Neutron scattering and scaling behavior in URu$_2$Zn$_{20}$ and YbFe$_2$Zn$_{20}$

C. H. Wang$^{1,2}$, A. D. Christianson$^3$, J. M. Lawrence$^1$, E. D. Bauer$^2$, E. A. Goremychkin$^4$, A. I. Kolesnikov$^3$, F. Trouw$^2$, F. Ronning$^2$, J. D. Thompson$^2$, M.D. Lumsden$^3$, N. Ni$^5$, E. D. Mun$^5$, S. Jia$^5$, P. C. Canfield$^5$

$^1$University of California, Irvine, California 92697, USA
$^2$Los Alamos National Laboratory, Los Alamos, NM 87545, USA
$^3$Oak Ridge National Laboratory, Oak Ridge, TN, 37831, USA
$^4$Argonne National Laboratory, Argonne, IL 60439, USA
$^5$Ames Laboratory, Iowa State University, Ames, IA, 50011

(Dated: June 8, 2009)

The dynamic susceptibility $\chi''(E)$, measured by inelastic neutron scattering measurements, shows a broad peak centered at $E_{\text{max}}=16.5$ meV for the cubic actinide compound URu$_2$Zn$_{20}$ and 7 meV at the (1/2, 1/2, 1/2) zone boundary for the rare earth counterpart compound YbFe$_2$Zn$_{20}$. For URu$_2$Zn$_{20}$, the low temperature susceptibility and magnetic specific heat coefficient $\gamma = C_{\text{mag}}/T$ take the values $\chi = 0.0123 \text{ emu/mole}$ and $\gamma = 190 \text{ mJ/mole-K}^2$ at $T=2 \text{ K}$. These values are roughly three times smaller, and $E_{\text{max}}$ is three times larger, than recently reported for the related compound UCo$_2$Zn$_{20}$, so that $\chi$ and $\gamma$ scale inversely with the characteristic energy for spin fluctuations, $T_{sf} = E_{\text{max}}/k_B$. While $\chi(T)$, $C_{\text{mag}}(T)$, and $E_{\text{max}}$ of the 4f compound YbFe$_2$Zn$_{20}$ are very well described by the Kondo impurity model, we show that the model works poorly for the URu$_2$Zn$_{20}$ and UCo$_2$Zn$_{20}$, suggesting that the scaling behavior of the actinide compounds arises from spin fluctuations of itinerant 5f electrons.

An important property of heavy fermion (HF) materials is a scaling law whereby the low temperature susceptibility $\chi$ and specific heat coefficient $\gamma = C/T$ vary as $1/T_{sf}$. Here $k_B T_{sf}$ is the spin fluctuation energy scale which can be directly observed as the maximum $E_{\text{max}}$ in the dynamic susceptibility $\chi''(E)$, measured through inelastic neutron scattering. Such scaling receives theoretical justification[1–4] from the Anderson impurity model (AIM), where the spin fluctuation temperature $T_{sf}$ is identified as the Kondo temperature $T_K$. This model assumes that fluctuations in local moments dominate the low temperature ground state properties of HF materials. For 4f electron rare earth HF compounds, the AIM appears to give an excellent description of much of the experimental behavior, including the temperature dependence of the magnetic contribution to the specific heat $C_m$, the susceptibility $\chi$, and the 4f occupation number $n_f$, as well as the energy dependence of the neutron scattering spectra $\chi''(E)$ of polycrystalline samples[5]. The theoretical calculations[1–4] show that these properties are highly dependent on the orbital degeneracy $N_f (= 2J + 1$ for rare earths). In particular, for large degeneracy ($N_f > 2$) both the calculated $\gamma(T)$ and $\chi(T)$ exhibit maxima at a temperature $\alpha T_K$ where $\alpha$ is a constant that depends on $N_f$. This kind of behavior is observed in rare earth compounds such as YbAgCu$_4$[5], CeSn$_3$ and YbCuAl$_6$[6] and YbFe$_2$Zn$_{20}$[7].

