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Crystal electric fields in heavy-electron metals: The specific heats of U_2Zn_{17} and $CeCu_6$ to 70 K

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Two temperature scales—(1) T_0 (single impurity) and/or T_F^* (degenerate Fermi) and (2) crystal-electric-field splitting(s) Δ—provide the basis for dividing the excitation spectra of heavy-electron systems into three regimes. They are (i) for $T_0 < T < \Delta$, the specific heat and susceptibility are that due to independent f atoms in a crystal field; (ii) for $T \sim T_0$, a bump appears in the specific heat due to the Kondo compensation; (iii) for $T < T_0$, interaction effects between the f atoms lead to a Fermi-liquid solid where band-structure effects show up in the low-temperature properties $(T < T_F^*/100)$. This paper is primarily concerned with region (i). In particular, we present new specific-heat data for U_2Zn_{17} up to 70 K and $CeCu_6$ to 80 K. In comparing these data with those for other heavy-electron materials we conclude that the specific heat for $T > T_0$ is qualitatively consistent with the existence of crystal-electric-field levels, but that the overall experimental entropy is too high. We present a tentative model to account for this discrepancy through the use of Fermi statistics to describe transitions in a stylized band model with two peaks. Crystal-field levels in most cases have *not* been confirmed by neutron scattering.

I. INTRODUCTION

Heavy-fermion materials are usually identified by their striking behavior at very low temperatures. The linear coefficient of specific heat and the low-temperature susceptibility indicate a huge effective mass; the resistivity bends over and tends toward zero, signaling coherence effects. However, unusual and distinctive features are also seen in these materials at higher temperatures. For example, even the conventionally dull specific heat shows structure at temperatures as high as 100 K. Structure at various temperature scales is also seen in other, nonequilibrium properties—thermopower and magnetoresistance are good examples. In this paper we concentrate on the specific heat.

Since heavy-fermion materials are rare-earth and actinide-element compounds, an obvious first scale is the single-impurity Kondo temperature. However, even a Kondo peak scaled to lattice concentrations does not fully explain the data. Rare earths, and Ce in particular, are well known to exhibit crystal-electric-field (CEF) effects. Experiments are now beginning to see CEF levels in uranium compounds as well. As a start at understanding energy scales in these materials, it seems reasonable to investigate theoretically the effect CEF will have on, for example, the static equilibrium properties. One must note, however, that without neutron scattering experiments it is often difficult to directly extract the existence, let alone the level spacings, of CEF levels from equilibrium measurements.

Specific-heat measurements are a more sensitive probe of crystal-electric-field levels than susceptibility measurements. The reason can be seen by contrasting these two quantities for a simple two-level system with the lower level having a degeneracy g_1 and effective magnetic moment p_1 (in units of μ_B) and the upper level, lying Δ above the lower, with degeneracy $g_2 = g_1 g_r$ (g_r is the relative degeneracy) and moment p_2. The specific heat $(\beta = 1/k_B T)$

$$\frac{C}{k_B} = \frac{g_r(\beta\Delta)^2 \exp(-\beta\Delta)}{[1 + g_r \exp(-\beta\Delta)]^2}$$

has a maximum at $\beta\Delta \sim 2$, while the susceptibility

$$\chi = \left[\frac{\mu_B^2}{3k_B T}\right] \frac{p_1^2 + p_2^2 g_r \exp(-\beta\Delta)}{1 + g_r \exp(-\beta\Delta)}$$

only deviates slightly from the otherwise expected Curie-law behavior. Combination of both measurements can permit the identification of the levels, their degeneracies, and effective moments, data which could also be obtained by more precise but also more difficult neutron-scattering measurements.

