td>J=9/2</td>
<td>208</td>
<td>69</td>
<td>36.5</td>
<td>12.1</td>
<td>0.0125</td>
</tr>
<tr>
<td>J=5/2</td>
<td>116</td>
<td>38</td>
<td>34</td>
<td>11</td>
<td>0.0135</td>
</tr>
<tr>
<td>J=1/2</td>
<td>23</td>
<td>7.6</td>
<td>20</td>
<td>6.8</td>
<td>0.0245</td>
</tr>
</tbody>
</table>

- **J=9/2** values for $\chi_0$ and $E_{max}$ are close to experiment: **K/AIM scaling good at low T?**

Fix $J = 9/2$, fix $T_0$ at the values given above.
Calculate the full $T$-dependence for $\chi$ and $\gamma$ using Rajan’s curves
  (use Cox for the $E$-dependence of $\chi''(E)/\chi''(E_{max})$)
AIM predictions for the temperature dependence of $\chi(T)$, $C_{mag}(T)$, and $S_{mag}(T)$ in the $J = 9/2$ case:
AIM predictions for the energy dependence of $\chi''(\Delta E) / \chi''(E_{\text{max}})$:
Summary so far:

- Kondo scaling, at both low and intermediate temperature, works extremely well for certain rare earth (RE) HF compounds. (YbAgCu$_4$; YbFe$_2$Zn$_{20}$)

-- For UAl$_2$, USn$_3$, UPt$_3$ and UBe$_{13}$ either $W = (\pi^2 R/3C_J) \chi(0)/\gamma$ or $E_{\text{max}} \gamma$ is wrong, i.e. too small or too large for $J = 9/2$ (or 8/2) low-T scaling.

For these uranium compounds, the susceptibility is always in the range 0.01 emu/mol, even though the specific heat $\gamma$ varies by an order of magnitude (from 0.1 to 1 J/mol-K$^2$).

-- $J = 9/2$ Low-T Kondo scaling works well for UCo$_2$Zn$_{20}$ and URu$_2$Zn$_{20}$, but the temperature dependence predicted by Kondo theory for $J = 9/2$, has serious problems:

  Theoretical values of the peak temperature for $C_{\text{mag}}(T)$ are higher than observed in the experiment.

  Experimental entropy is much smaller than predicted.

(For UCo$_2$Zn$_{20}$ the choice $J = 1/2$ gives a better fit to the specific heat maximum temperature and entropy and fits $\chi(0)$ but **seriously underestimates** the neutron peak energy $E_{\text{max}}$ Since this is the fundamental scale $k_B T_{sf}$, this is a serious failure.)
**What’s wrong with U-HF low-T scaling?**

1) **High temperature paramagnetic moment/entropy**

In cerium and Yb HF compounds, the high temperature “effective moment” $T\chi$ tends to the (Hund’s Rule) free ion (local moment) value $C_J = g^2 J(J+1) \mu_B^2/3k_B$ and the entropy tends to $R \ln N_J$ where $N_J = 2J+1$ is the degeneracy of the moment.

For some uranium HF compounds, $T\chi$ approaches the Hund’s Rule value at high T, but for others the paramagnetic moment appears to be seriously suppressed from the free ion value.

For UPt$_3$, $T\chi$ tends towards the $L=0$, $S=1$ value, as though $L$ is completely quenched as in transition metals.

Neutron form factor measurements (Lander) show that the orbital moment is much smaller than the Hund’s rule value in such compounds as UNi$_2$ and UFe$_2$.

The Curie constant and the entropy enter the scaling laws (in the rough approximation) as $E_{max, \gamma} \sim (3/2) R \ln N_J$ and $W = (\pi^2 R/3C_J) \chi(0)/\gamma$.

Hence we might improve agreement with scaling by using better values for $C_J$ and $N_J$.

