

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
 $k_B T_{sf} = E_{max}$, the maximum in the inelastic magnetic neutron scattering spectrum $\chi''(E)$
 \Rightarrow **The (Wilson) ratio $\chi(0)/\gamma$ and the product $E_{max} \gamma$ 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 T-dependence of $\chi(T)$ and $C(T)$ and *E*-dependence of $\chi''(E)$. (Example: $\text{YbFe}_2\text{Zn}_{20}$)

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

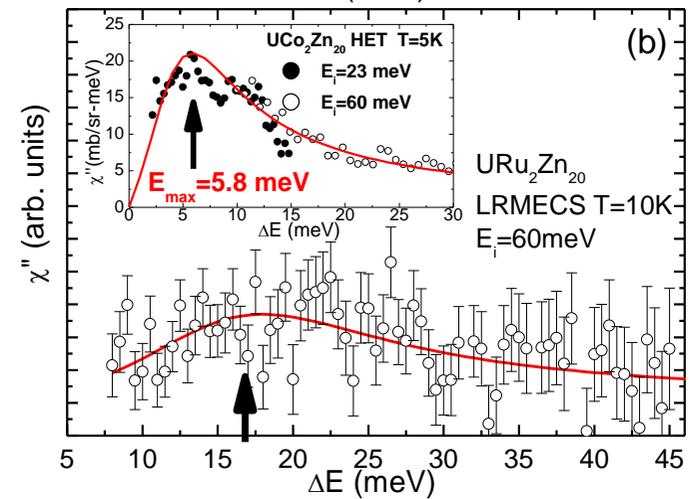
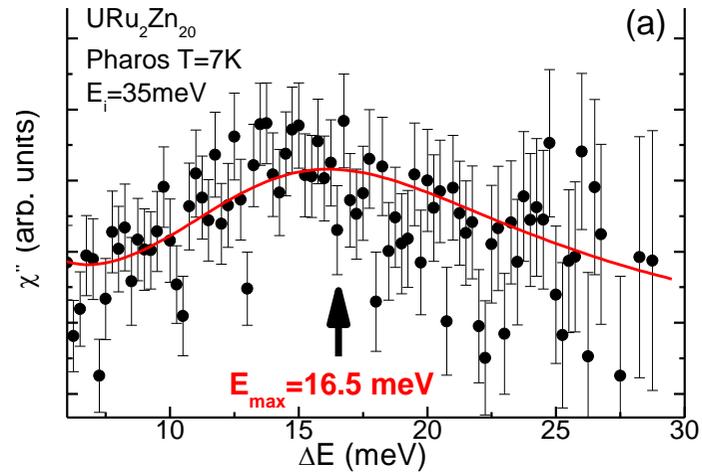
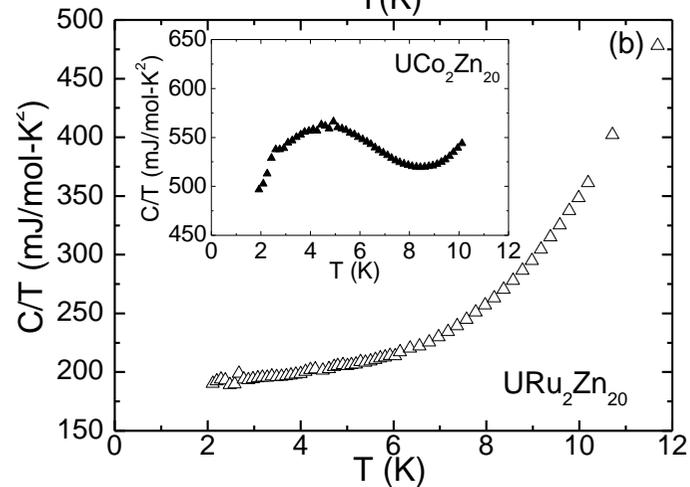
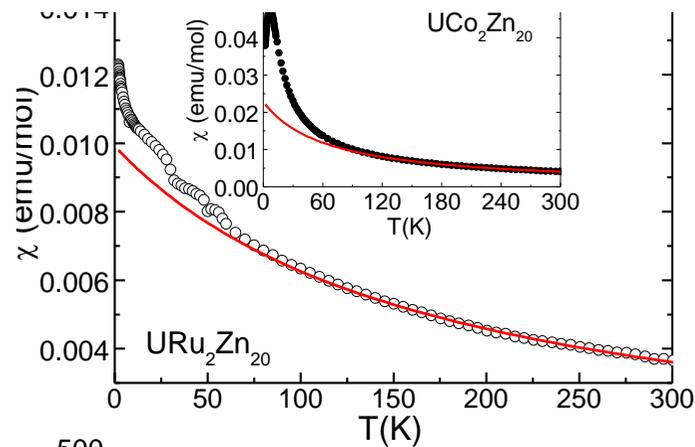
Fix up low-T scaling for U-HF:

Subtract out estimated contribution to γ 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: $U\text{Co}_2\text{Zn}_{20}$ vs. $URu_2\text{Zn}_{20}$



$$\chi(2K)_{Co} / \chi(2K)_{Ru} = 2.63$$

$$\chi(2K)_{Co} / \chi(2K)_{Ru} = 3.01$$

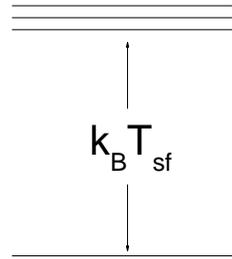
$$E_{max}(Ru) / E_{max}(Co) = 2.84$$

$$\chi(T_{max})_{Co} / \chi(2K)_{Ru} = 2.93$$

Approximate factor of three scaling: χ and γ scale as $1/k_B T_{sf} = 1/E_{max}$.

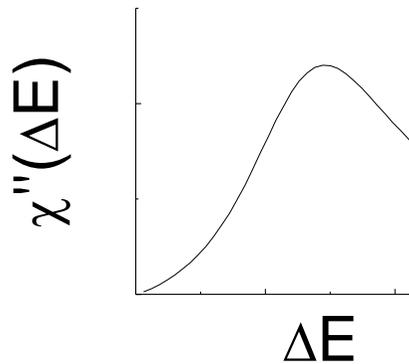
Poor but happy phenomenology of scaling

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



Nonmagnetic singlet ground state

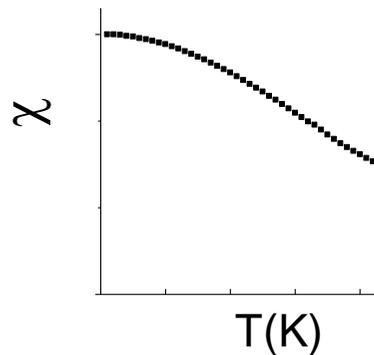
Damped oscillator spin fluctuations
(Fermionic, with $\chi'' \sim E$ at small E)



$$\chi''(E) = \frac{\chi(0)E(\Gamma/\pi)}{(E-E_0)^2 + \Gamma^2}$$

$E_0 = k_B T_{sf}$; Γ = SF damping rate

Van Vleck-like susceptibility



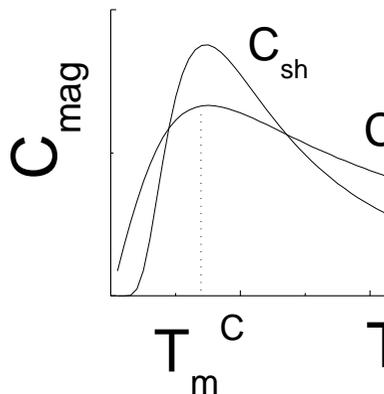
$\chi(0) \sim C_J/T_{sf}$ at low T ; $\chi(T) \sim C_J/(T+T_{sf})$ at high T

$$C_J = g^2 J(J+1) \mu_B^2 / 3k_B$$

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 ”: $\gamma T_m^C = 1/2 R \ln(2J+1)$

Together with $T_m^C \sim 1/3 T_{sf}$ (as for Schottky)

$$\Rightarrow \gamma T_{sf} = 3/2 R \ln(2J+1)$$

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]:*

$$\gamma_0 = \pi J R / 3 T_0$$

$$\chi_0 = (2J + 1) C_J / 2 \pi T_0$$

- *Cox's results (NCA) for the neutron scattering cross section:*

$$E_{max} = 1.36 T_0^{Cox} = 1.36 (T_0 / 1.15) = 1.18 T_0$$

Depends on J , $4f$ occupation n_f (degree of intermediate valence).

We assume $E_{max} = T_0$ is valid to 20%

V. T. Rajan et al., Phys. Rev. Lett., 51, 308 (1983).

D. L. Cox et al., J. Magn. Magn. Mater., 54, 333 (1986).

Low T scaling laws:

1) $E_{max}\gamma$

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

Kondo: $(\pi/3) R J$

J	Rough	Kondo
1/2	8.6	4.35
5/2	22.3	21.75
7/2	25.9	30.45
9/2	28.7	39.15

2) $W = (\pi^2 R / 3 C_J) \chi(0) / \gamma$

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

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

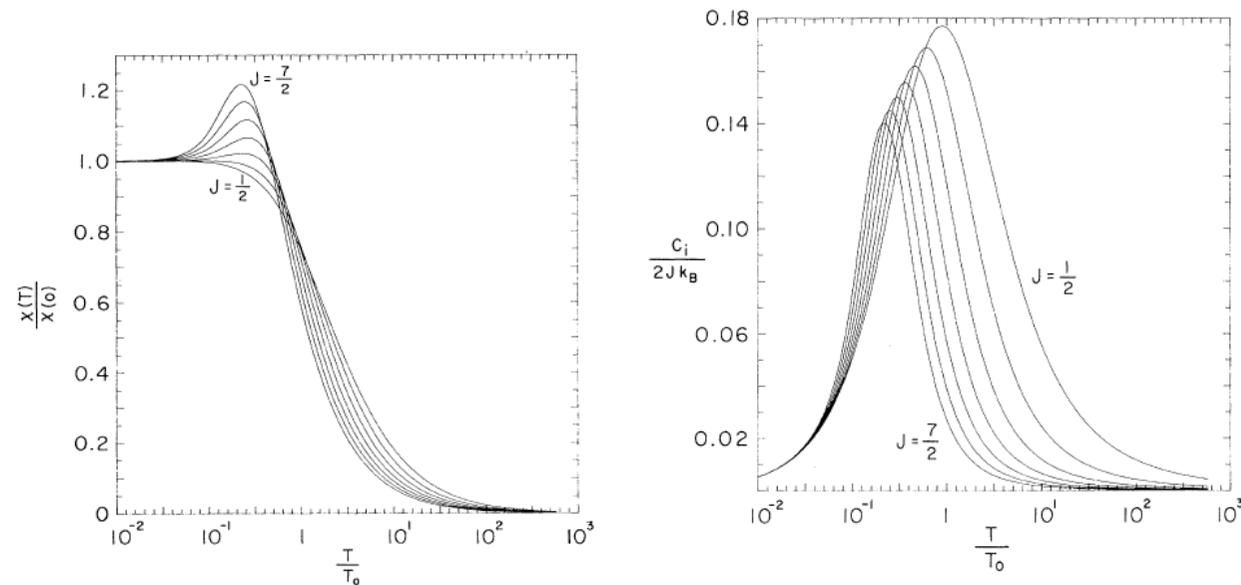
J	Rough	Kondo
1/2	3.15	2.00
5/2	1.22	1.20
7/2	1.05	1.14
9/2	0.95	1.11