It is reasonable to apply the AIM, which assumes local moments, to rare earth compounds where the 4f orbitals are highly localized and hybridize only weakly with the conduction electrons. On the other hand, in uranium compounds, the 5f orbitals are spatially extended and form dispersive bands through strong hybridization with the neighboring $s$, $p$, and $d$ orbitals. Hence, we might expect differences in the details of the behavior between the uranium and the rare-earth based heavy fermion materials, despite the common occurrence of scaling. Nevertheless, we have recently shown[8] that the actinide compound UCo$_2$Zn$_{20}$ exhibits a maximum in the susceptibility and a specific heat coefficient that are strikingly similar to those seen in the rare earth compound YbFe$_2$Zn$_{20}$. It is thus of interest to test whether a local moment AIM/Kondo description, which has been shown to give excellent agreement with the data for the Yb compound (see Ref. 7 and Fig. 3 of this paper), may also be valid for 5f HF compounds.

To accomplish this, we present herein the results of inelastic neutron scattering (INS) experiments on polycrystalline URu$_2$Zn$_{20}$ together with results for the magnetic susceptibility and specific heat of single crystalline samples. We also present the INS data on single crystal YbFe$_2$Zn$_{20}$. Both compounds belong to a new family of intermetallic compounds RX$_2$Zn$_{20}$ (R = lanthanide, Th, U; X = transition metal)[7, 9–12] which crystallize in the cubic CeCr$_2$Al$_2$ type structure (Fd$\bar{3}$m space group)[10, 13]. In this structure, every $f$-atom is surrounded by 16 zinc atoms in a nearly spherical array of cubic site symmetry, which leads to small crystal field splittings. Because the $f$-atom content is less than 5% of the total number of atoms, and the shortest $f/f$ spacing is $\sim 6 \text{ Å}$, these compounds are possible candidates for studying the Anderson impurity model in periodic $f$ electron compounds.

The crystals were grown in zinc flux[7, 8]. The magnetic susceptibility measurements were performed in a commercial superconducting quantum interference de-
vice (SQUID) magnetometer. The specific heat was measured in a Quantum Design PPMS system. For URu$_2$Zn$_{20}$, we performed inelastic neutron scattering on a 40 gram powder sample on the low resolution medium energy chopper spectrometer (LRMECS) at IPNS, Argonne National Laboratory, and on the High-Resolution Chopper Spectrometer (Pharos) at the Lujan center, LANSCE, at Los Alamos National Laboratory. For YbFe$_2$Zn$_{20}$ the INS spectrum was obtained for two co-aligned crystals of total mass 8.5 grams, using the HB-3 triple-axis spectrometer at the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (ORNL); the final energy was fixed at $E_f = 14.7$ meV, and the scattering plane was $(H, H, L)$. The data have been corrected for scattering from the empty holder but have not been normalized for absolute cross section. For URu$_2$Zn$_{20}$, we used the non-magnetic counterpart compound ThCo$_2$Zn$_{20}$ to determine the scaling of the nonmagnetic scattering between low $Q$ and high $Q$; for YbFe$_2$Zn$_{20}$, we measured at $Q = (1.5,1.5,1.5)$ and $(4.5,4.5,4.5)$ and assumed that the phonon scattering scales as $Q^2$. Assuming that the magnetic scattering scales with the $Q$-dependence of the $4f$ or $5f$ form factor, we iterated several times between the low $Q$ and the high $Q$ data, subtracting the nonmagnetic component to obtain the magnetic scattering function $S_{mag}(\Delta E)$.

The magnetic susceptibility $\chi(T)$ and the specific heat $C/T$ of URu$_2$Zn$_{20}$ are displayed in Fig. 1 and compared to the data for UCo$_2$Zn$_{20}$. Fits of the data to a Curie-Weiss law (Fig. 1 (a)) at high temperature give the effective moments $\mu_{eff} = 3.69 \mu_B$ for URu$_2$Zn$_{20}$ and 3.44 $\mu_B$ for UCo$_2$Zn$_{20}$. The Curie-Weiss temperatures are $\theta = -172$ K and $-65$ K for the Ru and Co cases, respectively. For URu$_2$Zn$_{20}$, the magnetic susceptibility $\chi(T)$ increases monotonically as the temperature decreases to the value $\chi(2K) \approx 0.0123$ emu/mole. At 2 K, the susceptibility of UCo$_2$Zn$_{20}$ is about 0.037 emu/mole, which is 3 times larger than for the Ru case. The specific heat is plotted as $C/T$ vs $T$ in Fig. 1 (b). For URu$_2$Zn$_{20}$ $C/T$ has the magnitude $\gamma \approx 190$ mJ/mole-K$^2$ at 2 K. At low temperature $C/T$ follows the $T^2$ behavior expected for a phonon contribution, which permits the extrapolation of the Sommerfeld coefficient to the value $\gamma \approx 188$ mJ/mole-K$^2$. From the inset to Fig. 1 (b), it can be seen that for UCo$_2$Zn$_{20}$, $\gamma(2K)$ is approximately 500 mJ/mole-K$^2$ at 2 K, while at $T_{max} = 4.1$ K, $\gamma = 558$ mJ/mole-K$^2$; these values are 2.6 and 2.9 times larger than for URu$_2$Zn$_{20}$, respectively.

