Neutron scattering and scaling behavior in URu₂Zn₂₀ and YbFe₂Zn₂₀

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

Y. Qiu,^{6,7} and J. R. D. Copley⁶

¹University of California, Irvine, California 92697, USA

²Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

³Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA

⁴Argonne National Laboratory, Argonne, Illinois 60439, USA

⁵Ames Laboratory, Iowa State University, Ames, Iowa 50011, USA

⁶National Institute of Standards and Technology, Gaithersburg, Maryland 20899, USA

⁷University of Maryland, College Park, Maryland 20742, USA

(Received 23 February 2010; published 5 November 2010)

The dynamic susceptibility $\chi''(\Delta E)$, measured by inelastic neutron-scattering measurements, shows a broad peak centered at E_{max} =15 meV for the cubic actinide compound URu₂Zn₂₀ and 7 meV at zone center and at the (1/2, 1/2, 1/2) zone boundary for the rare-earth counterpart compound YbFe₂Zn₂₀. For URu₂Zn₂₀, the low-temperature susceptibility and magnetic specific-heat coefficient $\gamma = C_{mag}/T$ take the values χ =0.011 emu/mole and γ =190 mJ/mole K² 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 UCo₂Zn₂₀, 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 4*f* compound YbFe₂Zn₂₀ are very well described by the Kondo impurity model, we show that the model works poorly for URu₂Zn₂₀ and UCo₂Zn₂₀, suggesting that the scaling behavior of the actinide compounds arises from spin fluctuations of *itinerant* 5*f* electrons.

DOI: 10.1103/PhysRevB.82.184407

PACS number(s): 71.27.+a, 72.15.Qm, 75.20.Hr

I. INTRODUCTION

An important property of heavy-fermion (HF) materials is a scaling law whereby the low-temperature magnetic 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 $\chi''(\Delta E)$, measured through inelastic neutron scattering (INS). Such scaling receives theoretical justification¹⁻⁴ from the Anderson impurity model (AIM), where the spinfluctuation 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 intermediate valence 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_{mag} , the susceptibility χ , and the 4f occupation number n_f , as well as the energy dependence of the INS spectra $\chi''(\Delta E)$ of polycrystalline samples.^{5,6} This is true despite the fact that the rare 4f ions are not impurities but form a lattice. Under these circumstances, properties which depend crucially on the lattice periodicity, such as transport or de Haas-van Alphen signals, behave very differently from the predictions of the AIM. The quantities C_{mag} , χ , n_f , and $\chi''(\Delta E)$, however, depend primarily on the spin fluctuations, which are sufficiently localized in these rareearth compounds that coherence appears to have only a minor effect at low temperature.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 conduc-

tion 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 dorbitals. Photoemission spectroscopy in 4f electron systems shows clear signals from local moment states at energies well below the Fermi level; the weak hybridization between the f electron and the conduction electron leads to emission near the Fermi energy ϵ_F that can be described in the context of the Anderson impurity model as a Kondo resonance.⁸ In 5f electron systems, no local states are seen, but rather broad 5f band emission is observed near ϵ_F . The Anderson lattice model is sometimes employed to understand the *f*-derived band in actinide systems⁹ while in some systems itinerantelectron band models are employed.10 Hence, despite the common occurrence of scaling, we might expect differences between the uranium and the rare-earth-based heavy-fermion materials in the details of the thermodynamics and the spin fluctuations. Nevertheless, we have recently shown¹¹ that the temperature dependence of the susceptibility and specific heat of the actinide compound UCo2Zn20 are strikingly similar to those seen in the rare-earth compound YbFe2Zn20 where Kondo theory does an excellent job of fitting the data.⁶ 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. 6 and also Fig. 3 of this paper), may also be valid for 5f HF compounds.

To accomplish this, we present herein the results of 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 YbFe₂Zn₂₀. Both compounds belong to a new family of intermetallic compounds RX_2Zn_{20} (*R*=lanthanide,

Th, and U; X=transition metal) (Refs. 6 and 12–15) which crystallize in the cubic CeCr₂Al₂₀-type structure ($Fd\bar{3}m$ space group).^{13,16} 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 *R*-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.