Crystal-electric-field levels are more usually of interest for magnetic ions in insulators than in metals where the contributions of the conduction electrons (and phonons for the specific heat) typically dominate the equilibrium properties. However, the partially filled f-electron states in the rare earths and actinides are so localized that the CEF splittings are small enough $(\Delta \sim 100$ K) [compared

to the transition metals where the CEF splittings of the d electrons exceed 1000 K] that they can be detected by specific-heat measurements. There is extensive literature[1,2] for the levels of these atoms as impurities in insulators, but very little concerning the levels for such impurities in metals. In addition, there is almost no experimental evidence for U CEF levels in metallic compounds, whereas there is evidence for Ce CEF levels (cf. Table IV for references). In fact, this paper offers one of the first examples of crystal-electric-field-like levels in U_2Zn_{17} (see Sec. IV). We note also that CEF levels have been reported in UBe_{13}.[3]

An essential difficulty in detecting CEF levels in metals is the presence of a large lattice contribution to the specific heat, in addition to that from the conduction electrons. Accordingly, it is always necessary to subtract the specific heat of an isostructural compound having no f electrons and similar lattice constant. For example, we use data for Th_2Zn_{17} to approximate the lattice contribution in U_2Zn_{17}. In addition, Th_2Zn_{17} is a metal, so in the calculation of $\Delta C(T)$ which is done on a per mole basis,

$$\Delta C = C(U_2ZN_{17}) - C(Th_2Zn_{17}) ,$$

we are necessarily removing some of the conduction-electron contribution. If we could be sure that the number of non-f-electrons were the same in the heavy-electron and comparison compounds (as might be the case if U has a f^2 configuration), then the resulting $\Delta C(T)$ would contain essentially only contributions from the f electrons. But given that any heavy-electron compound contains admixtures of at least two valences and that, furthermore, the conduction electrons are hybridized with the f electrons, we know that $\Delta C(T)$ must contain, besides the f-electron contribution, some positive residue of the conduction electrons. Still we suppose that this residue has the weak electronic contribution expected of ordinary metals.[4] Accordingly, we can integrate $\Delta C(T)$ to extract the entropy difference $\Delta S(T)$ due to the f electrons,

$$\Delta S(T) = \int_0^T dT' \frac{\Delta C(T')}{T'} .$$

This $\Delta S(T)$ is thus a measure of the number of degrees of freedom per f atom $[N_{dof}(T)]$, according to the relation

$$\Delta S(T)/R = \ln[N_{dof}(T)] .$$

As we shall see the deduced $N_{dof}(T)$ is consistent with a CEF model for Ce compounds, but leads to a serious inconsistency in the case of U_2Zn_{17}.

In order to identify the features of the specific heat, models are needed. To start, we include the obvious effects first, and then consider possible refinements. In the case of heavy fermions, these "obvious" effects are single-impurity Kondo behavior, coherency effects beginning at the effective degeneracy temperature, and crystal-field effects. The plan of the paper is as follows. Section II describes a "standard model" for the excitations of heavy-electron metals with emphasis on relevant energy scales. The experimental methods are described

in Sec. III, and results for U_2Zn_{17} and $CeCu_6$ are presented in Sec. IV. Section V contains discussion of these results, with reference to the "standard model" of Sec. II. Conclusions appear in Sec. VI.

II. "STANDARD MODEL" FOR EXCITATIONS IN HEAVY-ELECTRON METALS

In the absence of any comprehensive theory for heavy-electron metals, a series of models has evolved to cover various temperature ranges. Here we attempt to combine the various points of view into a "standard model" which will serve as a basis both for summarizing the data and for discussing flaws in the model itself. At the outset we stress that the "standard model" has developed in response to the data for Ce compounds. There is little evidence for its applicability in U compounds; in fact, one aim of this paper is to stimulate more high- (and low-) temperature measurements on U systems.

Roughly speaking there are two important temperatures: T_0 characterizing the single-impurity behavior and T_F^* characterizing the very-low-temperature "coherent" behavior. Table I lists these temperatures, together with the effective masses that can be associated with the low-temperature specific heat. The characteristic numbers are deduced in the following ways.

