Neutron scattering and scaling behavior in URu_2Zn_{20} and $YbFe_2Zn_{20}$

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The dynamic susceptibility $\chi''(E)$, measured by inelastic neutron scattering measurements, shows a broad peak centered at $E_{max} = 16.5$ meV for the cubic actinide compound URu_2Zn_{20} and 7 meV at the (1/2, 1/2, 1/2) zone boundary for the rare earth counterpart compound $YbFe_2Zn_{20}$. For URu_2Zn_{20}, the low temperature susceptibility and magnetic specific heat coefficient $\gamma = C_{mag}/T$ take the values $\chi = 0.0123$ emu/mole and $\gamma = 190$ mJ/mole-K^2 at $T = 2$ K. These values are roughly three times smaller, and E_{max} is three times larger, than recently reported for the related compound UCo_2Zn_{20}, so that χ and γ scale inversely with the characteristic energy for spin fluctuations, $T_{sf} = E_{max}/k_B$. While $\chi(T)$, $C_{mag}(T)$, and E_{max} of the $4f$ compound $YbFe_2Zn_{20}$ are very well described by the Kondo impurity model, we show that the model works poorly for the URu_2Zn_{20} and UCo_2Zn_{20}, suggesting that the scaling behavior of the actinide compounds arises from spin fluctuations of *itinerant* $5f$ electrons.

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An important property of heavy fermion (HF) materials is a scaling law whereby the low temperature susceptibility χ 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_{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 χ, 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 $N_J (= 2J + 1$ for rare earths). In particular, for large degeneracy ($N_J > 2$) both the calculated $\gamma(T)$ and $\chi(T)$ exhibit maxima at a temperature αT_K where α is a constant that depends on N_J. This kind of behavior is observed in rare earth compounds such as $YbAgCu_4$[5], $CeSn_3$ and YbCuAl[6] and $YbFe_2Zn_{20}$[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 occurence of scaling. Nevertheless, we have recently shown[8] that the actinide compound UCo_2Zn_{20} exhibits a maximum in the susceptibility and a specific heat coefficient that are strikingly similar to those seen in the rare earth compound $YbFe_2Zn_{20}$. 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_2Zn_{20} together with results for the magnetic susceptibility and specific heat of single crystalline samples. We also present the INS data on single crystal $YbFe_2Zn_{20}$. Both compounds belong to a new family of intermetallic compounds RX_2Zn_{20} (R = lanthanide, Th, U; X = transition metal)[7, 9–12] which crystallize in the cubic $CeCr_2Al_{20}$ 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 ~ 6 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-

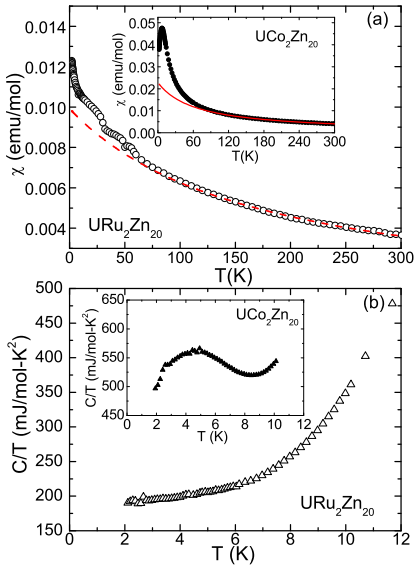

FIG. 1: (a) Magnetic susceptibility $\chi(T)$ for URu_2Zn_{20}. The lines are Curie-Weiss fits. (b) Specific heat coefficient C/T vs T of URu_2Zn_{20}. Insets: Susceptibility and specific heat coefficient of UCo_2Zn_{20}; the data are from Bauer *et al*[8].

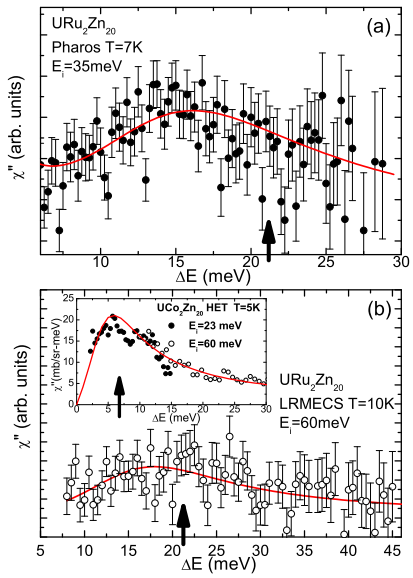

FIG. 2: Low temperature dynamic susceptibility χ'' vs ΔE of URu_2Zn_{20}. (a) Pharos data at T=7 K (E_i = 35 meV). (b) LRMECS data at T=10 K (E_i = 60 meV). The lines represent Lorentzian fits with E_0=13.5 meV and Γ= 9.5 meV. Inset: low temperature dynamic susceptibility of UCo_2Zn_{20}; the data are from Bauer *et al*[8]. The line is a fit to a Lorentzian with E_0=3 meV and Γ= 5 meV. The arrows indicate the peak positions predicted by the AIM for $N_J = 10$ (See Table I).