Low T Scaling in Ce, Yb and U compounds

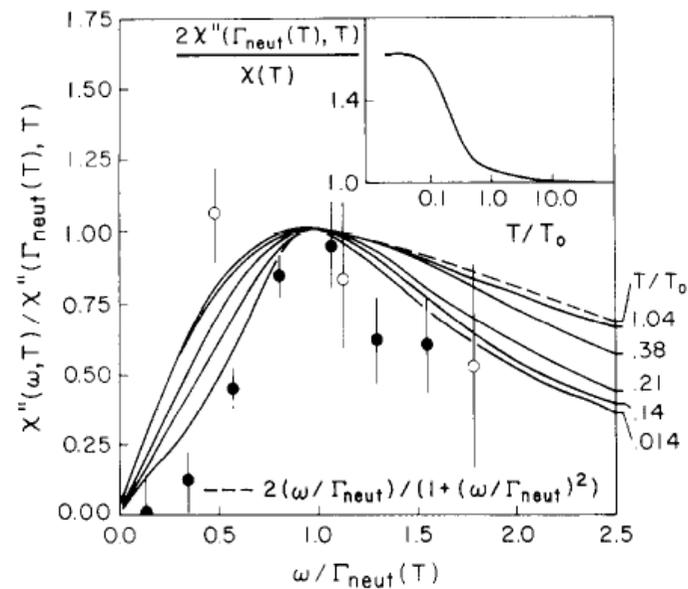
Compound	γ (J/mol-K ²)	E_0 (K)	γE_0 (J/mol-K)	$\chi(0)$ (emu/mol)	Wilson
CeRu ₂ Si ₂	0.35	23.2	8.12	0.016	
Ce ₃ In	0.7	17	11.9	0.06	
CeCu ₂ Si ₂	1.1	8.7	9.57	0.008	
CeCu ₆	1.6	3.5	5.6	0.027	
CeAl ₃	1.62	7.66	12.4092	0.036	
CePd ₃	0.03	700	21	0.002	1.52
CeSn ₃	0.042	491	20.622	0.00185	1.49
YbAl ₃	0.04	634	25.36	0.005	1.32
YbInCu ₄	0.041	507	20.787	0.006	1.54
YbMgCu ₄	0.053	545	28.885	0.004	0.8
YbAgCu ₄	0.199	139	27.661	0.017	0.9
YbFe ₂ Zn ₂₀	0.52	70	36.4	0.05	1.02
UAl ₂	0.14	243	34.02	0.004	0.48
USn ₃	0.17	60	10.2	0.01	0.99
URu ₂ Zn ₂₀	0.188	191	35.908	0.012	1.08
UPt ₃	0.45	70	31.5	0.009	0.34
UCo ₂ Zn ₂₀	0.558	70	39.06	0.047	1.42
UBe ₁₃	1.1	150	165	0.015	0.23

Note: Rough Approximation gives values close to Kondo for $E_{max}\gamma$ and W

Temperature-dependent scaling in Kondo model



Temperature dependence of χ and C_m for different J [Rajan]



Energy dependence of the dynamic susceptibility at different temperatures. Γ_{neutron} is the peak position E_{max} [Cox].

The validity of the AIM for the rare earth 4f compound $\text{YbFe}_2\text{Zn}_{20}$

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 $\chi''(E)$

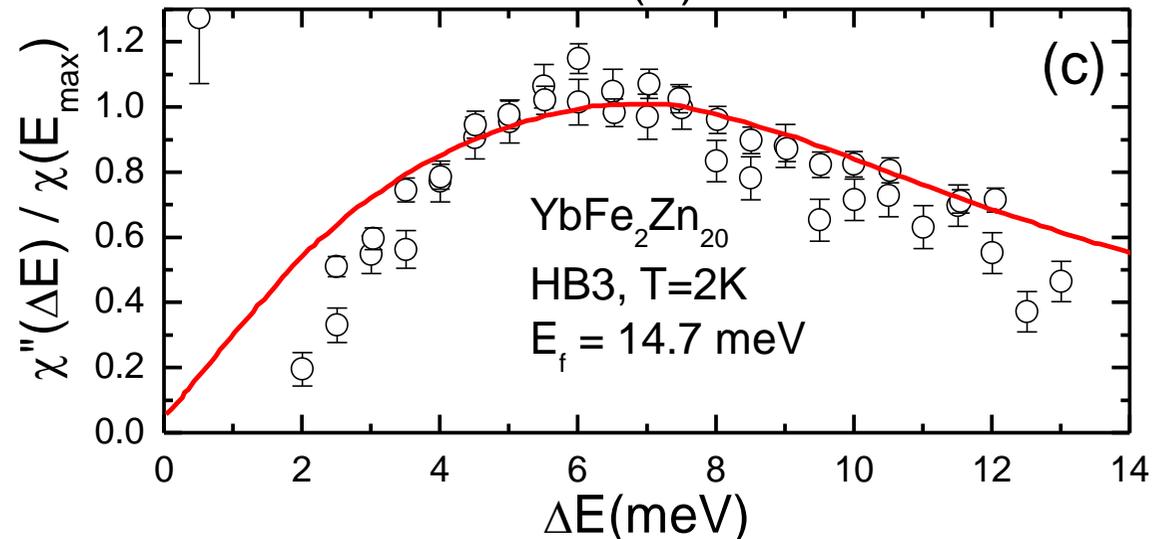
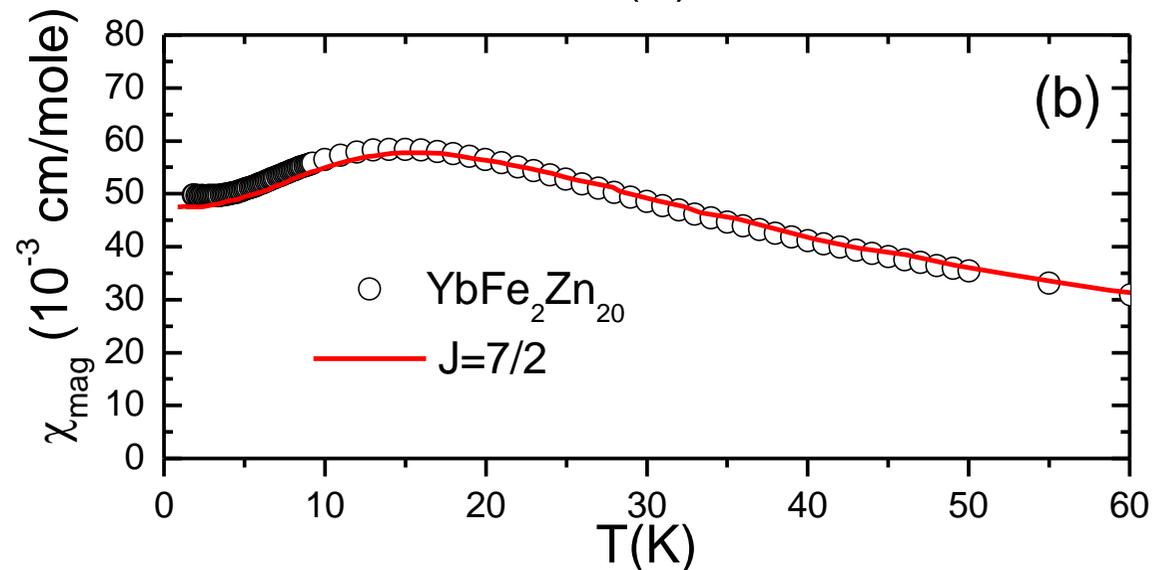
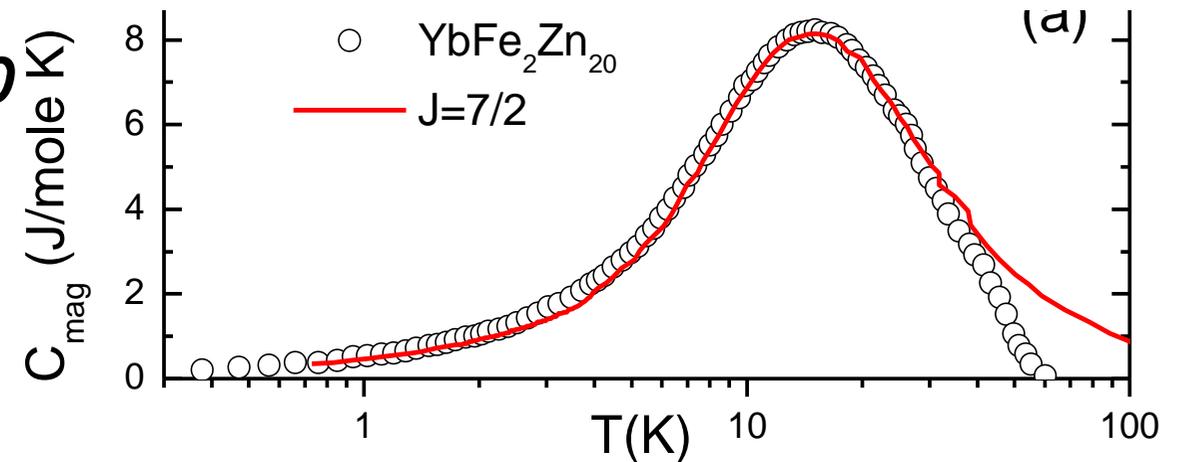
$$E_{\text{max}} = 1.18 T_0 = 82\text{K} = 7\text{ meV}.$$