T_0: The inverse of the high-temperature magnetic susceptibility versus temperature has the following interpolation formulas for spin $S = \frac{1}{2}$:

$$\chi^{-1}(T) \propto T + 1.82 T_0 \quad \text{for } 0.6T_0 < T < 20T_0 .$$

Since the bulk of the ground state levels are doublets (only CeB_6 is believed to have a quartet ground state) and the crystal-field splitting usually exceeds T_0, this formula is a reasonable guide to T_0. In Table I the negative temperature intercept of $\chi^{-1}(T)$ is taken to be $2T_0$ for simplicity.

T_F^*: At temperatures far below the effective degeneracy temperature $k_B T_F^*[\equiv(\hbar k_F)^2/(2m^*)]$, $C(T)/T$ approaches the zero-temperature specific-heat coefficient $\gamma(0)$ ($= m^* k_F k_B^2/3\hbar^2$) where the value of k_F is deduced via $n = k_F^3/3\pi^2$. In Table I we assume that both Ce and U have one conduction electron. [Since $T_F^* \propto n/\gamma(0)$ other choices for n can easily be translated into T_F^*.]

To date, there is no theoretical connection between T_0 and T_F^*, nor does Table I suggest one. There are at least two problems connected with deducing T_0, under the assumption that the single-impurity susceptibility is valid for the concentrated system. First, there is always the possibility that there may be yet another energy scale associated with the RKKY interactions between U atoms, denoted by I_{RKKY}. The reliability of the formula above for the inverse susceptibility will then depend on I_{RKKY} being much less than T_0. One way of judging whether or not we are in this limit (i.e., $I_{RKKY} \ll T_0$) is by comparing the antiferromagnetic transition temperatures $(T_N \sim I_{RKKY})$ with T_0. Tables I and II suggest that the Néel temperature T_N is generally smaller than T_0 for Ce compounds and generally larger than T_0 for U compounds.

TABLE I. Single-impurity temperature T_0 and effective degeneracy temperature T_F^* for some heavy-electron metals. See discussion in the text (Sec. II) for methods used to extract T_0 from the high-temperature susceptibility and T_F^* from the specific-heat coefficient $\gamma(T)=C(T)/T$ "extrapolated" to zero temperature. Note that T_0 may be larger or smaller than T_F^*. All T_0 are deduced from the $\chi(T)$ data in Ref. 41 unless otherwise indicated. All T_F^* and m^*/m values are from Ref. 42; the source of the $\gamma(0)$ value used in the calculation is cited.

	$CeAl_3$	$CeCu_2Si_2$	$CeCu_6$	UBe_{13}	UPt_3	U_2Zn_{17}
T_0 (K)	20[a]	60	20	35[b]	>25[c]	<65[d]
T_F^* (K)	34[e]	41[f]	28[g]	37[h]	91[i]	50[j]
m^*/m	440	540	670	430	270	300

[a]Reference 45.
[b]Reference 46.
[c]Reference 47.
[d]Reference 48.
[e]Reference 19.
[f]Reference 49.
[g]Reference 20.
[h]Reference 50.
[i]Reference 51.
[j]Based on the low-temperature extrapolated value for $\gamma(0)$ of 350 mJ/mol U K^2 found in this work.

The second possible problem is that the value of T_0 deduced from the inverse susceptibility may be influenced by the presence of a comparable CEF level. In the one case for which relevant data exists we can clearly illustrate this. The high-temperature inverse susceptibility for $Ce_xLa_{1-x}B_6$ (per mole of Ce) has the concentration-independent value of $T_0=50$ K.[6] On the other hand, at low concentrations of Ce the low-temperature inverse susceptibility yields $T_0\sim 1$ K^6. We know that there is a crystal-field splitting of 500 K between the low-lying quartet and the doublet.[7] So we ex-

pect that this splitting has worked to produce a higher value of T_0.[8] In addition we cannot discount that the RKKY and other interactions between the Ce atoms have raised the low T_0 (~ 1 K) for the dilute system to a higher value in the concentrated one.[9]

Given that T_0 and T_F^* are comparable, the simplest procedure is to consider temperature scales that are large compared to T_0, comparable to T_0, and small compared to T_0 and T_F^*. These three regimes characterize the "standard model" summarized in Table III.