II. EXPERIMENT DETAILS

The crystals were grown in zinc flux.^{6,11} The magneticsusceptibility measurements were performed in a commercial superconducting quantum interference device magnetometer. The specific-heat experiments were performed in a commercial measurement system that utilizes a relaxational (time constant) method. For URu₂Zn₂₀, we performed inelastic neutron scattering on a 40 g powder sample on the lowresolution medium-energy chopper spectrometer (LRMECS) at IPNS, Argonne National Laboratory, on the highresolution chopper spectrometer (Pharos) at the Lujan center, LANSCE, at Los Alamos National Laboratory, and on the time-of-flight disk chopper spectrometer (DCS) at the NIST Center for Neutron Research. For YbFe2Zn20 the INS spectrum was obtained for two coaligned crystals of total mass 8.5 g, using the HB-3 triple-axis spectrometer at the high flux isotope reactor 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 the Pharos and LRMECS measurements of URu₂Zn₂₀, we used the nonmagnetic counterpart compound ThCo2Zn20 to determine the scaling of the nonmagnetic scattering between low Q and high Q; for YbFe₂Zn₂₀, 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 $Q^{2.17}$ Assuming that the magnetic scattering scales with the Q dependence of the 4f or 5f form factor, we subtracted the nonmagnetic component to obtain the magnetic scattering function $S_{mag}(\Delta E)$.^{5,18,19}

III. RESULTS AND DISCUSSION

The magnetic susceptibility $\chi(T)$ and the specific heat C/T of URu₂Zn₂₀ are displayed in Fig. 1 and compared to the data reported earlier for UCo₂Zn₂₀. Fits of the data to a Curie-Weiss law [Fig. 1(a)] at high temperature give the effective moments μ_{eff} =3.61±0.005 μ_B for URu₂Zn₂₀ and 3.44±0.011 μ_B for UCo₂Zn₂₀. The Curie-Weiss temperatures are θ =-145±0.5 K and -65±2 K for the Ru and Co cases, respectively. For URu₂Zn₂₀, the magnetic susceptibility $\chi(T)$ increases monotonically as the temperature decreases to the value $\chi(2K) \approx 0.0111$ emu/mole. At 2 K, the susceptibility of UCo₂Zn₂₀ is about 0.0372 emu/mole, which is 3.3 times larger than for the Ru case. The specific heat is plotted as C/T vs T in Fig. 1(b). For URu₂Zn₂₀ C/T has the magnitude $\gamma \approx 190$ mJ/mole K² at 2 K. At low temperature C/T follows the T² behavior expected for a phonon contri-



FIG. 1. (Color online) (a) Magnetic susceptibility $\chi(T)$ for URu₂Zn₂₀. The lines are Curie-Weiss fits. (b) Specific heat C/T vs *T* of URu₂Zn₂₀. Insets: susceptibility and specific heat of UCo₂Zn₂₀; the data are from Bauer *et al.* (Ref. 11).

bution, which permits the extrapolation of the Sommerfeld coefficient to the value $\gamma \approx 188 \text{ mJ/mole K}^2$. From the inset to Fig. 1(b), it can be seen that for UCo₂Zn₂₀, $\gamma(2K)$ is approximately 500 mJ/mole K² while at T_{max} =4.1 K, γ =558 mJ/mole K²; these values are 2.6 and 2.9 times larger than for URu₂Zn₂₀, respectively.

As mentioned above, the characteristic energy for spin fluctuations can be determined from the inelastic neutronscattering experiments. In Fig. 2 we plot the Q-averaged dynamic susceptibility $\chi''(\Delta E)$ of URu₂Zn₂₀ as a function of energy transfer ΔE . This is determined from the scattering function through the formula $S_{mag} = A \lfloor n(\Delta E)$ $+1]f^{2}(Q)\chi''(\Delta E)$, where $[n(\Delta E)+1]=1/[1-\exp(-\Delta E/k_{B}T)]$ is the Bose factor and $f^2(Q)$ is the U 5f form factor. Both the Pharos data and the LRMECS data for $\chi''(\Delta E)$ for URu₂Zn₂₀ exhibit broad peaks with peak position E_{max} at an energy transfer $\Delta E \simeq 15$ meV. A widely used function for the inelastic neutron-scattering spectra entails a Lorentzian power function^{20,21}

$$\chi''(\Delta E) = \chi'(0)\Delta E \left(\frac{\Gamma}{2\pi}\right) \left[\frac{1}{\left[(\Delta E - E_0)^2 + \Gamma^2\right]} + \frac{1}{\left[(\Delta E + E_0)^2 + \Gamma^2\right]}\right].$$