Red line: $\chi''(\Delta E) / \chi''(E_{\text{max}})$ was as determined from Fig. 4 of Cox

➤ **Only one adjustable parameter (T_0)**

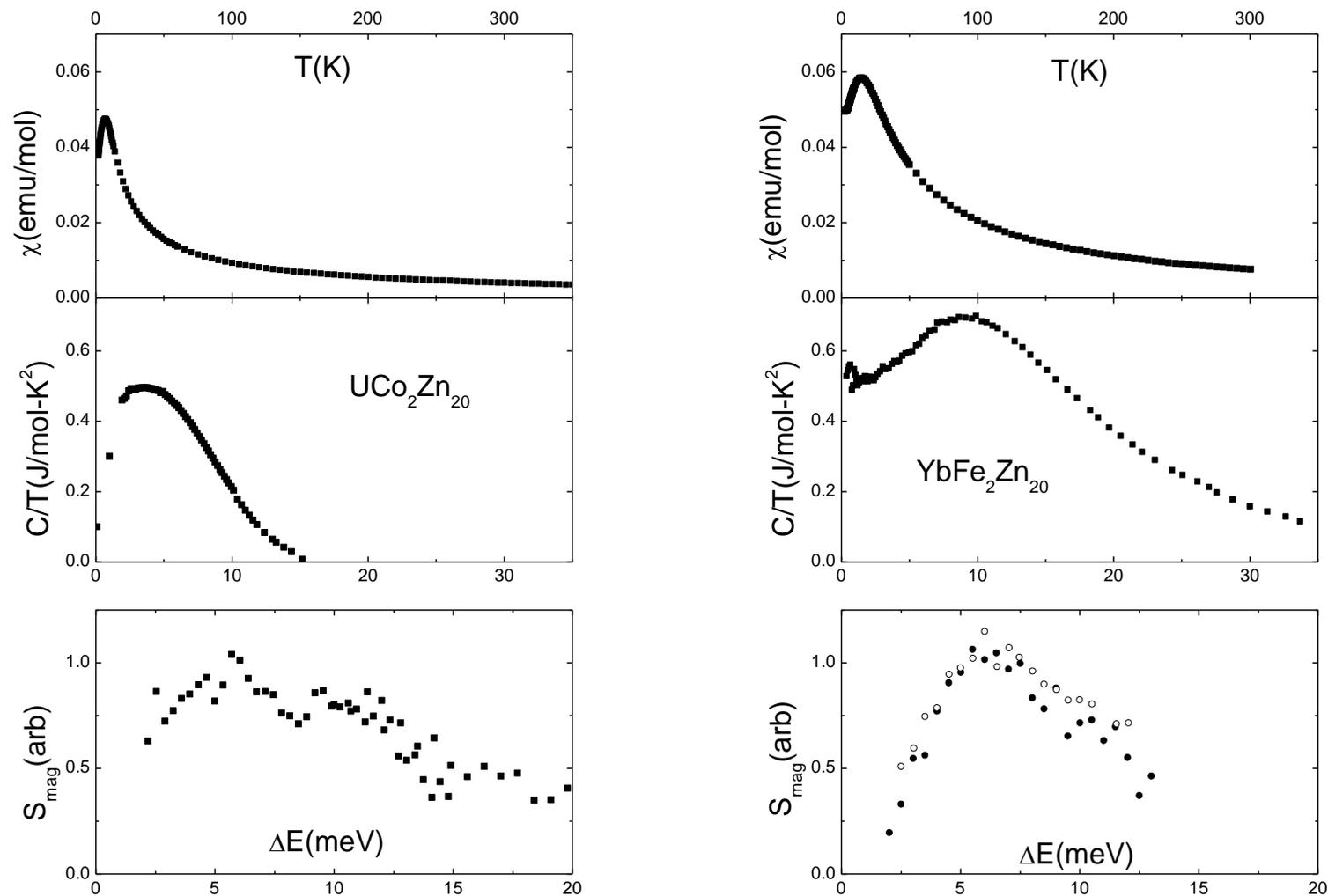
$$(T_0 = 69.2\text{ K.})$$

➤ **WOW!**



YbFe₂Zn₂₀ compared to UCo₂Zn₂₀

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



Since Kondo works so well for YbFe₂Zn₂₀, we next examine how it works for UCo₂Zn₂₀

Note: URu₂Zn₂₀ and YbFe₂Zn₂₀ 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 $\sim 6\text{\AA}$, 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 $U\text{Co}_2\text{Zn}_{20}$ and $URu_2\text{Zn}_{20}$:

Use Rajan and measured value of γ

➤ For each J, estimate T_0 (called T_K in the table) from $\gamma = \pi J R / 3 T_0$.

➤ For J= 5/2 and 9/2, use the free ion value Curie constant for C_J .

(For J = 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

	$T_K(K)$		$T_{max}^C(K)$		$\chi_0(\frac{emu}{mole})$		$T_{max}^X(K)$		$E_{max}(meV)$	
	Ru	Co	Ru	Co	Ru	Co	Ru	Co	Ru	Co
experiment			6.8	7.1	0.0123	0.037		7	16.5	5.8
J=9/2	208	69	36.5	12.1	0.0125	0.0378	39.2	13.0	21.3	7.1
J=5/2	116	38	34	11	0.0135	0.0412	30	10	11.9	3.9
J=1/2	23	7.6	20	6.8	0.0245	0.0402			2.4	0.8