**Problem:** For Hund’s Rule $J$ or for $L=0$ $S=1$ (or 3/2), we know how to determine $C_J$ and $N_J$ but for partially quenched $L$, what is $N_J$?
What’s wrong with U-HF low-T scaling? (continued)

2) Antiferromagnetic fluctuations in uranium compounds

In addition to the spin fluctuation peak at $E_{\text{max}} \sim 10 - 20$ meV several U-HF compounds exhibit a low energy peak at $E_{\text{AF}} \leq 1$ meV with a $Q$-dependence characteristic of antiferromagnetic (AF) fluctuations near a quantum critical point (QCP) for a transition into a nearby AF phase.

Example: UPt$_3$

Main spin fluctuation at $E_{\text{max}} \sim 6$ meV

This shows a moderate $Q$-dependence – a little larger at the zone boundary

AF fluctuation at $E_{\text{AF}} \sim 0.2$ meV

Sharp $Q$-dependence: narrow peak near $(1/2, 0, 1)$

Peak in intensity near $T_N = 5$K, disappears above 20K

[Graphs and figures showing experimental data and theoretical predictions are included here.]

FIG. 1. Constant-energy scans along $(0,0,1)$ above and below $T_N$. (a) Taken at an energy transfer of 0.5 meV. Note that while the scattering at 30 K no longer peaks at $(\pm 1, 0, 1)$, its level is higher than that seen between the maxima for $T=1.8$ K. (b) Elastic scattering with a PG and Be filtered beam. The line through these points is a resolution-corrected Lorentzian. The horizontal bar indicates the spectrometer resolution as measured by a/2 scattering from the nuclear $(10\bar{2})$ Bragg reflection.
What’s wrong with U-HF low-T scaling? (continued)

2) (continued) AF fluctuations and the specific heat:

Such AF fluctuations can contribute to the low temperature specific heat, enhancing it above the value expected from the scaling relation $\gamma = (\pi/3) R J/ E_{\text{max}}$. Assuming that the susceptibility is not strongly affected by AF fluctuations, this means that the Wilson ratio of $\chi/\gamma$ will be much smaller, and the product $\gamma E_{\text{max}}$ much larger, than expected.

To correct for this, we can try to separate $C/T$ into two regions: a low temperature region (say $T < 10$ K) where $C/T$ increases rapidly with decreasing temperature due to AF fluctuations, and a region at higher temperature (say $10 < T < 20$ K) where the linear coefficient $\gamma_{\text{HiT}}$ corresponds to the value arising from the spin fluctuation at $E_{\text{max}} = k_B T_{sf}$.
Example: UBe$_{13}$

\[ \chi(0) = 0.015 \text{ emu/mol} \]
\[ \gamma = 1.1 \text{ J/mol-K}^2 \]
\[ E_{\text{max}} = 13 \text{ meV} = 150 \text{ K} \]

\[ \Rightarrow \]
\[ W = (\pi^2 R/3C_J) \chi(0)/\gamma = 0.23 \]
\[ E_{\text{max}} \gamma = 165 \text{ J/mol-K} \]

But for \( \gamma_{\text{HiT}} = 0.1825 \text{ J/mol-K}^2 \)

\[ W = (\pi^2 R/3C_J) \chi(0)/\gamma_{\text{HiT}} = 1.38 \]
\[ E_{\text{max}} \gamma_{\text{HiT}} = 27.4 \text{ J/mol-K} \]

Notes:

a) These values are much closer to the expected values in the rough approximation for the \( J = 9/2 \) case where \( W = 1.11 \) and \( E_{\text{max}} \gamma = 28.7 \).

b) Use of the \( J = 9/2 \) Curie constant appears reasonable at high T.

The Curie Weiss \( \theta \approx E_{\text{max}}/k_B \).

c) The AF fluctuations account for most of \( \gamma \) at low-T, affecting \( \gamma \) only below 8 K and generating only 25% of R ln2.