Using this function, the fitting results in the parameters $E_0=12.4$ meV and $\Gamma=8.9$ meV, giving $E_{max}=15.3$ meV. Fits of the data for UCo₂Zn₂₀ to the Lorentzian power function (Fig. 2, inset) give $E_0=2.2$ meV and $\Gamma=4.8$ meV, for which $E_{max}=5.3$ meV. We note that these values of E_{max} are nearly equal to the values of $k_B\theta$ derived from the high temperature susceptibility; i.e., the temperature scale for the sup-



FIG. 2. (Color online) Low-temperature dynamic susceptibility χ'' vs ΔE of URu₂Zn₂₀. Error bars in all figures represent $\pm \sigma$. (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 = 12.4 \pm 0.6 meV and Γ =8.9 \pm 0.8 meV. Inset: low-temperature dynamic susceptibility of UCo₂Zn₂₀; the data are from Bauer *et al.* (Ref. 11). The line is a Lorentzian fit with E_0 =2.2 \pm 0.5 meV and Γ =4.8 \pm 0.2 meV. The arrows indicate the peak positions predicted by the AIM for N_I =10 (See Table I).

pression of the moment is identical to the energy scale of the spin fluctuation.

Given that $\gamma(2 \text{ K})_{\text{Co}}/\gamma(2 \text{ K})_{\text{Ru}}=2.6$, that $\chi(2 \text{ K})_{\text{Co}}/\chi(2 \text{ K})_{\text{Ru}}=3.3$, and that $E_{max}(\text{Ru})/E_{max}(\text{Co})=2.8$, we see that at low temperature these compounds exhibit a factor of 3 scaling of χ , γ , and E_{max} to an accuracy of about 10–15 %.

The scaling laws represent the relationship between the energy E_{max} and degeneracy 2J+1 of the primary, Kondo-type²² spin fluctuation and the low-temperature specific heat and moment. Two aspects of the physics can affect the applicability of such scaling: crystal fields (discussed further below) and magnetic correlations. The latter can make a contribution to the low temperature specific heat and susceptibility over and beyond that of the primary spin fluctuation.²³ It is very plausible that the low-temperature peaks in C/T and χ seen in UCo₂Zn₂₀ represent the onset of such correlations. This creates some uncertainty as to the correct value of the scaling ratios [e.g., $\gamma(T_{max})_{Co}/\gamma(2 \ K)_{Ru}=2.9$] without invalidating the basic concept of scaling.

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 rareearth 4*f* compound YbFe₂Zn₂₀. 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 $C_{mag}(T)$ and $\chi_{mag}(T)$ (where the data for LuFe₂Zn₂₀ has been subtracted to determine the magnetic



FIG. 3. (Color online) (a) Specific heat $C_{mag}(T)$ [from Torikachvili *et al.* (Ref. 6) and (b) magnetic susceptibility $\chi_{mag}(T)$ for YbFe₂Zn₂₀. (c) The inelastic neutron-scattering spectra of YbFe₂Zn₂₀ at two different **Q** in the Brillouin zone. The nonmagnetic scattering has been subtracted, as described in the text. The results show little **Q**dependence. The lines are fits, for the J=7/2case, to Rajan's predictions for C_{mag} and χ_{mag} and to Cox's predictions for $\chi''(\Delta E)/\chi''(E_{max})$. In all three cases, there is only one common adjustable parameter T_0 , set to the value 69 K to give the best agreement with experiment.

contribution) to Rajan's predictions for the J=7/2 case.³ In these fits, the only adjustable parameter is a scaling parameter T_0 ; we find that the value 69 K gives the best agreement with experiment.

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 $E_{max} = 1.36k_BT_0^{Cox}$ (see Fig. 5 in Cox *et al.*⁴). The scaling temperature T_0^{Cox} is related to Rajan's scaling temperature T_0 via $T_0^{Cox} = T_0/1.15$ (see the caption of Fig. 2 in Cox *et al.*⁴). Hence for the J=5/2 case, we have $E_{max}=1.18k_BT_0$. In the absence of comparable theoretical results for other values of J, we will assume that this relationship between T_0 and E_{max} is approximately true for the J=7/2 and 9/2 cases; the error is probably of order 20%. For YbFe₂Zn₂₀, we then expect $E_{max}=7$ meV.