As mentioned above, the characteristic energy for spin fluctuations can be determined from the inelastic neutron scattering experiments. In Fig. 2 we plot the $Q$-averaged dynamic susceptibility $\chi''(\Delta E)$ of URu$_2$Zn$_{20}$ as a function of energy transfer $\Delta E$. This is determined from the scattering function through the formula $S_{mag} = A(n(\Delta E) + 1)f^2(Q)\chi''(\Delta E)$, where $(n(\Delta E) + 1)$ is the Bose factor and $f^2(Q)$ is the U 5f form fac-

FIG. 1: (a) Magnetic susceptibility $\chi(T)$ for URu$_2$Zn$_{20}$. The lines are Curie-Weiss fits. (b) Specific heat coefficient $C/T$ vs $T$ of URu$_2$Zn$_{20}$. Insets: Susceptibility and specific heat coefficient of UCo$_2$Zn$_{20}$; the data are from Bauer et al.

FIG. 2: Low temperature dynamic susceptibility $\chi''$ vs $\Delta E$ of URu$_2$Zn$_{20}$. (a) Pharos data at $T=7$ K ($E_i = 35$ meV). (b) LRMECS data at $T=10$ K ($E_i = 60$ meV). The lines represent Lorentzian fits with $E_0 = 13.5$ meV and $\Gamma = 9.5$ meV. Inset: low temperature dynamic susceptibility of UCo$_2$Zn$_{20}$; the data are from Bauer et al. The line is a fit to a Lorentzian with $E_0 = 3$ meV and $\Gamma = 5$ meV. The arrows indicate the peak positions predicted by the AIM for $N_f = 10$ (See Table I).
that these compounds exhibit scaling behavior with data to an inelastic Lorentzian give $E_{\text{max}}$ for UCo$_2$Zn$_{20}$. In these fits, the only adjustable parameter is a

As shown in the inset to Fig. 2, for UCo$_2$Zn$_{20}$, $\chi''(\Delta E)$ shows a peak centered near $E_{\text{max}} = 6$ meV. Fits of this data to an inelastic Lorentzian give $E_0 = 3$ meV with $\Gamma = 5$ meV, for which $E_{\text{max}} = 5.8$ meV.

Given that $\gamma(2K)_{\text{Co}}/\gamma(2K)_{\text{Ru}} = 2.63$ (alternatively $\gamma(T_{\text{max}})_{\text{Co}}/\gamma(2K)_{\text{Ru}} = 2.93$), that $\chi(2K)_{\text{Co}}/\chi(2K)_{\text{Ru}} = 3.01$, and that $E_{\text{max}}(\text{Ru})/E_{\text{max}}(\text{Co}) = 2.84$, it is clear that these compounds exhibit scaling behavior with $\chi$ and $\gamma$ scaling as $1/k_BT_{\text{eff}} = 1/E_{\text{max}}$.

We next examine whether such scaling arises due to the applicability of the AIM to these actinide compounds. Before doing so, we first check the validity of the AIM for the rare earth 4f compounds YbFe$_2$Zn$_{20}$. We apply Rajan’s Coqblin-Schrieffer model[3], which is essentially the AIM in the Kondo limit ($n_f \approx 1$) for large orbital degeneracy. In Fig. 3, we compare the data for $C_{\text{mag}}(T)$ and $\chi(T)$ to Rajan’s predictions for the $J=7/2$ case[3]. Here the LuFe$_2$Zn$_{20}$ data has been subtracted for $\chi(T)$ data. In these fits, the only adjustable parameter is a scaling parameter $T_0$, which we fix at 69.2 K. To fit to the dynamic susceptibility $\chi''(\Delta E)$ we use the results of Cox et al[4], obtained using the noncrossing approximation (NCA) to the AIM. This calculation, which was performed for the $J=5/2$ case, gives the peak position of the dynamic susceptibility at low temperature as $E_{\text{max}} = 1.36 T_0$ (Cox) where $T_0(\text{Cox}) = T_0 / 1.15$ so that $E_{\text{max}} = 1.18 T_0$. Assuming a similar result for the $J=7/2$ case, we then expect $E_{\text{max}} = 7$ meV for YbFe$_2$Zn$_{20}$. The lineshape for $\chi''(\Delta E)/\chi''(E_{\text{max}})$ determined from Fig. 4 in Cox et al using this value for $E_{\text{max}}$. Albeit we have only determined $\chi''(\Delta E)$ at one location in the zone, it is clear from these plots that the $N_J = 8$ AIM in the Kondo limit does an excellent job of fitting the susceptibility $\chi(T)$, magnetic specific heats $C_{\text{mag}}$, and characteristic energy $E_{\text{max}}$ of this rare earth compound.