Neutrons see \( \Gamma_{\text{AF}} \sim 1-2 \text{ meV for T < 20 K} \)
Example: UAl$_2$

\[ \chi(0) = 0.004 \text{ emu/mol} \]
\[ \gamma = 0.14 \text{ J/mol-K}^2 \]
\[ E_{\text{max}}/k_B = 243 \text{K} \]

\[ \Rightarrow \text{for } J = 9/2 \ (C = 1.62 \text{ emu-K/mol}) \]
\[ W = (\pi^2 R/3C_J) \chi(0)/\gamma = 0.48 \]
\[ E_{\text{max}} \gamma = 34 \text{ J/mol-K} \]

But for \( \gamma_{\text{HiT}} = 0.07 \text{ J/mol-K}^2 \)
and \( C_{\text{HiT}} = 1.2 \text{ emu-K/mol} \)

\[ \Rightarrow W = (\pi^2 R/3C_J) \chi(0)/\gamma_{\text{HiT}} = 1.30 \]
\[ E_{\text{max}} \gamma_{\text{HiT}} = 17.0 \text{ J/mol-K} \]

Notes:

a) The Curie constant is smaller than the \( J = 4 \) or 9/2 value (1.62 emu-K/mol) but a little larger than the \( L=0 \ S=1 \ g =2 \) value of 1.00 emu-K/mol.

b) If we take this to mean that the degeneracy \( N_J \) is a little larger than 3, then \( E_{\text{max}} \gamma \) should be a bit larger than 13.7 (the rough appx \( J = 3 \) value) and \( W \) should a bit smaller than 1.5 (the Kondo \( J=3 \) value)

c) The Curie Weiss \( \theta = 250 \text{ K} \), the low temperature \( T_{\text{sf}} = 300 \text{K} \) in \( \chi(T) = \chi(0) \ (1-(T/ T_{\text{sf}})^2) \) and the low-T value of \( C_{\text{HiT}}/ \chi(0) \) all correspond to \( E_{\text{max}}/k_B \)

d) The AF fluctuations have not been seen by neutrons. We assume they arise below 10K and account for half of the low-T \( \gamma \); they generate entropy of order only 5% of \( R \ln 2 \).
Example: UPt$_3$

$\chi(0) = 0.009$ emu/mol
$\gamma = 0.45$ J/mol-K$^2$
$E_{\text{max}}/k_B = 70$ K

$\Rightarrow$ for $J = 9/2$ ($C = 1.62$ emu-K/mol)  
$W = (\pi^2 R/3C_J) \chi(0)/\gamma = 0.34$
$E_{\text{max}} \gamma = 31.5$ J/mol-K

But for $\gamma_{\text{HiT}} = 0.225$ J/mol-K$^2$
and $C_{\text{HiT}} = 1$ emu-K/mol

$W = (\pi^2 R/3C_J) \chi(0)/\gamma_{\text{HiT}} = 1.10$
$E_{\text{max}} \gamma_{\text{HiT}} = 15.7$ J/mol-K

Notes:

a) The Curie constant is equal to the $L=0$ $S=1$ $g =2$ value of 1.00 emu-K/mol.

b) If we take this to mean that the degeneracy $N_J$ is 3, then $E_{\text{max}} \gamma$ should be 13.7 (the rough appx $J = 3$ value) and $W$ should a bit smaller than 1.5 (the Kondo $J=3$ value).

c) The Curie Weiss $\theta = 60$ K, and the low-T value of $C_{\text{HiT}}/ \chi(0) = 111$ K correspond to $E_{\text{max}}/k_B$

d) As shown above, the AF fluctuations arise below 20K; they account for half of the low-T $\gamma$; they generate entropy of order 20% of R ln2. They do not peak at $T_N = 5$ K.
Antiferromagnetic uranium compounds:

Compounds such as $\text{U}_2\text{Zn}_{17}$ exhibit antiferromagnetic order with healthy ($\sim 1 \mu_B$) magnetic moments. These compounds show both critical scattering from the AF fluctuations on a small ($\sim 1$ meV) scale and demagnetizing spin fluctuations on a larger energy scale ($\sim 10$ meV). In both cases, there is a large linear coefficient of specific heat as $T \to 0$ in the AF phase. We assume that this arises from the high energy spin fluctuation, and analyze the specific heat and scaling laws in a similar manner as in UBe$_{13}$ and UAl$_2$ by trying to separate the contribution of the AF fluctuations from the low-T specific heat.
Example: $U_2Zn_{17}$: An HF antiferromagnet