In Fig. 3(c) we compare the predicted line shape (as determined from Fig. 4 in Cox *et al.*⁴ using the value E_{max} =7 meV) to the data for YbFe₂Zn₂₀ at zone center and at the (1/2, 1/2, 1/2) zone-boundary point. The near identity of the spectra at these two **Q** suggests that the spin fluctuations have only a very weak *Q* dependence, i.e., that they are essentially localized. As mentioned in Sec. I, this is a necessary condition for the applicability of the AIM to periodic HF compounds. It is clear from Fig. 3 that the N_J =8 AIM in



FIG. 4. (Color online) (a) Magnetic susceptibility $\chi(T)$, (b) magnetic specific heat $C_{mag}(T)$, and (c) entropy $S_{mag}(T)$ for URu₂Zn₂₀; the insets show the same quantities for UCo₂Zn₂₀ (Ref. 11). The lines are fits using Rajan's predictions for J=9/2. The open symbols in (b) and (c) represent data corrected for the energy of the Einstein mode at 8 (7) meV in the U (Th) compounds (Ref. 24). (d): The dynamic susceptibility $\chi''(\Delta E)/\chi''(E_{max})$ of URu₂Zn₂₀; the inset shows the data for UCo₂Zn₂₀. The lines are obtained using Cox's results, as explained in the text.

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 calculations³ for a 2J+1 Kondo impurity give the following zero-temperature limits for the specific heat, and magnetic susceptibility:

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

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

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 $\mu_{eff}=3.62 \ \mu_B \ (C_J=1.64 \ \text{emu K/mole})$ or a possible $5f^2$ state for which J=4 and $\mu_{eff}=3.58 \ \mu_B \ (C_J=1.60 \ \text{emu K/mole})$. The high-

temperature Curie-Weiss fit of $\chi(T)$ for URu₂Zn₂₀ gives an experimental value for the Curie constant close to these free ion values. In what follows, we choose J=9/2 but we note that the analysis is not significantly different for the J=4 case. 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 $E_{max}=1.18T_0$, which as mentioned we expect to be correct here to 20%. The calculated results are listed in Table I, along with the 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 (solid lines) for the temperature dependence of $\chi(T)$ and C_{mag} (where the data for the corresponding Th compound have been subtracted to determine the magnetic contribution²⁴) in the J=9/2 case. For the energy dependence of $\chi''(\Delta E)/\chi''(E_{max})$ 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 lowtemperature specific-heat coefficient as equal to 208 K for the Ru case and 70 K for the Co case. The fits are very poor in several respects. First, no peak is observed in the susceptibility of URu₂Zn₂₀ as expected on the basis of the AIM/ Kondo model for J=9/2; furthermore the peak that is observed in UCo₂Zn₂₀ occurs at a temperature T_{max}^{χ} that is a factor of 2 smaller than expected. Similarly, for both compounds the peaks in the specific heat occur at temperatures T_{max}^{C} that are much smaller than expected (see Table I). Even more significant is the fact that the magnitude of the specific heat is much smaller than expected. Hence there appears to be a very serious discrepancy between the data and the Kondo model.

In Ce (Yb) HF compounds, the J=5/2 (7/2) AIM calculations fail to describe the data when the Kondo energy T_K is smaller than the crystal-field splitting. Indeed, in our previous paper,¹¹ we attempted to compare the data for UCo₂Zn₂₀ to the predictions of the AIM calculated using the NCA. The calculation assumed mixed valence between the J=4 and 9/2 states, and assumed that a large crystal-field splitting resulted in a sixfold degeneracy (effective J=5/2 behavior) at low temperature. In that work, we measured the inelastic neutron spectrum of UCo₂Zn₂₀ to energy transfers as large as 100

TABLE I. Experimental and theoretical values of key quantities for URu₂Zn₂₀ and UCo₂Zn₂₀. The values for the scaling temperature T_0 are obtained using γ_{2K} =188 mJ/mol K² for URu₂Zn₂₀ and γ_{max} =558 mJ/mol K² for UCo₂Zn₂₀. For J=9/2 and 5/2, the Curie constant used in the calculation is the 5 f^3 free ion value while for J=1/2, C_J is obtained from the Curie-Weiss fit to the low-temperature magnetic susceptibility.