Turning now to the actinide compounds, we note that Rajan’s calculations[3] for a 2J+1 Kondo impurity give the following zero-temperature limits for the specific heat, and magnetic susceptibility:

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

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

where $R$ is the gas constant and $C_J$ is the Curie constant. To test these scaling laws, we first note that Uranium has a possible $5f^3$ state for which $J = 9/2$ and $\mu_{eff} = 3.62\mu_B$ ($C_J = 1.64$ emu K/mole) or a possible $5f^2$ state for which $J = 4$ and $\mu_{eff} = 3.58\mu_B$ ($C_J = 1.60$ emu K/mole). Since the high temperature Curie-Weiss fit of $\chi(T)$ for URu$_2$Zn$_{20}$ gives an experimental value for the Curie constant close to the free ion value, we take $J = 9/2$. We estimate $T_0$ from the low temperature value for $\gamma$, and then determine $\chi_0$. To estimate $E_{\text{max}}$ we use the above-stated rule $E_{\text{max}} = 1.18 T_0$, which while developed for $J=5/2$ should be correct here to 10%. The results are listed in Table I, along with similar results for $J=7/2$ and $J=1/2$.

From Table I, we can see that the expected values for $\chi_0$ and $E_{\text{max}}$ are closer to the experimental values for the $J=9/2$ case than for either the $J=5/2$ or $1/2$ cases. In Fig. 4 we compare the experimental data to the predictions (red dashed lines) for the temperature dependence of $\chi(T)$ and $C_{\text{mag}}$ in the $J=9/2$ case. For the energy dependence of $\chi''(\Delta E)/\chi''(E_{\text{max}})$ at low temperature, we utilize the results of Cox et al[4], as outlined above. Again, there is only one adjustable parameter, $T_0$, which is determined from the low temperature specific heat coefficient as equal to 208 K for the Ru case and 69 K for the Co case. The fitting is very poor in several respects. First, the expected values of $T_{\text{max}}$ for both $\chi(T)$ and $C_{\text{mag}}(T)$ are much higher than observed in the experiment, and indeed for URu$_2$Zn$_{20}$ there is even no maximum in the experimental curve for $\chi(T)$. Even more significant is the fact that the experimental

FIG. 3: (a) Specific heat $C_{\text{mag}}$ and (b) magnetic susceptibility $\chi(T) = \chi(\gamma(2K)_{\text{Co}} - \chi(2K)_{\text{Ru}}$) for YbFe$_2$Zn$_{20}$. The $C_{\text{mag}}$ data is taken from Torikachvili et al[7]. (c) The dynamic susceptibility $\chi''(\Delta E)/\chi''(E_{\text{max}})$ determined at the $(3/2, 3/2, 3/2)$ zone boundary point. The lines are fits, for the $J=7/2$ case, to Rajan’s predictions for $C_{\text{mag}}$ and $\chi(T)$ and to Cox’s predictions for $\chi''(\Delta E)/\chi''(E_{\text{max}})$. In all three cases, there is only one common adjustable parameter $T_0$, fixed at 69.2 K.
TABLE I: Experimental and theoretical values of key quantities for URu$_2$Zn$_{20}$ and UC$_2$O$_2$Zn$_{20}$. The values for the scaling temperature $T_0$, are obtained using $\gamma_{2K} = 188$ mJ/mol-$K^2$ for URu$_2$Zn$_{20}$ and $\gamma_{max} = 558$ mJ/mol-$K^2$ for UC$_2$O$_2$Zn$_{20}$. For $J=9/2$ and $5/2$, the Curie constant used in the calculation is the $5^3$ free ion value while for $J=1/2$, $C_J$ is obtained from the Curie-Weiss fit to the low temperature magnetic susceptibility.