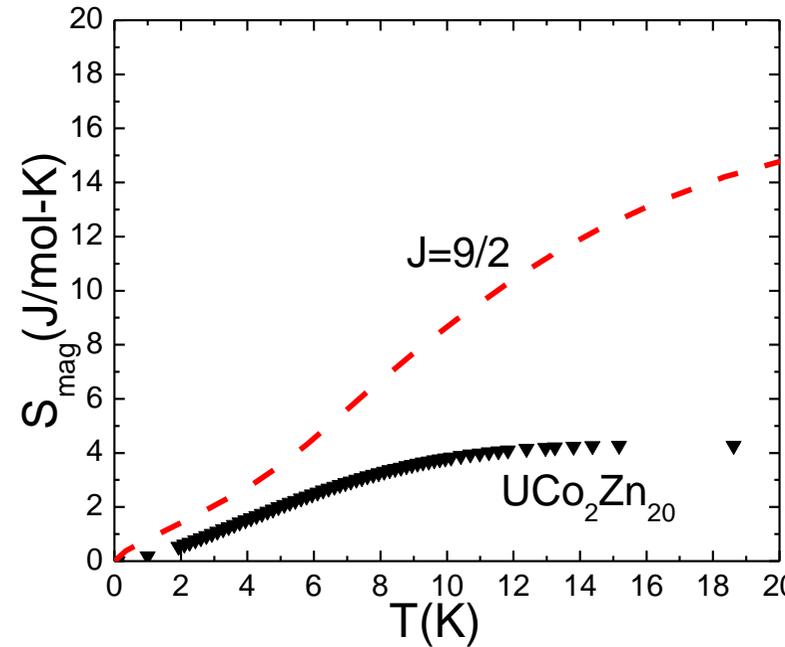
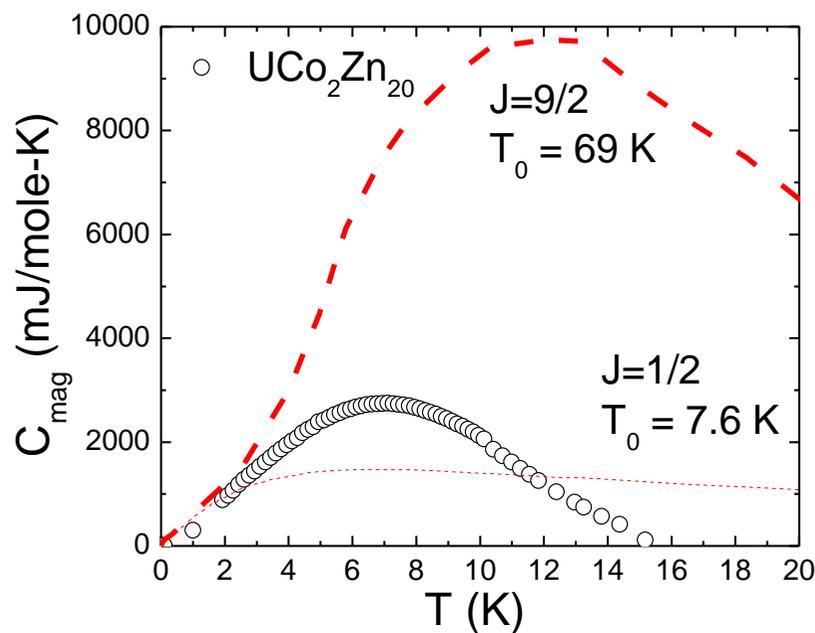
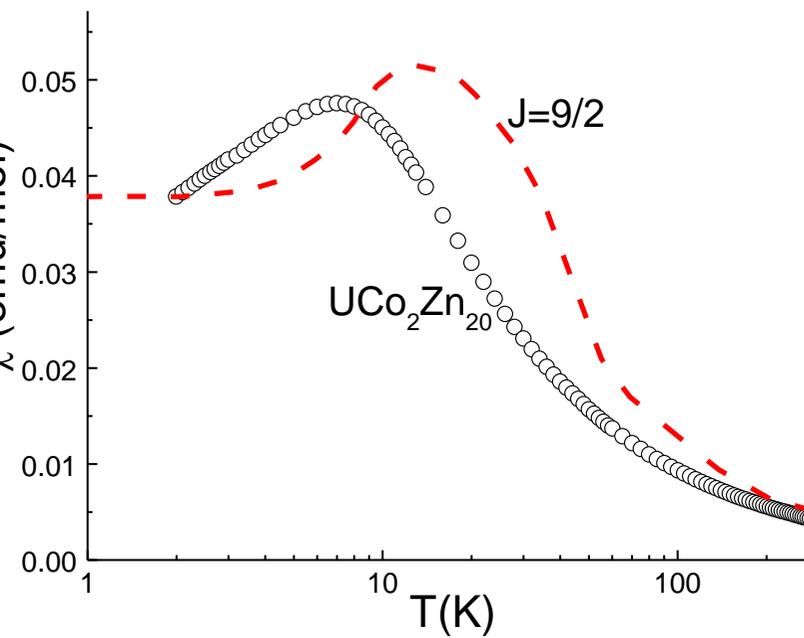
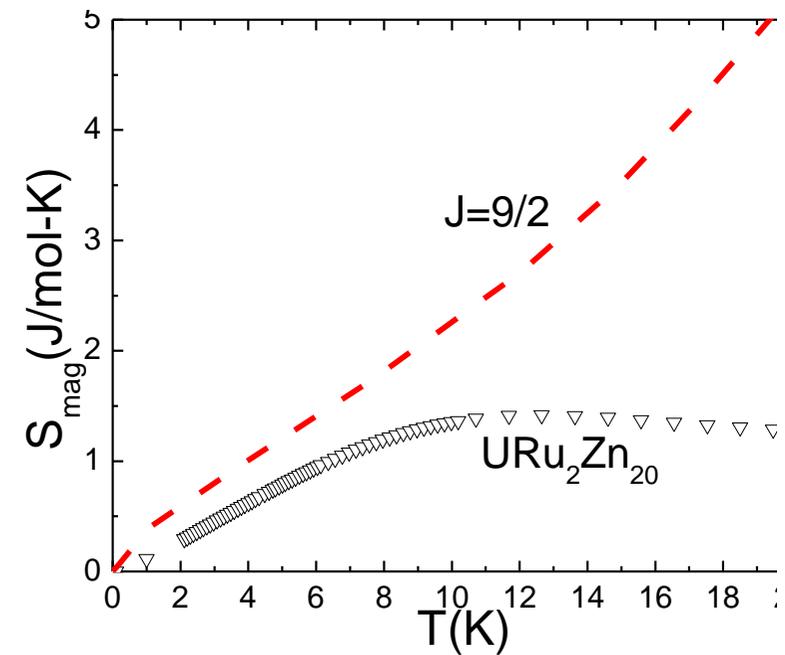
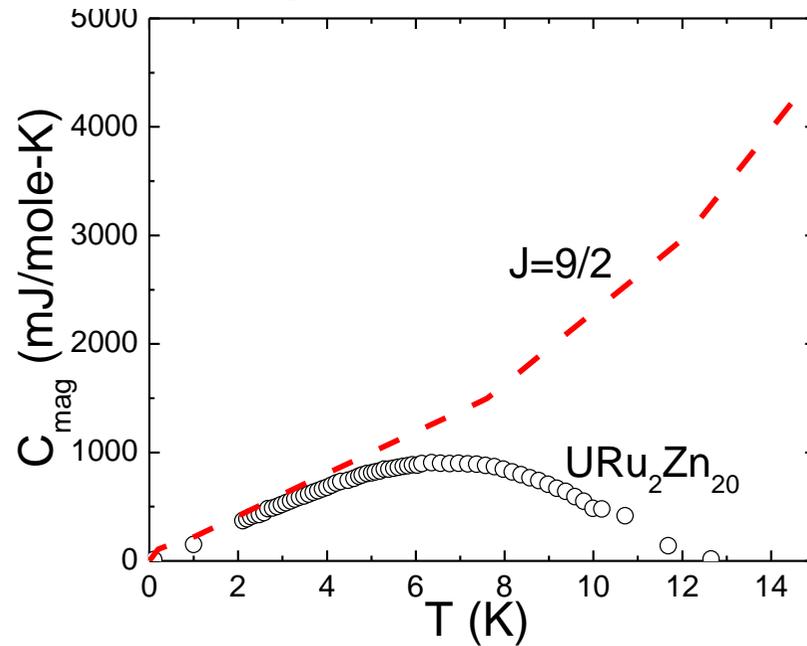
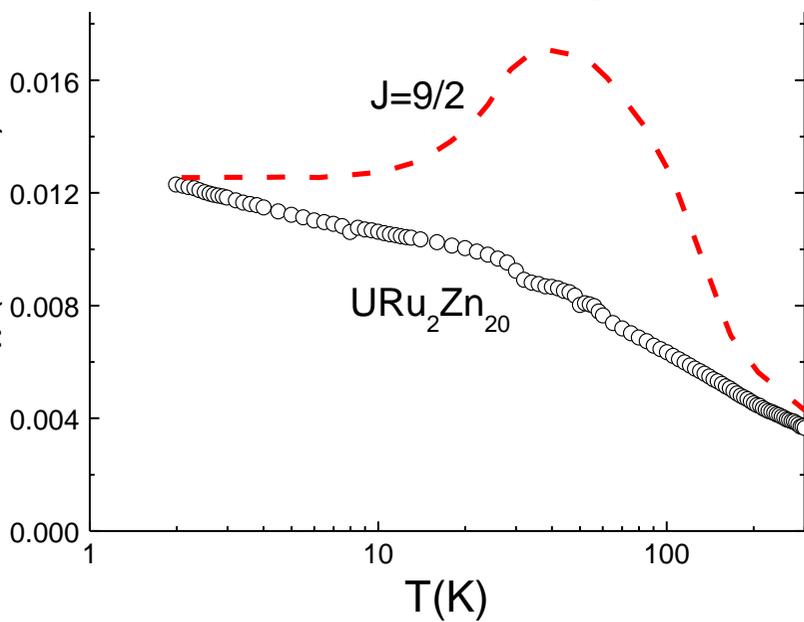
➤ **J=9/2** values for χ_0 and E_{max} are close to experiment: **K/AIM scaling good at low T?**

Fix J = 9/2, fix T_0 at the values given above.

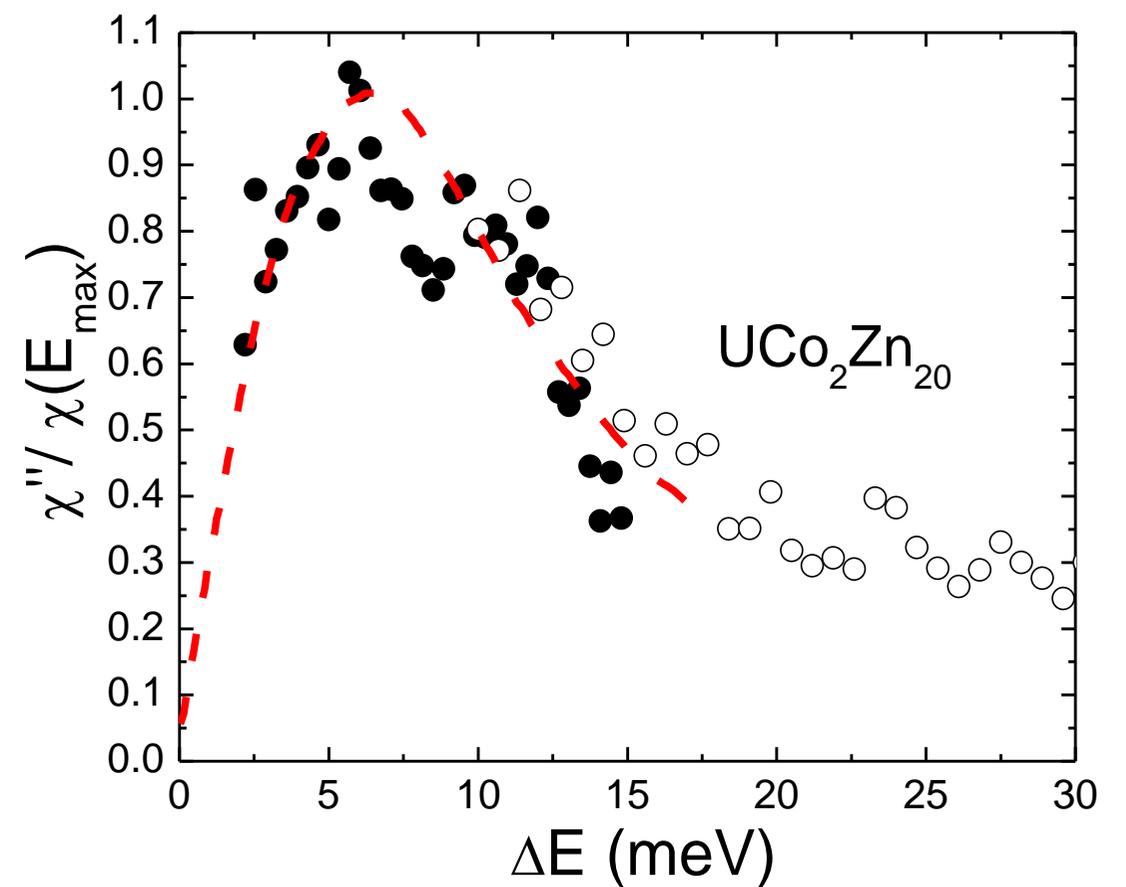
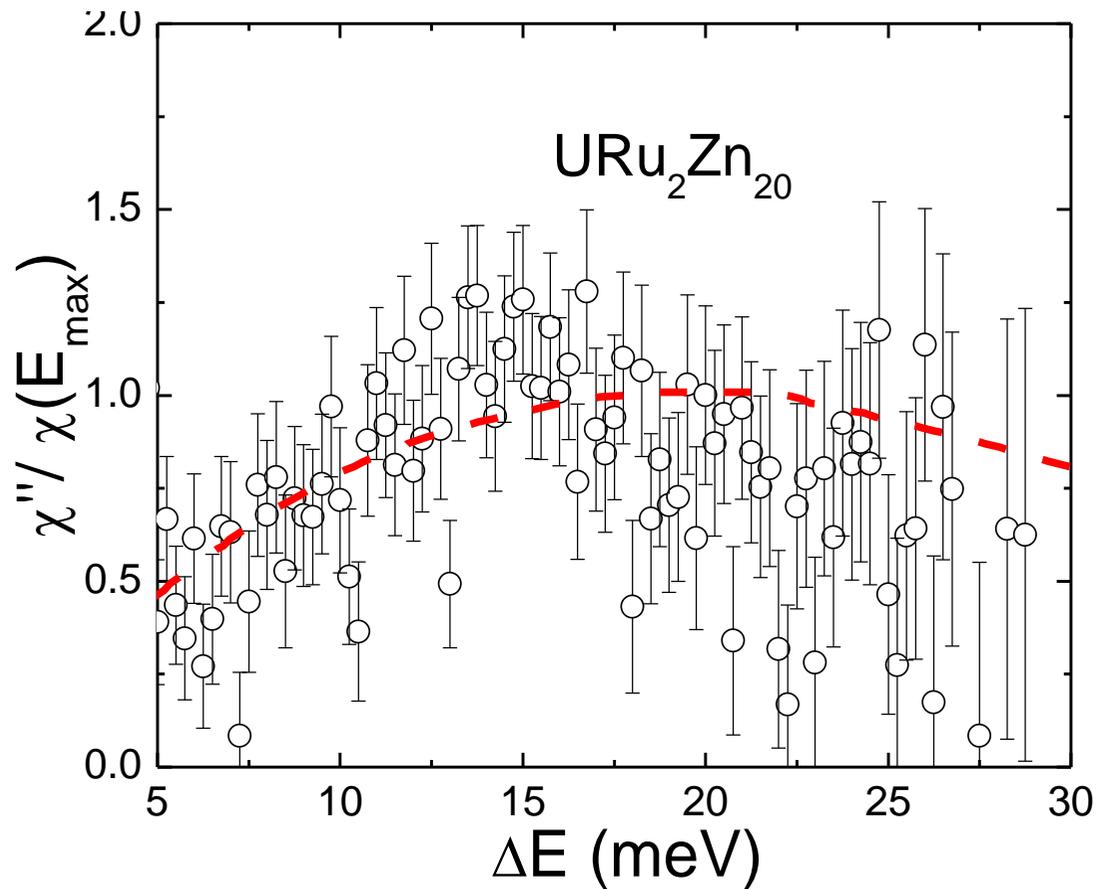
Calculate the full T- dependence for χ and γ using Rajan's curves