$\chi_{\text{max}} = 0.0125 \text{ emu/mol}$

$\gamma_{Hi} = 0.55 \text{ J/mol-K}^2$

$\gamma_{Lo} = 0.20 \text{ J/mol-K}^2$

$E_{\text{max}}/k_B = 100 \text{ K}$

$\Rightarrow$ for $J = 9/2$ ($C = 1.62 \text{ emu-K/mol}$)

$W = (\pi^2 R/3C_J) \chi(0)/\gamma_{Lo} = 1.05$

$E_{\text{max}} \gamma_{Lo} = 20 \text{ J/mol-K}$

Notes:

a) The high-T behavior of $T\chi$ suggests free ion $f^2$ (or $f^3$) behavior. A Curie Weiss law with $\theta = 100 \text{ K}$ and $C(f^2)$ is close to the data and corresponds to $E_{\text{max}}/k_B = 100\text{K}$.

b) The AF fluctuations ($T_{AF} \sim 1 \text{ meV}$) appear above $T_N = 9.7\text{K}$; the AF ordering wipes out more than half of the low-T $\gamma$. The entropy at $T_N$ is close to $R \ln 2$.

c) If we use $\gamma_{Hi}$ in the scaling laws, then $W = 2.88$ and $E_{\text{max}} \gamma_{Lo} = 55 \text{ J/mol-K}$; both values are much too large. If we use $\gamma_{Lo}$, then the Wilson ratio is good but $E_{\text{max}} \gamma_{Lo}$ is a little small. The AF transition has removed some (but not most) of the entropy that is associated with the 100K spin fluctuation, which remains as a large linear coefficient below $T_N$. 
Repairing low-T scaling in U-HF compounds:

1) Separation of AF contribution to C/T: In UBe$_{13}$, UPt$_3$ and UAl$_2$, the values of the Wilson ration $W$ and the scaling product $E_{\text{max} \gamma}$ can be brought in line with the rough scaling approximation if we use the value of specific heat coefficient $\gamma_{\text{HiT}}$ extrapolated for $10 < T < 20$ K. We assume that the upturn in $C/T$ below $10$K is due to AF fluctuations, carrying only a fraction of $R \ln 2$ for entropy.

2) Estimating the moment degeneracy: In correcting these scaling parameters, we have to estimate the moment degeneracy $N_J$. We use the measured High T Curie constant $C_{\text{HiT}}$. Since, for UPt$_3$ and UAl$_2$, $C_{\text{HiT}}$ is of order of the $L=0$, $S=1$, $g=2$ value, we assume that the degeneracy $N_J$ that goes in the formulae for $W$ and $E_{\text{max} \gamma}$ is of order $3$ – or, rather, considerably less than the $f^2$ ($f^3$) Hund’s rule values 9-10.

3) Validity in U antiferromagnets: The procedure also seems to be valid in U$_2$Zn$_{17}$, where there is AF order at 10 K, and where there are both low energy scale AF fluctuations and higher energy scale spin fluctuations seen by neutrons. We attribute the large coefficient of specific heat seen for $0 < T < T_N$ to the presence of high energy scale spin fluctuations in the AF phase.
How good is all this?

1) No distinction between itinerant and local moment low-T scaling: The main ingredients of the “rough approximation” are the energy scale for and the degeneracy of the spin fluctuations. As such, the low-T scaling that it predicts does not distinguish between itinerant and local spin fluctuations. To distinguish the two cases, we need to look elsewhere – e.g. in the Q-dependence of the spin fluctuations. For the main (higher energy) spin fluctuation in uranium compounds, this has only been accomplished for UPt$_3$. Direct comparison with appropriate rare earths would be very compelling: e.g. UCo$_2$Zn$_{20}$ vs YbFe$_2$Zn$_{20}$ or USn$_3$ vs CeSn$_3$.