	<i>T</i> ₀ (K)		$\begin{array}{c} T^C_{max} \\ (\mathrm{K}) \end{array}$		$\left(rac{\chi_0}{ ext{mole}} ight)$		T^{χ}_{max} (K)		E _{max} (meV)	
	Ru	Co	Ru	Co	Ru	Co	Ru	Co	Ru	Co
Experiment			6.8	7.1	0.0111	0.0372		7.0	16.5	5.8
J = 9/2	208	70	36.5	12.1	0.0125	0.0378	39.2	13.0	21.3	7.1
J = 5/2	116	39	34.4	11.3	0.0135	0.0412	30.3	10.2	11.9	3.9
J = 1/2	23	7.8	20	6.8	0.0245	0.0402			2.4	0.8



FIG. 5. (a) The INS spectra of URu₂Zn₂₀ and ThCo₂Zn₂₀ taken on Pharos with incident energies E_i =400 and 700 meV. The diamond is the estimated magnetic scattering χ'' , obtained as described in the text. (b) The INS spectra of URu₂Zn₂₀ in the energy range 0.1–4 meV taken on DCS with incident energies E_i =2.3 and 7.1 meV. The near equality of the high-Q and low-Q scattering suggests that all the scattering observed in this energy range is due to background.

meV but saw no additional excitations beyond the one at 5 meV. To confirm whether crystal-field excitations are present in URu₂Zn₂₀, we measured that compound together with ThCo2Zn20 on Pharos using large incident energies. In Fig. 5(a) we show the INS spectra for energy transfers up to $\Delta E = 550$ meV. The results exhibit no sign of crystal-field excitations beyond the one at 15 meV. Given that the J=9/2 (8/2) states should exhibit several excitations in a crystal field but that none are observed over such a broad energy range in either compound, we think it extremely unlikely that the discrepancies between our data and the predictions of the AIM/Kondo model arise from crystal fields. Indeed, we would like to point out that well-defined crystal fields are almost never seen in metallic uranium-based heavy-fermion compounds. What is universally seen is a single broad Kondo-type magnetic excitation peak, similar to what we see in URu₂Zn₂₀ and UCo₂Zn₂₀. In certain very heavy compounds, additional low-energy (1 meV or less) peaks due to magnetic correlations are observed, as discussed further below.

Hence, while the J=7/2 AIM works extremely well⁶ for the susceptibility and specific heat and also reproduces the characteristic energy E_{max} of the neutron spectrum of YbFe₂Zn₂₀, however, for the actinide compounds considered here, namely, URu₂Zn₂₀ and UCo₂Zn₂₀, the J=9/2 (or J=4) 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 calculations based on smaller values of N_J , the characteristic energy E_{max} is badly underestimated by the theory (see Table I). 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. Furthermore as we have argued that crystal-field/magnetic fluctuations, etc., are not likely to be the reasons behind the failure of the AIM in the description of the 5f compounds studied here, the most likely remaining candidate is that of itinerant 5f electrons subject to correlation enhancement.

In further support of this itineracy, we note that when uranium compounds such as UPd₃ exhibit local moments, then intermultiplet excitations can be observed at energies near 400 mev; no such excitation is seen for metallic compounds such as UPt₃.²⁵ The lack of such excitations in the Pharos data [Fig. 5(a)] for URu₂Zn₂₀ gives further evidence that the 5*f* electrons are itinerant, not localized, in that compounds.

Given these considerations, we believe the characteristic INS energies of 5.3 and 15.3 meV that we have observed in UCo_2Zn_{20} and URu_2Zn_{20} represent Kondo-type²² spin fluctuations of itinerant electrons as observed in many uranium compounds such as UBe_{13} ,²⁶ UPt_3 ,²⁷ and USn_3 .²⁸

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 wave vector $\mathbf{Q}_{\mathbf{N}}$, and which yield only a fraction of $R \ln 2$ for the entropy. In this regard, the behavior is similar to that of UBe_{13} or UPt_3 , where Q-dependent antiferromagnetic fluctuations occur on a much smaller energy scale ($\sim 1 \text{ meV}$ for UBe_{13} and 0.2 meV for UPt_3) than the scale of the Kondo-type²² fluctuations [13 meV for UBe₁₃ and 6 meV for UPt₃ (Refs. 26 and 27)]. Such antiferromagnetic fluctuations are large only in the vicinity of the wave vector Q_N and contain only a small fraction of the spectral weight compared to the Kondo-type fluctuations. Hence, it is not surprising that the polycrystalline averaged INS spectra in Fig. 5(b)shows no obvious excitation in the energy transfer range 0.1-4 meV; careful measurements on single crystals are required to reveal such low-energy, low-spectral-weight Q-dependent magnetic fluctuations.