(use Cox for the E-dependence of $\chi''(E)/\chi''(E_{max})$)

AIM predictions for the temperature dependence of $\chi(T)$ $C_{\text{mag}}(T)$ and $S_{\text{mag}}(T)$ in the $J = 9/2$ case:



AIM predictions for the energy dependence of $\chi''(\Delta E) / \chi''(E_{\max})$:



Summary so far:

- - Kondo scaling, at both low and intermediate temperature, works extremely well for certain rare earth (RE) HF compounds. (YbAgCu₄; YbFe₂Zn₂₀)

-- For UAl₂, USn₃, UPt₃ and UBe₁₃ either $W = (\pi^2 R/3C_J) \chi(0)/\gamma$ or $E_{\max} \gamma$ 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 γ varies by an order of magnitude (from 0.1 to 1 J/mol-K²).

-- $J = 9/2$ Low-T Kondo scaling works well for UCo₂Zn₂₀ and URu₂Zn₂₀, but the temperature dependence predicted by Kondo theory for $J = 9/2$, has serious problems:

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

Experimental entropy is *much* smaller than predicted.

(For UCo₂Zn₂₀ 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_{\max} . Since this is the fundamental scale $k_B T_{sf}$, this is a serious failure.)

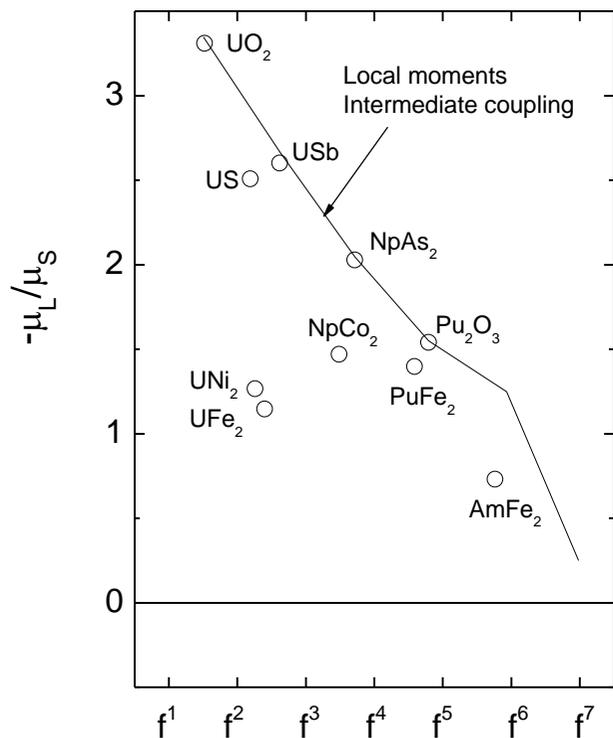
What's wrong with U-HF low-T scaling?

1) High temperature paramagnetic moment/ entropy

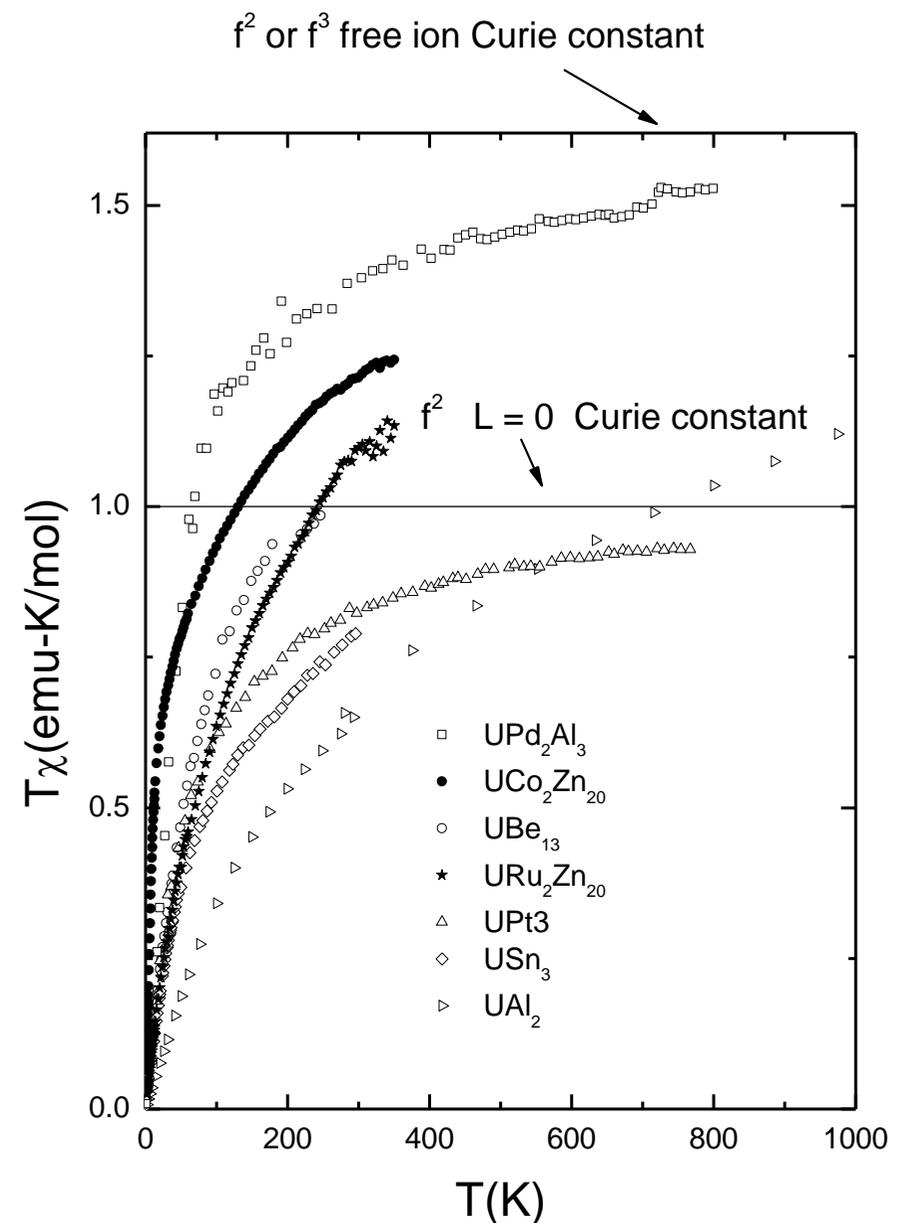
In cerium and Yb HF compounds, the high temperature “effective moment” $T\chi$ tends to the (Hund’s Rule) free ion (local moment) value $C_J = g^2 J(J+1) \mu_B^2 / 3k_B$ and the entropy tends to $R \ln N_J$ where $N_J = 2J+1$ is the degeneracy of the moment.

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



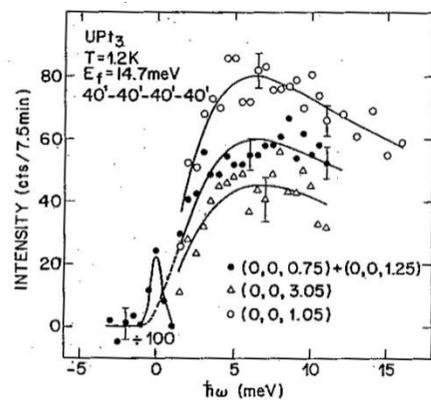
The Curie constant and the entropy enter the scaling laws (in the rough approximation) as $E_{max} \gamma \sim (3/2)R \ln N_J$ and $W = (\pi^2 R / 3 C_J) \chi(0) / \gamma$. Hence we might improve agreement with scaling by using better values for C_J and N_J .