vice (SQUID) magnetometer. The specific heat was measured in a Quantum Design PPMS system. For URu_2Zn_{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 $YbFe_2Zn_{20}$ 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_2Zn_{20}, we used the non-magnetic counterpart compound $ThCo_2Zn_{20}$ to determine the scaling of the nonmagnetic scattering between low Q and high Q; for $YbFe_2Zn_{20}$, 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_2Zn_{20} are displayed in Fig. 1 and compared to the data for UCo_2Zn_{20}. Fits of the data to a Curie-Weiss law (Fig. 1 (a)) at high temperature give the effective moments μ_{eff} = 3.69 μ_B for URu_2Zn_{20} and 3.44 μ_B for UCo_2Zn_{20}. The Curie-Weiss temperatures are θ = -172 K and -65 K for the Ru and Co cases, respectively. For URu_2Zn_{20}, the magnetic susceptibility $\chi(T)$ increases monotonically as the temperature decreases to the value $\chi(2K) \simeq 0.0123$ emu/mole. At 2 K, the susceptibility of UCo_2Zn_{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_2Zn_{20} C/T has the magnitude $\gamma \simeq 190$ 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 $\gamma \simeq 188$ mJ/mole-K^2. From the inset to Fig. 1 (b), it can be seen that for UCo_2Zn_{20}, $\gamma(2K)$ is approximately 500 mJ/mole-K^2 at 2 K, while at T_{max} = 4.1 K, γ = 558 mJ/mole-K^2; these values are 2.6 and 2.9 times larger than for URu_2Zn_{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_2Zn_{20} as a function of energy transfer ΔE. 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-

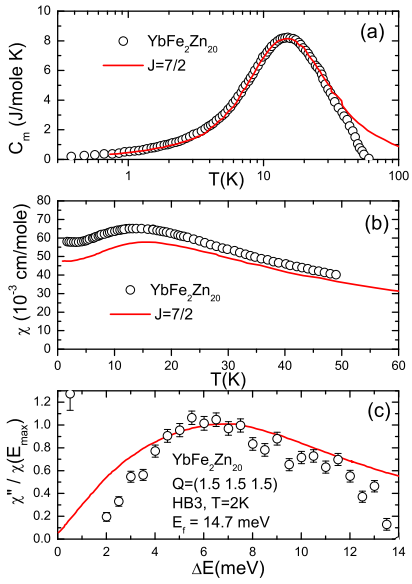

FIG. 3: (a) Specific heat C_{mag} and (b) magnetic susceptibility $\chi(T)(=\chi_{YbFe_2Zn_{20}}-\chi_{LuFe_2Zn_{20}})$ for $YbFe_2Zn_{20}$. The C_{mag} data is taken from Torikachvili *et al*[7]. (c) The dynamic susceptibility $\chi\prime\prime(\Delta E)/\chi\prime\prime(E_{max})$ determined at the (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_{mag} and $\chi(T)$ and to Cox's predictions for $\chi\prime\prime(\Delta E)/\chi\prime\prime(E_{max})$. In all three cases, there is only one common adjustable parameter T_0, fixed at 69.2 K.

tor. Both the Pharos data and the LRMECS data for $\chi''(\Delta E)$ for URu_2Zn_{20} exhibit broad peaks with peak position E_{max} at an energy transfer $\Delta E \simeq$ of order 16 meV. The dynamic susceptibility $\chi''(\Delta E)$ can be fit by a Lorentzian power function as $\chi''(\Delta E)= \chi'(0)\Delta E(\Gamma/\pi)/[(\Delta E-E_0)^2+\Gamma^2]$ with the parameters E_0 = 13.5 meV and Γ= 9.5 meV, giving E_{max} = 16.5 meV. As shown in the inset to Fig. 2, for UCo_2Zn_{20}, $\chi''(\Delta E)$ shows a peak centered near E_{max} = 6 meV. Fits of this data to an inelastic Lorentzian give E_0 = 3 meV with Γ = 5 meV, for which E_{max} = 5.8 meV.