Single-impurity regime. ($T > T_0$). The susceptibility

TABLE II. Antiferromagnetic transition temperatures T_N for Ce compounds (Ref. 43) and U compounds (Ref. 44), arranged by crystal structure of decreasing symmetry.

Crystal structure	Symmetry	Ce compound (typical examples)	T_N (K)	U compound (typical examples)	T_N (K)
NaCl	cubic			UN	52
				UP	125
				UAs	127
				USb	241
CsCl	cubic	CeMg	18–20		
		CeZn	29–36		
$MgCu_2$	cubic	$CeAl_2$	4–4.6	UMn_2	260
		$CePt_2$	1.6		
$AuCu_3$	cubic	$CeIn_3$	11	UIn_3	100
				UGa_3	70
		$CeTl_3$	no	UTl_3	85
		$CePb_3$	no	UPb_3	32
FeB	orth	CeCu	2.7		
		CeGe	10		
$CeCu_2$	orth	$CeZn_2$	7		
$BaCd_{11}$		$CeCd_{11}$	no	UCd_{11} [a]	15
Th_2Zn_{17}				U_2Zn_{17} [b]	10

[a]Reference 41.
[b]Reference 41.

TABLE III. Description of the standard model for the excitations in a heavy-electron metal in terms of the temperature ranges, nature of the excitation, form of the specific heat, and typical examples.

Temperature range	Nature of excitations	Expected specific heat	Typical examples
$T_0 < T < \Delta$	f atoms interacting with conduction electrons and CEF levels	Peaks in $\Delta C(T)$ $T_{peak} \sim \Delta/2.4$	$CeAl_3$ $CeCu_2Si_2$ UBe_{13}
$T \sim T_0$	Conduction electrons Kondo screen f-atom spin	Peak in $\Delta C(T)$ $T_{peak} \sim T_0$	$CeAl_3$
$T \ll T_F^*, T_0$	Fermi liquid with band structure	Low-temperature structure in properties	$CeAl_3$ $CeCu_6$

is well explained as the sum of susceptibilities for single f atoms, each interacting only with the conduction electrons. As the temperature decreases, $\chi(T)$ changes from a Curie law to the interpolation formula used to deduce T_0 due to interactions with the conduction electrons. At still lower temperatures the impurity spin ($S = \frac{1}{2}$ usually assumed in theories) would be completely screened by the conduction electrons, if interactions effects in concentrated systems did not affect the susceptibility.

For $T > T_0$, the specific heat should be that of independent f atoms—i.e., we expect to see the effect of the crystal-field levels as peaks in the specific heat. The position of the peak T_{peak} can be used to estimate the crystal-electric-field splitting Δ according to $\Delta = $ (prefactor)T_{peak}; the prefactor 2.4 is only weakly dependent on the relative degeneracy. (For example, for Ce with $J = \frac{5}{2}$ in a cubic field, the prefactor is 2.23 if the quartet is lower and 2.65 if the doublet is lower. For U this factor can be as large as 3.0 if two quartets lie above the ground-state doublet.)

This simple-minded approach works best for $CeAl_3$ (see Table IV) where the CEF splitting[10] is roughly twice the temperature of the peak in the electronic specific heat. However, even in this case there is difficulty with

TABLE IV. Electronic entropy and temperature of electronic specific-heat maximum, with comparison of the smallest measured crystal-electric-field splitting, for the heaviest heavy-electron compounds. With two exceptions only those compounds are listed for which a comparison compound is available. If the peak in $\Delta C(T)$ can be interpreted as a Schottky anomaly then, assuming only doublet levels, the smallest CEF splitting should be $\sim 2.4 T_{peak}$.