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 $E_{\max} \sim 10 - 20$ 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.



Unpolarized, constant-Q spectra for single crystal sample of UPt₃ (from ref. [7]).

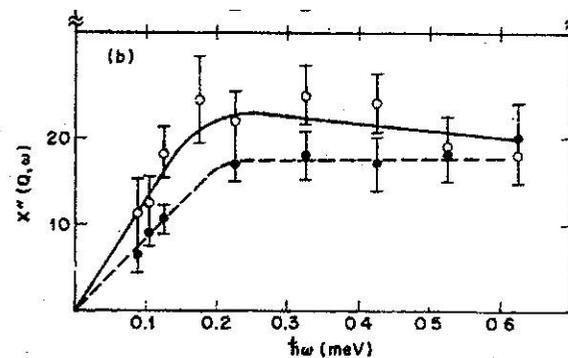


FIG. 4. (a) Constant- Q scans at $Q=(0.3,0,1)$ at $T=0.5$ K and $Q=(0.53,0,1)$ at $T=5$ K and $T=0.5$ K. (b) $\chi''(Q,\omega)$ at the same two temperatures deduced from these data as described in the text.

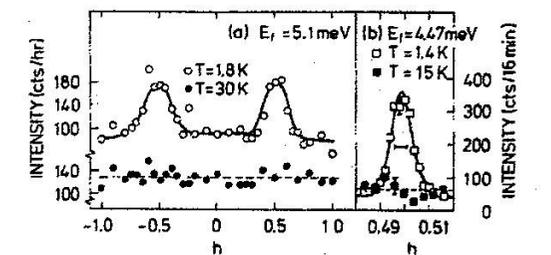


FIG. 5. Constant-energy scans along $(h,0,1)$ above and below T_N . (a) Taken at an energy transfer of 0.5 meV. Note that while the scattering at 30 K no longer peaks at $(\pm \frac{1}{2}, 0, 1)$, its level is higher than that seen between the maxima for $T=1.8$ K. (b) Elastic scattering with a PG and Be filtered beam. The line through these points is a resolution-corrected Lorentzian. The horizontal bar indicates the spectrometer resolution as measured by $\lambda/2$ scattering from the nuclear (102) Bragg reflection.

Example: UPt₃

Main spin fluctuation at $E_{\max} \sim 6$ 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 $(1/2, 0, 1)$

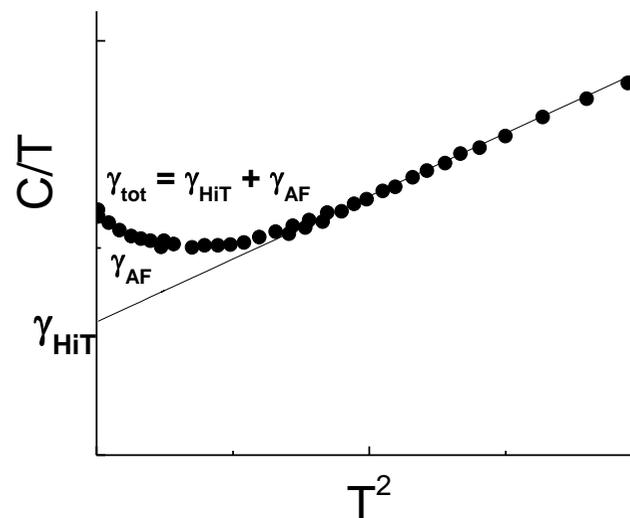
Peak in intensity near $T_N = 5$ K, disappears above 20K

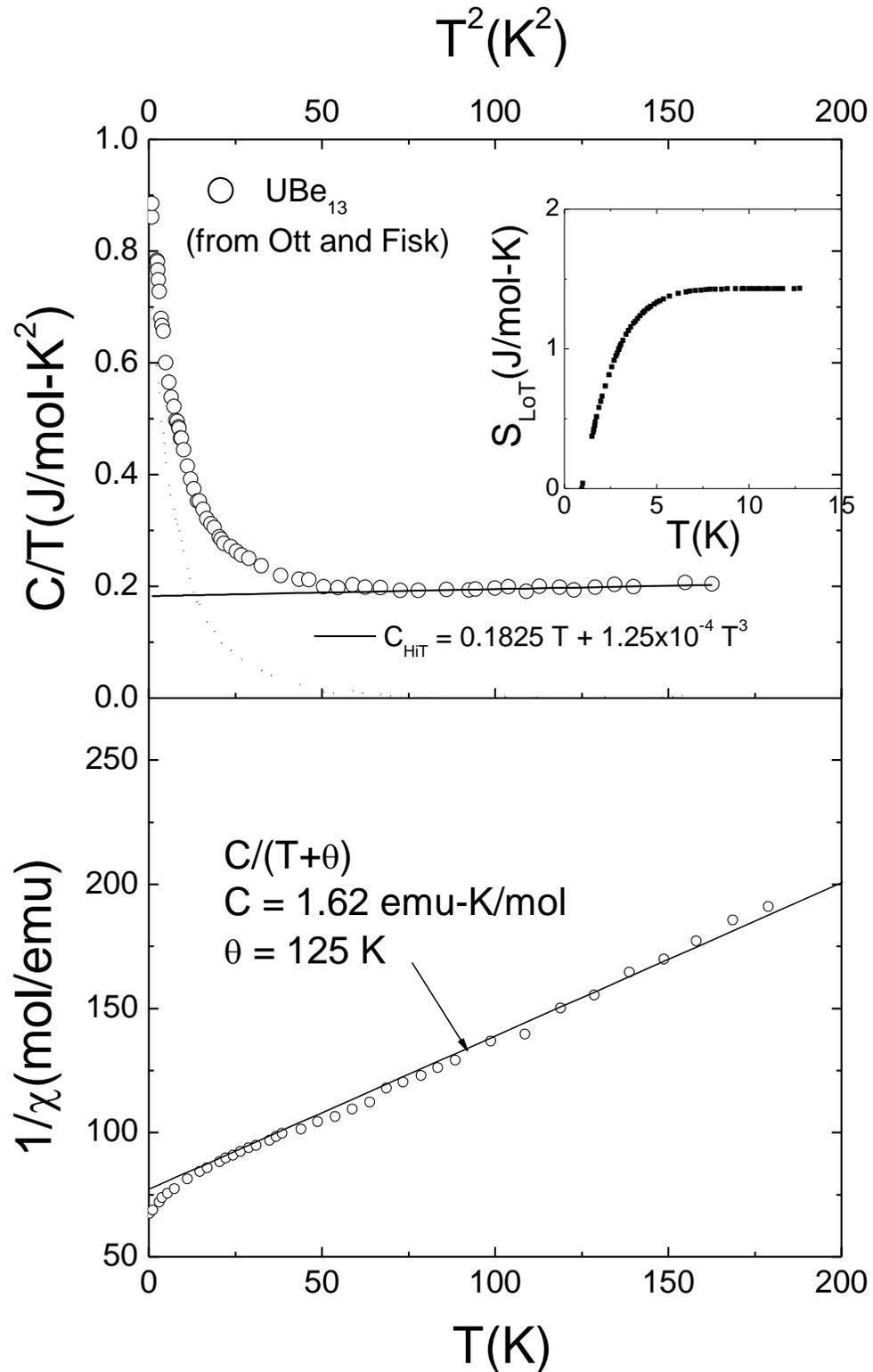
What's wrong with U-HF low-T scaling? (continued)

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 $\gamma = (\pi/3) R J/ E_{\max}$. Assuming that the susceptibility is not strongly affected by AF fluctuations, this means that the Wilson ratio of χ/ γ will be much smaller, and the product γE_{\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 γ_{HiT} corresponds to the value arising from the spin fluctuation at $E_{\max} = k_B T_{\text{sf}}$.





Example: UBe_{13}

$$\chi(0) = 0.015 \text{ emu/mol}$$

$$\gamma = 1.1 \text{ J/mol-K}^2$$

$$E_{\text{max}} = 13 \text{ meV} \equiv 150 \text{ K}$$

\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:

- These values are much closer to the expected values in the rough approximation for the $J = 9/2$ case where $W = 1.11$ and $E_{\text{max}} \gamma = 28.7$.
- Use of the $J = 9/2$ Curie constant appears reasonable at high T .

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

- The AF fluctuations account for most of γ at low- T , affecting γ only below 8 K and generating only 25% of $R \ln 2$.

Neutrons see $\Gamma_{\text{AF}} \sim 1\text{-}2 \text{ meV}$ for $T < 20 \text{ K}$