IV. CONCLUSION

The static and dynamic magnetic susceptibility and the specific heat of URu_2Zn_{20} and $YbFe_2Zn_{20}$ compounds have been presented. The results show that the AIM works very well to describe the magnetic susceptibility, specific heat, and dynamic susceptibility well of the compound $YbFe_2Zn_{20}$ where the 4f electrons are localized. In the actinide compounds $URu_2Zn_{20}(UCo_2Zn_{20})$, however, the fits to the AIM temperature dependence are very poor even though the low-temperature scaling behavior expected for a J=9/2 Kondo impurity was observed. An associated problem is that the magnetic entropy generated by 20 K is too small compared to the expected value. These results suggest that the spin fluctuations in these actinide compounds arise from itinerant rather than localized 5f electrons. Antiferromagnetic fluctua-

tions may affect the specific heat. While our neutron scattering results for a polycrystalline sample saw no signs of these fluctuations in the 0.1–4 meV range, they may be observable as a small spectral weight signal in single-crystal experiments.

ACKNOWLEDGMENTS

Research at UC Irvine was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of

- ¹A. C. Hewson, *The Kondo Problem to Heavy Fermions* (Cambridge University Press, Cambridge, England, 1997).
- ²N. E. Bickers, D. L. Cox, and J. W. Wilkins, Phys. Rev. B **36**, 2036 (1987).
- ³V. T. Rajan, Phys. Rev. Lett. **51**, 308 (1983).
- ⁴D. L. Cox, N. E. Bickers, and J. W. Wilkins, J. Magn. Magn. Mater. **54-57**, 333 (1986).
- ⁵J. M. Lawrence, P. S. Riseborough, C. H. Booth, J. L. Sarrao, J. D. Thompson, and R. Osborn, Phys. Rev. B **63**, 054427 (2001).
- ⁶M. S. Torikachvili, S. Jia, E. D. Mun, S. T. Hannahs, R. C. Black, W. K. Neils, Dinesh Martien, S. L. Bud'ko, and P. C. Canfield, Proc. Natl. Acad. Sci. U.S.A. **104**, 9960 (2007).
- ⁷J. M. Lawrence, Mod. Phys. Lett. B **22**, 1273 (2008).
- ⁸ A. Koitzsch, S. V. Borisenko, D. Inosov, J. Geck, V. B. Zabolotnyy, H. Shiozawa, M. Knupfer, J. Fink, B. Buchner, E. D. Bauer, J. L. Sarrao, and R. Follath, Phys. Rev. B 77, 155128 (2008).
- ⁹J. D. Denlinger, G.-H. Gweon, J. W. Allen, C. G. Olson, M. B. Maple, J. L. Sarrao, P. E. Armstrong, Z. Fisk, and H. Yamagami, J. Electron Spectrosc. Relat. Phenom. **117-118**, 347 (2001).
- ¹⁰E. Guziewicz, T. Durakiewicz, M. T. Butterfield, C. G. Olson, J. J. Joyce, A. J. Arko, J. L. Sarrao, D. P. Moore, and L. Morales, Phys. Rev. B **69**, 045102 (2004); T. Durakiewicz, C. D. Batista, J. D. Thompson, C. G. Olson, J. J. Joyce, G. H. Lander, J. E. Gubernatis, E. Guziewicz, M. T. Butterfield, A. J. Arko, J. Bonca, K. Mattenberger, and O. Vogt, Phys. Rev. Lett. **93**, 267205 (2004).
- ¹¹E. D. Bauer, C. Wang, V. R. Fanelli, J. M. Lawrence, E. A. Goremychkin, N. R. de Souza, F. Ronning, J. D. Thompson, A. V. Silhanek, V. Vildosola, A. M. Lobos, A. A. Aligia, S. Bobev, and J. L. Sarrao, Phys. Rev. B **78**, 115120 (2008).
- ¹²E. D. Bauer, J. D. Thompson, J. L. Sarrao, and M. F. Hundley, J. Magn. Magn. Mater. **310**, 449 (2007).
- ¹³ V. M. T. Thiede, W. Jeitschko, S. Niemann, and T. Ebel, J. Alloys Compd. **267**, 23 (1998).
- ¹⁴A. P. Gonçalves, J. C. Waerenborgh, A. Amaro, M. Godinho, and M. Almeida, J. Alloys Compd. **271-273**, 456 (1998).
- ¹⁵S. Jia, S. L. Bud'ko, G. D. Samolyuk, and P. C. Canfield, Nat. Phys. **3**, 334 (2007).
- ¹⁶S. Niemann and W. Jeitschko, J. Solid State Chem. **114**, 337 (1995).
- ¹⁷Alhough the coherent approximation (Q^2 dependence) is not expected to be valid for a single crystal, the resulting magnetic