2) Difficulty of direct determination of magnetic specific heat: We have assumed the rough approximation to be valid on the 25% level. To test it directly would require measuring the specific heat over the range $0 < T < E_{\text{max}}/k_B$, and showing that the full $R \ln(N_J)$ entropy is generated on this scale. Unfortunately it is difficult to subtract the nonmagnetic contribution to the specific heat accurately for $T > 20K$, since this involves a small difference between two large numbers, e.g. $C(\text{UCo}_2\text{Zn}_{20}) - C(\text{ThCo}_2\text{Zn}_{20})$.

3) UCo$_2$Zn$_{20}$ as a strong violation of the rough approximation: The spin fluctuation energy in this compound is small enough (60-70K) that most of the expected $R \ln 10$ entropy should be observed for $T < 20K$, but the observed entropy is much smaller than this. If a smaller moment degeneracy than $N_J = 10$ in the spin fluctuation peak is at the origin of the discrepancy then why does the $J = 9/2$ scaling work at low $T$? And, why does the high temperature Curie constant $C_{\text{HiT}}$ appear to approach the $J = 4$ or $J = 9/2$ value?
How good is all this (continued)

4) What is the moment degeneracy associated with the spin fluctuation?
We need to better understand the relation between $C_{\text{HiT}}$, $\mu_L/\mu_S$, and the degeneracy for these itinerant uranium systems. One way to determine the high temperature moment: use the neutron sum rule $\int S(Q,E) \, dQ \, dE \sim J(J+1)$

5) Proof needed that the low-T upturn in $C/T$ is due to AF fluctuations:
The existence of AF correlations in compounds such as UAl$_2$ and USn$_3$ remain to be demonstrated by neutron scattering. In compounds such as UPt$_3$, and U$_2$Zn$_{17}$ where neutrons have seen the AF fluctuations, it remains to be seen whether the upturn in the specific heat correlates quantitatively with the temperature dependence of the AF fluctuations seen in neutrons, by comparing the spectral weight in these fluctuations to the weight in the upturn in $C/T$).

6) U-HF ~ RE intermediate valence, but AF fluctuations never seen in RE-IV:
The spin fluctuation peaks seen in $\chi''(E)$ in the range 5-20 meV in uranium HF compounds are very similar in lineshape and energy scale to what is seen in intermediate valence cerium and ytterbium compounds. AF fluctuations are only seen in cerium HF compounds that have very large specific heat coefficients and very small spin fluctuation energies $E_{\text{max}} \sim 1-2$ meV. AF fluctuations are never seen in RE-IV compounds.

Perhaps, due to the itineracy of the $5f$’s, the intersite $f$-$f$ coupling energy $J$ is larger relative to $k_B T_{sf}$ for U compounds than for Ce and Yb IV compounds, so that AF order can exist in the U compounds, where it is not seen for RE-IV.
What does theory have to say?

Three kinds of theory are appropriate here:

1) **Band theory:** Norman, Albers and collaborators investigated UPt$_3$ and found:
   a) The theory gets the lattice constant and the Fermi surface topology correctly
   b) The measured specific heat is enhanced over the calculated band value by $m^*/m_b = 22$
   c) The dHvA experimental mass enhancements are in the range $m^*/m_b \sim 12$-$29$ with an average enhancement 18 for eight branches.
   d) After rescaling the energy axis by an enhancement factor of 20 ($\Delta E \rightarrow \Delta E/20$), the calculated dynamic susceptibility peaks at an energy twice as large as experiment.
   e) The theory does correctly predict that $\chi''(\Delta E)$ is largest at the (0,0,2) zone boundary.
   f) The theory strongly overestimates the magnitude of the ordered moment – giving $0.27 \mu_B$ compared to the experimental value $0.02 \mu_B$. It also does not see any enhancement at the ordering wavevector ($2\pi/\sqrt{3} a, 0, 2\pi/c$).