Example: UAl_2

$$\chi(0) = 0.004 \text{ emu/mol}$$

$$\gamma = 0.14 \text{ J/mol-K}^2$$

$$E_{\text{max}}/k_B = 243\text{K}$$

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

$$W = (\pi^2 R/3C_J) \chi(0)/\gamma = 0.48$$

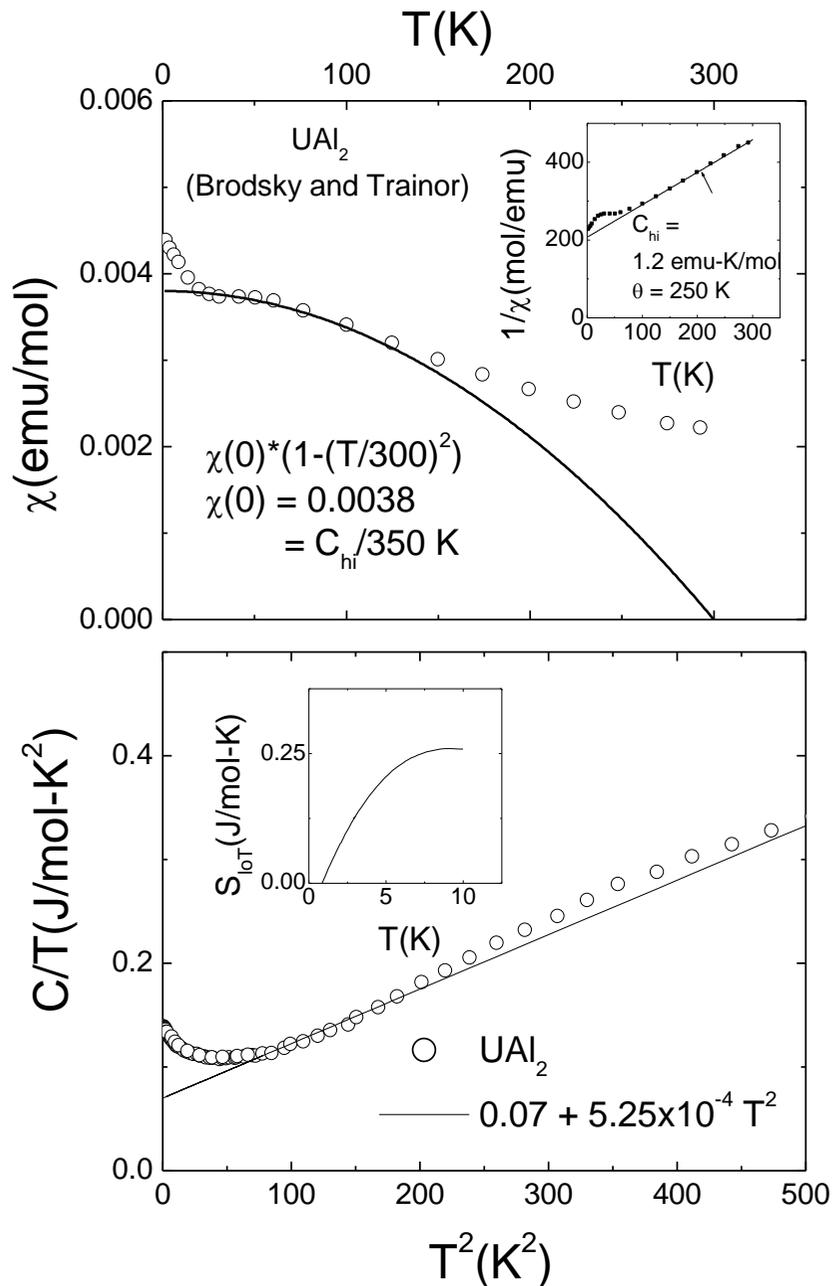
$$E_{\text{max}} \gamma = 34 \text{ J/mol-K}$$

$$\text{But for } \gamma_{\text{HiT}} = 0.07 \text{ J/mol-K}^2$$

$$\text{and } C_{\text{HiT}} = 1.2 \text{ emu-K/mol}$$

$$\Rightarrow W = (\pi^2 R/3C_J) \chi(0)/\gamma_{\text{HiT}} = 1.30$$

$$E_{\text{max}} \gamma_{\text{HiT}} = 17.0 \text{ J/mol-K}$$



Notes:

- 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.
- 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 be a bit smaller than 1.5 (the Kondo $J=3$ value)
- The Curie Weiss $\theta = 250$ 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_{max}/k_B
- The AF fluctuations have not been seen by neutrons. We assume they arise below 10K and account for half of the low- T γ ; they generate entropy of order only 5% of $R \ln 2$.

Example: UPt₃

$$\chi(0) = 0.009 \text{ emu/mol}$$

$$\gamma = 0.45 \text{ J/mol-K}^2$$

$$E_{\text{max}}/k_B = 70 \text{ K}$$

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

$$W = (\pi^2 R/3C_J) \chi(0)/\gamma = 0.34$$

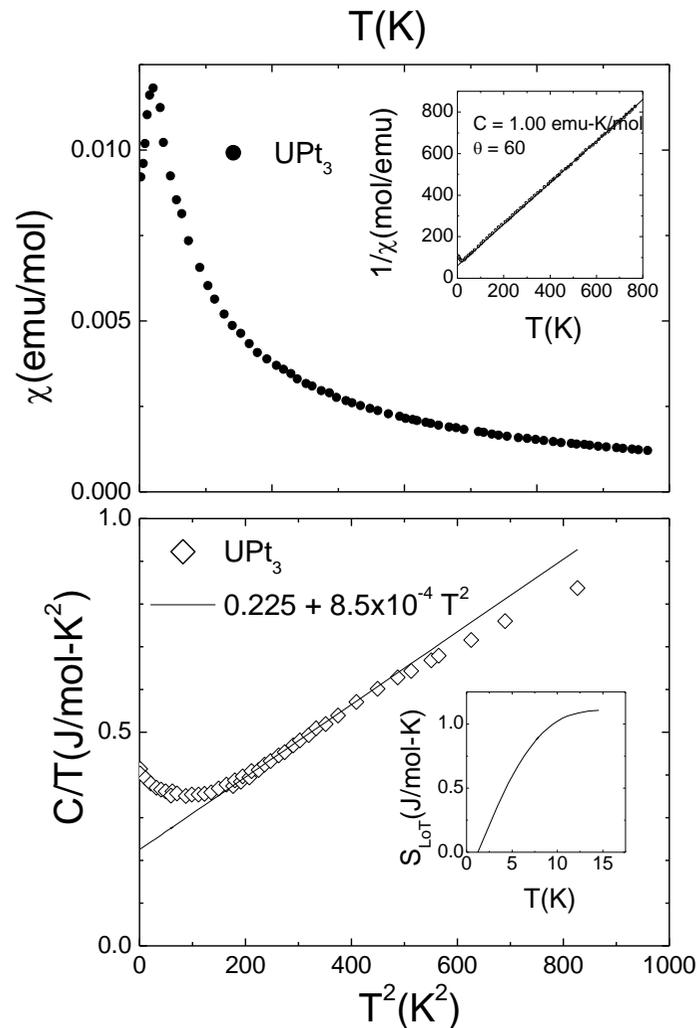
$$E_{\text{max}} \gamma = 31.5 \text{ J/mol-K}$$

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

$$\text{and } C_{\text{HiT}} = 1 \text{ emu-K/mol}$$

$$W = (\pi^2 R/3C_J) \chi(0)/\gamma_{\text{HiT}} = 1.10$$

$$E_{\text{max}} \gamma_{\text{HiT}} = 15.7 \text{ J/mol-K}$$



Notes:

- The Curie constant is equal to the $L=0$ $S=1$ $g=2$ value of 1.00 emu-K/mol.
- 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 be a bit smaller than 1.5 (the Kondo $J=3$ value).
- The Curie Weiss $\theta = 60$ K, and the low-T value of $C_{\text{HiT}}/\chi(0) = 111$ K correspond to E_{max}/k_B
- As shown above, the AF fluctuations arise below 20K; they account for half of the low-T γ ; they generate entropy of order 20% of $R \ln 2$. They do *not* peak at $T_N = 5$ K.

Antiferromagnetic uranium compounds:

Compounds such as U_2Zn_{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 (~ 1 meV) scale and demagnetizing spin fluctuations on a larger energy scale (~ 10 meV). In both cases, there is a large linear coefficient of specific heat as $T \rightarrow 0$ 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.

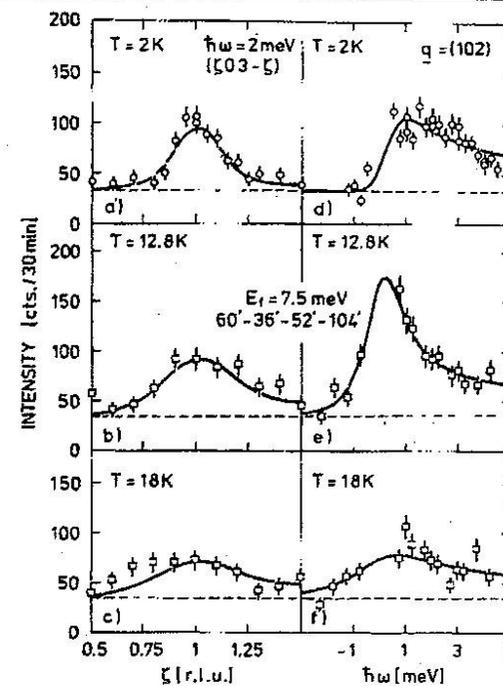
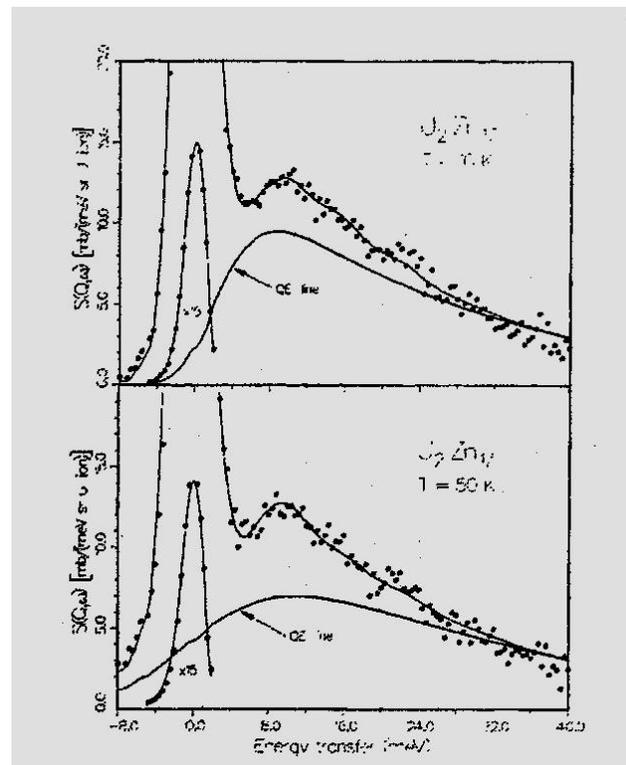


FIG. 2. Constant $\hbar\omega = 2$ meV and constant $Q = (1,0,2)$ scans at temperatures above and below T_N . The solid lines result from the model calculations described in text.