Materials Sciences and Engineering under Award No. DE-FG02-03ER46036. Work at ORNL was supported by the Scientific User Facilities Division Office of Basic Energy Sciences (BES), DOE. Work at ANL was supported by DOE-BES under Contract No. DE-AC02-06CH11357. Work at the Ames Laboratory was supported by the DOE-BES under Contract No. DE-AC02-07CH11358. Work at Los Alamos, including work performed at the Los Alamos Neutron Science Center, was also supported by the DOE-BES. Work at NIST utilized facilities supported in part by the National Science Foundation under Agreement No. DMR-0454672.

scattering in this single crystal is identical to that obtained in polycrystals [A. D. Christianson *et al.* (unpublished)] where the approximation is valid. This may reflect the very complex unit cell for this compound which includes 184 atoms/unit cell.

- ¹⁸A. P. Murani, Phys. Rev. B 28, 2308 (1983).
- ¹⁹E. A. Goremychkin and R. Osborn, Phys. Rev. B **47**, 14280 (1993).
- ²⁰J.-Y. So, S.-J. Oh, J.-G. Park, D. T. Adroja, K. A. McEwen, and T. Takabatake, Phys. Rev. B **71**, 214441 (2005).
- ²¹C.-K. Loong, B. H. Grier, S. M. Shapiro, J. M. Lawrence, R. D. Parks, and S. K. Sinha, Phys. Rev. B **35**, 3092 (1987).
- ²²We call these fluctuations "Kondo-type" because they represent excitation of the magnetic moment at temperatures $T > E_{max}/k_B$ but where the moment arises from itinerant rather than local electrons.
- ²³C. H. Wang, J. M. Lawrence, A. D. Christianson, E. A. Goremychkin, V. R. Fanelli, K. Gofryk, E. D. Bauer, F. Ronning, J. D. Thompson, N. R. de Souza, A. I. Kolesnikov, and K. C. Littrell, Phys. Rev. B **81**, 235132 (2010).
- ²⁴In our recent paper (Ref. 11) we found that the optic phonon peak seen at 8 meV in UCo₂Zn₂₀ occurs at 7 meV in ThCo₂Zn₂₀. This shift arises from the contribution of the 5*f* electrons to the crystal bonding. The specific heat for this optic mode will thus be larger for the latter compound, causing an error at low temperature in the magnetic specific heat obtained by the U-Th subtraction. The open symbols in Fig. 4 have corrected for this effect, assuming Einstein modes with three degrees of freedom. The effect is small for T < 20 K, hence the small magnetic entropy cannot be explained on this basis.
- ²⁵M. J. Bull, K. A. McEwen, R. Osborn, and R. S. Eccleston, Physica B **223-224**, 175 (1996).
- ²⁶S. Coad, A. Hiess, D. F. McMorrow, G. H. Lander, G. Aeppli, Z. Fisk, G. R. Stewart, S. M. Hayden, and H. A. Mook, Physica B 276-278, 764 (2000).
- ²⁷ A. I. Goldman, G. Shirane, G. Aeppli, E. Bucher, and J. Hufnagl, Phys. Rev. B **36**, 8523 (1987); G. Aeppli, E. Bucher, C. Broholm, J. K. Kjems, J. Baumann, and J. Hufnagl, Phys. Rev. Lett. **60**, 615 (1988).
- ²⁸M. Loewenhaupt and C. K. Loong, Phys. Rev. B **41**, 9294 (1990).