The point here is that adding an *ad hoc* Stoner enhancement to band theory gets the specific heat and the dynamic susceptibility in the right ballpark, and hence comes close (within a factor of 2) to the measured low-$T$ scaling $E_{\text{max}}\gamma$.

A key problem is that the band theory is not expected to be reliable for excitations or at finite temperatures. For example, the band theory has uncoupled $j = 5/2$ bands, which may not be appropriate, especially for calculating the Curie constant.
2) “SCR” theory: This widely used approach, due to Moriya, amounts to a mean field phenomenology of an AF transition near a QCP. Its starting point is a local spin fluctuation susceptibility

$$
\chi_L(E) = \frac{\chi_L}{1 - iE/\Gamma_L}
$$

which then is subjected to site-site exchange coupling $J_Q$

$$
\chi(Q,E) = \frac{\chi_L(E)}{1 - J_Q \chi_L(E)}
$$

The assumption is made that $\chi(Q,0)$ will diverge at the QCP; a finite value of $\chi(Q,0)$ is related to the “control parameter” or distance from the QCP. A small number of parameters is required to characterize the behavior.

This has been recently applied to USn$_3$. It gives appropriate scaling between $\gamma$ and $E_{\text{max}}$. Hence, the low value of $\gamma E_{\text{max}}$ observed for this compound may be due to AF fluctuations. (The Curie constant suggests that $N_J$ may also be small.) The theory assumes that the 5 meV peak seen in the neutron scattering is due to AF fluctuations, and predicts a bare spin fluctuation at 560 K (48 meV). The appropriate $Q$-dependence for a 5 meV AF peak, as well as the existence of the 48 meV peak need to be confirmed by neutrons.

**Fig. 7.** (Color online) Closed circles are the $T$ dependence of the electronic specific heat $C_p/T$ from Ref. 4, which has been estimated by subtraction of the phonon part from the total specific heat. The solid line is $C_p/T$ calculated using the spin fluctuation parameters in Table II and the equation for $C_p/T$ (Ref. 14).

**Fig. 9.** (Color online) $T$ dependence of the relaxation rate $\Gamma_{\text{neutron}}(T) - 2\pi T \gamma(T)$ estimated from $\gamma(T)$ in Fig. 8. For comparison, the relaxation rate $\Gamma(T)$ determined in neutron inelastic scattering measurements (Ref. 17) is presented.
Theory comments, continued:

3) **Anderson/Kondo Lattice**: Many versions. The theory can, in principle, handle large orbital degeneracy. However, it is not equipped to handle realistic $5f$ band dispersion. It serves best as a qualitative guide as to how the correlations work.

**A speculation:**
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--We showed above that the K/AIM worked very well for rare earth compounds such as YbAgCu$_4$ and YbFe$_2$Zn$_{20}$, but was poor for the temperature dependence of UCo$_2$Zn$_{20}$ and URu$_2$Zn$_{20}$.

--We remind the listener that in the early days of Kondo physics, strong efforts were made to have Kondo impurities in such compounds as Au$_{1-x}$Fe$_x$ on the ppm level. The argument was made – and experimental evidence given – that higher levels of impurities led to d-d coupling $J$ and spin glass or magnetic order.

--The fact that K/AIM works so well for rare earth IV compounds suggests that the intersite f-f coupling is very small for these compounds, relative to $k_B T_{sf}$, presumably due to the fact that the $4f$ electrons are so localized. Under these circumstances, the Kondo/Anderson Lattice gives results close to K/AIM behavior.

--For uranium compounds, however, the spatial extent of the $5f$ orbits should result in larger intersite coupling. Perhaps this leads to a situation where the Kondo/Anderson lattice does not look like the K/AIM, and perhaps this is at the root of the discrepancy seen between the K/AIM predictions and the temperature dependence of UCo$_2$Zn$_{20}$ and URu$_2$Zn$_{20}$. 