Example: U_2Zn_{17} : An HF antiferromagnet

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

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

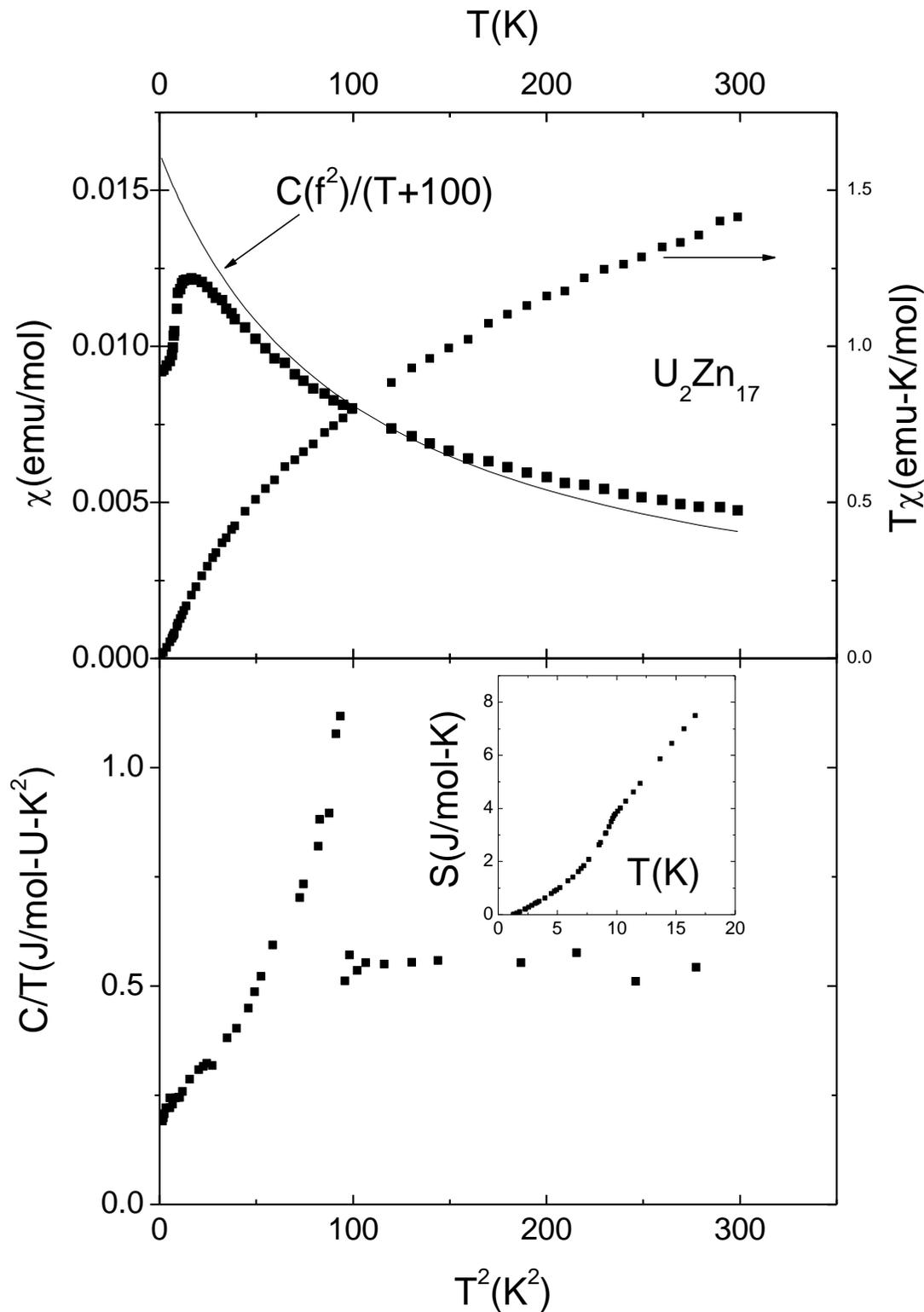
$$\gamma_{\text{Lo}} = 0.20 \text{ J/mol-K}^2$$

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

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

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

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



Notes:

- The high-T behavior of $T\chi$ suggests free ion f^2 (or f^3) behavior. A Curie Weiss law with $\theta = 100$ K and $C(f^2)$ is close to the data and corresponds to $E_{\max}/k_B = 100$ K.
- The AF fluctuations ($\Gamma_{\text{AF}} \sim 1$ meV) appear above $T_N = 9.7$ K; the AF ordering wipes out more than half of the low-T γ . The entropy at T_N is close to $R \ln 2$.
- If we use γ_{Hi} in the scaling laws, then $W = 2.88$ and $E_{\max} \gamma_{\text{Lo}} = 55$ J/mol-K; both values are much too large. If we use γ_{Lo} , then the Wilson ratio is good but $E_{\max} \gamma_{\text{Lo}}$ 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 ratio 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 γ_{HiT} extrapolated for $10 < T < 20$ K. We assume that the upturn in C/T below 10K 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_{HiT} . Since, for UPt_3 and UAl_2 , C_{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_2Zn_{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 UPt₃. Direct comparison with appropriate rare earths would be very compelling: e.g. UCo₂Zn₂₀ vs YbFe₂Zn₂₀ or USn₃ vs CeSn₃.

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 $T > 20\text{K}$, 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) UCo₂Zn₂₀ 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 $T < 20\text{K}$, 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_{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_{HiT} , μ_L/μ_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_2Zn_{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:

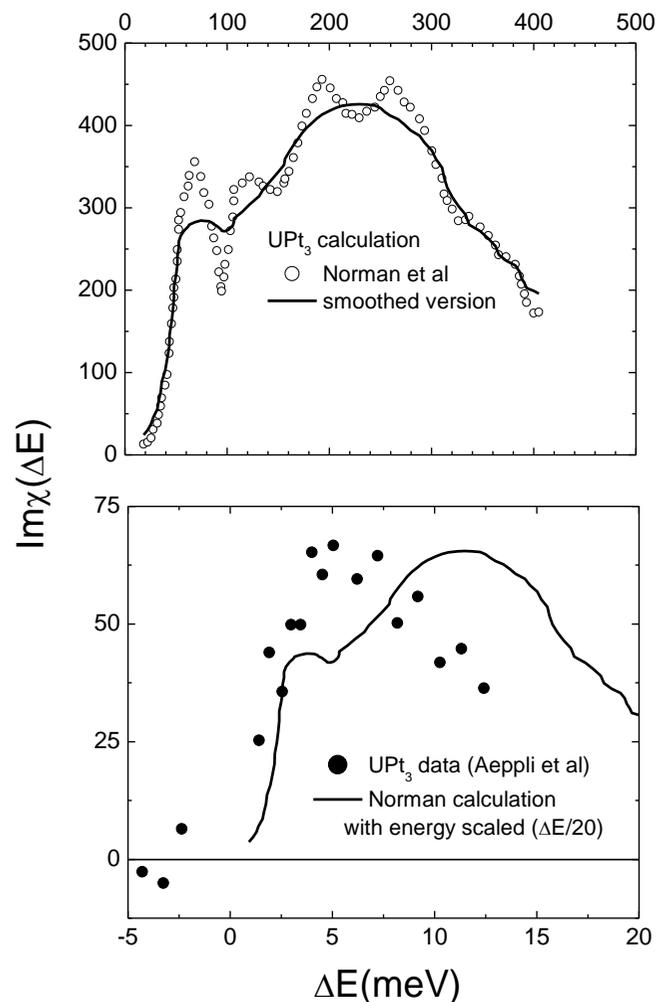
The spin fluctuation peaks seen in $\chi''(E)$ in the range 5-20 meV in uranium HF compounds are very similar in lineshape and energy scale to what is seen in intermediate valence cerium and ytterbium compounds. AF fluctuations are only seen in cerium HF compounds that have very large specific heat coefficients 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 itinerancy of the $5f$'s, the intersite f - f coupling energy J is larger relative to $k_B T_{\text{sf}}$ 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 $E_{\max}\gamma$.

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.

Theory Comments, continued

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

$$\chi_L(\mathbf{E}) = \chi_L / (1 - i\mathbf{E}/\Gamma_L)$$

which then is subjected to site-site exchange coupling J_Q

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

The assumption is made that $\chi(\mathbf{Q}, 0)$ will diverge at the QCP; a finite value of $\chi(\mathbf{Q}, 0)$ 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 γ and E_{max} . Hence, the low value of γE_{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.

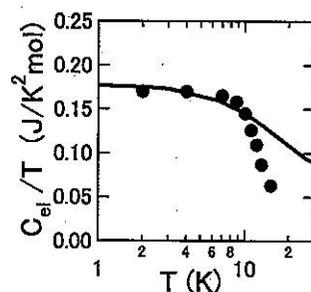


FIG. 7. (Color online) Closed circles are the T dependence of the electronic specific heat C_{el}/T from Ref. 4, which has been estimated by subtraction of the phonon part from the total specific heat. The solid line is C_{el}/T calculated using the spin fluctuation parameters in Table II and the equation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

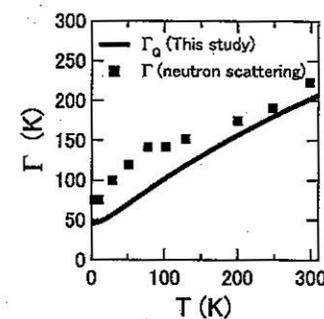


FIG. 9. (Color online) T dependence of the relaxation rate $\Gamma_Q(T) \sim 2\pi T \gamma(T)$ estimated from $\gamma(T)$ in Fig. 8. For comparison, the relaxation rate $\Gamma(T)$ determined in neutron inelastic scattering measurements (Ref. 17) is presented.

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:

--We showed above that the K/AIM worked very well for rare earth compounds such as YbAgCu_4 and $\text{YbFe}_2\text{Zn}_{20}$, but was poor for the temperature dependence of $\text{UCo}_2\text{Zn}_{20}$ and $\text{URu}_2\text{Zn}_{20}$.

--We remind the listener that in the early days of Kondo physics, strong efforts were made to have Kondo impurities in such compounds as $\text{Au}_{1-x}\text{Fe}_x$ on the ppm level. The argument was made – and experimental evidence given – that higher levels of impurities led to d-d coupling \mathbf{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 $\text{UCo}_2\text{Zn}_{20}$ and $\text{URu}_2\text{Zn}_{20}$.