<table>
<thead>
<tr>
<th>$T_0$(K)</th>
<th>$T_C^{max}$(K)</th>
<th>$\chi_0^{(en)}$(emu/mol)</th>
<th>$T_{max}$(K)</th>
<th>$E_{max}$(meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ru</td>
<td>Co</td>
<td>Ru</td>
<td>Co</td>
<td>Ru</td>
</tr>
<tr>
<td>J=9/2</td>
<td>208</td>
<td>6.8</td>
<td>7.1</td>
<td>0.0124</td>
</tr>
<tr>
<td>J=5/2</td>
<td>116</td>
<td>3.8</td>
<td>3.4</td>
<td>0.0135</td>
</tr>
<tr>
<td>J=1/2</td>
<td>23</td>
<td>7.6</td>
<td>2.0</td>
<td>0.0245</td>
</tr>
</tbody>
</table>

FIG. 4: (a) Magnetic susceptibility $\chi(T)$, (b) magnetic specific heat $C_{mag}(T)$, and (c) entropy $S_{mag}(T)$ for URu$_2$Zn$_{20}$; the insets show the same quantities for UC$_2$O$_2$Zn$_{20}$[8]. The lines are fits using Rajan’s predictions for $J=9/2$. (d): The dynamic susceptibility $\chi(\Delta E)/\chi(E_{max})$ of URu$_2$Zn$_{20}$; the inset shows the data for UC$_2$O$_2$Zn$_{20}$. The lines are obtained using Cox’s results, as explained in the text.

entropy developed below 20 K is much smaller than expected. Indeed the experimental entropy appears to be closer to Rln2, which is as expected for a two-fold degeneracy (J=1/2). However, the calculations utilizing J=1/2 require very small values of $T_0$ to reproduce the specific heat coefficients, and hence yield values for the characteristic energy $E_{max}$ that disagree markedly with the experimental value (see table 1). Since $E_{max}$ is essentially equal to $k_B T_0$, this represents a very serious discrepancy. (We note in passing that our recent paper[8] attempted to compare the data for UC$_2$O$_2$Zn$_{20}$ to the predictions of the J = 5/2 AIM, calculated using the NCA. It is clear from Table 1 that such an approach will overestimate $T_{max}$, underestimate $E_{max}$ and badly overestimate the entropy.) Hence, while the J = 7/2 AIM works extremely well [7] for the susceptibility and specific heat and also reproduces the characteristic energy $E_{max}$ of the neutron spectrum of YbFe$_2$Zn$_{20}$, for these actinide compounds, the J = 9/2 AIM works well only for the low temperature scaling, but very poorly for the overall temperature dependence of $\chi(T)$ and $C(T)$; in particular the theory badly overestimates the entropy. For smaller values of $N_f$, the characteristic energy $E_{max}$ is badly underestimated by the theory.

These results suggest that the physics responsible for the low temperature heavy mass behavior in these actinide compounds is not that of local moments subject to the Kondo effect, as for the 4f electron compounds, but is that of itinerant 5f electrons subject to correlation enhancement. Since the peaks observed in $C_{mag}(T)$ for both the Ru and Co cases and in $\chi(T)$ for the Co case occur at a much lower temperature than the characteristic temperature $E_{max}/k_B$, they are very probably associated with low temperature magnetic correlations, which exist only in the vicinity of some critical wavevector $Q_N$, and which yield only a fraction of Rln2 for the entropy. In this regard, the behavior is similar to that of UBe$_{13}$, where Q-dependent antiferromagnetic fluctuations occur on a much smaller energy scale (∼ 1 meV) than the scale of the demagnetizing fluctuations (13 meV)[14]. We expect that careful measurements on single crystals of UC$_2$O$_2$Zn$_{20}$ and URu$_2$Zn$_{20}$ should reveal such low energy Q-dependent fluctuations.

Work at UC Irvine is supported by U. S. DOE Grant No. DE-FG02-03ER46036. Work at ORNL was supported by the Scientific User Facilities Division Office of Basic Energy Sciences, DOE and was managed by UT-Battelle, LLC, for DOE under Contract DE-AC05-000R22725. Work at Los Alamos was performed under the auspices of the U. S. DOE. Work at ANL is supported by DOE-BES under contract DE-AC02-06CH11357. Work at the Ames Laboratory was supported by the DOE BES under Contract No. DE-AC02-07CH11358.