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

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 $4f$ compound $YbFe_2Zn_{20}$. We apply Rajan's Coqblin-Schrieffer model[3], which is essentially the AIM in the Kondo limit ($n_f \simeq 1$) for large orbital degeneracy. In Fig. 3, we compare the data for $C_{mag}(T)$ and $\chi(T)$ to Rajan's predictions for the J=7/2 case[3]. Here the $LuFe_2Zn_{20}$ 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 $\chi\prime\prime(\Delta E)$ 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_{max} = 1.36 $T_0(Cox)$ where $T_0(Cox) = T_0$ / 1.15 so that E_{max} = 1.18 T_0. Assuming a similar result for the $J = 7/2$ case, we then expect E_{max} = 7 meV for $YbFe_2Zn_{20}$. The lineshape for $\chi\prime\prime(\Delta E)/\chi\prime\prime(E_{max})$ was determined from Fig. 4 in Cox *et al* using this value for E_{max}. Albeit we have only determined $\chi\prime\prime(\Delta E)$ 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_{mag}, and characteristic energy E_{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:

$$\gamma_0 = \pi JR/3T_0$$

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

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 μ_{eff}=3.62μ_B (C_J= 1.64 emu K/mole) or a possible $5f^2$ state for which J = 4 and μ_{eff}=3.58μ_B (C_J= 1.60 emu K/mole). Since the high temperature Curie-Weiss fit of $\chi(T)$ for URu_2Zn_{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 γ, and then determine χ_0. To estimate E_{max} we use the above-stated rule E_{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 = 5/2 and J = 1/2.

From Table I, we can see that the expected values for χ_0 and E_{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_{mag} in the J = 9/2 case. For the energy dependence of $\chi\prime\prime(\Delta E)/\chi\prime\prime(E_{max})$ 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_{max} for both $\chi(T)$ and $C_{mag}(T)$ are much higher than observed in the experiment, and indeed for URu_2Zn_{20} there is even no maximum in the experimental curve for $\chi(T)$. Even more significant is the fact that the experimental

TABLE I: Experimental and theoretical values of key quantities for URu_2Zn_{20} and UCo_2Zn_{20}. The values for the scaling temperature T_0 are obtained using γ_{2K} = 188 mJ/mol-K^2 for URu_2Zn_{20} and γ_{max} = 558 mJ/mol-K^2 for UCo_2Zn_{20}. For J=9/2 and 5/2, the Curie constant used in the calculation is the $5f^3$ free ion value while for J=1/2, C_J is obtained from the Curie-Weiss fit to the low temperature magnetic susceptibility.

	T_0(K)		T^C_{max}(K)		$\chi_0(\frac{emu}{mole})$		$T^\chi_{max}(K)$		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

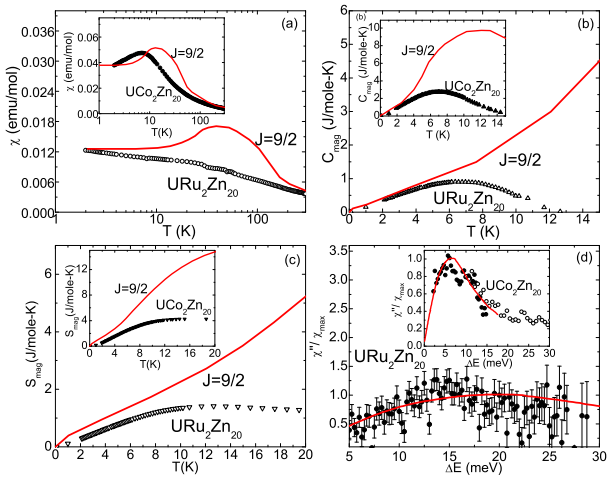

FIG. 4: (a) Magnetic susceptibility $\chi(T)$, (b) magnetic specific heat $C_{mag}(T)$, and (c) entropy $S_{mag}(T)$ for URu_2Zn_{20}; the insets show the same quantities for UCo_2Zn_{20}[8]. The lines are fits using Rajan's predicitions for J=9/2. (d): The dynamic susceptibility $\chi''(\Delta E)/\chi''(E_{max})$ of URu_2Zn_{20}; the inset shows the data for UCo_2Zn_{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_BT_0[1], this represents a very serious discrepancy. (We note in passing that our recent paper[8] attempted to compare the data for UCo_2Zn_{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^C_{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 Emax of the neutron spectrum of $YbFe_2Zn_{20}$, 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_J, 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 ($\sim$ 1 meV) than the scale of the demagnetizing fluctuations (13 meV)[14]. We expect that careful measurements on single crystals of UCo_2Zn_{20} and URu_2Zn_{20} should reveal such low energy Q-dependent fluctuations.

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