Heavy-electron compound	Comparison compound	$\Delta S/R$ at stated temperature	Temperature of ΔC peak (K)	Smallest CEF splitting (K)
$CeAl_3$	$LaAl_3$	ln6 at 200 K[a]	30[a]	60[b]
$CeAl_2$	$LaAl_2$	ln2 at 200 K[c]	40[c]	100[d]
$CeCu_2Si_2$	$LaCu_2Si_2$	ln6 at 400 K[e,f]	150[e]	863[g]
$CeRu_2Si_2$	$LaRu_2Si_2$	$>$ln2 at 100 K[h]		
$CeCu_6$	$LaCu_6$	$<$ln4 at 70 K[i] $>$ln4 at 80 K[k]	27[j] 26,70[k]	
CeB_6	LaB_6	ln6 at 300 K[l]		530[m]
UBe_{13}	$ThBe_{13}$	ln1.5 at 10 K[n] $<$ln3 at 40 K[q] $<$ln4 at 120 K[r]	3[n] 70[r]	180[o,p]
UPt_3	none	$<$ln4 at 30 K[s]		
U_2Zn_{17}	Th_2Zn_{17}	ln10 at 70 K[k]	$\sim$50[k]	not seen[t]
UCd_{11}	none	$\sim$ln4 at 13 K[u]		

[a]Reference 35.
[b]Reference 10.
[c]Reference 52.
[d]Reference 53.
[e]Reference 36.
[f]Reference 54.
[g]Reference 55.
[h]Reference 56.
[i]Reference 20.
[j]Reference 57.
[k]This work.

[l]Reference 58.
[m]Reference 59.
[n]Reference 60.
[o]Reference 3.
[p]Reference 61.
[q]Reference 62.
[r]Reference 63.
[s]Reference 64.
[t]Reference 34.
[u]Reference 63.

this interpretation. In particular, the specific-heat peak, supposedly due to a CEF, is smaller in magnitude and wider in temperature than predicted.

In contrast, for $CeCu_2Si_2$ the peak is not connected with any known CEF level splitting. Furthermore, in this case the measured peak is larger in magnitude and narrower than a CEF level would predict, i.e., just the reverse of $CeAl_3$. We will return to the problem of detecting CEF levels from the specific heat after discussing the new data presented in Sec. IV.

"*Kondo regime*" $(T \sim T_0)$. For temperatures comparable to the single-impurity temperature T_0 the conduction electrons screen the impurity spin associated with the lowest CEF level.[11] From the theoretical work on the single-impurity problem, we know that in the f-projected density of states there is a peak at $\sim T_0$ above the Fermi level. This peak, which is temperature dependent, gives rise to a peak in the specific heat at $T \sim T_0$. Such peaks have been seen in many dilute-impurity systems and may also be present in heavy-electron materials.

The effects of crystal fields on the Kondo-based peak of the specific heat have been studied by Desgranges and Rasul[12,13] for the cases of two doublets and three doublets (equally spaced). As the CEF increases, splitting a fourfold level into two doublets, a shoulder develops in the specific heat, and eventually two peaks form on either side of the original fourfold level. The lower peak can be associated with the Kondo effect involving the low-lying doublet, and the upper with a Schottky anomaly. They obtain similar results for the sixfold case splitting into three doublets.

The main features of the resulting specific heats are that, except for $\Delta \lesssim T_0$, the main peak is shifted to lower in temperature, and its height is decreased from the isolated Schottky peak. The most prominent feature is the height decrease, and for several compounds ($CeAl_3$, $CeAl_2$, $CeCu_6$) there is qualitative agreement in this aspect. The shift in peak position becomes less pronounced as Δ becomes smaller, and, in fact, becomes positive for $\Delta \lesssim 0.5T_0$ due to the overlap of the Kondo peak. However, even using the shape of the specific-heat curve to estimate Δ/T_0 there is not good agreement. The experimental peak is at a higher temperature for $CeAl_3$ ($\Delta/T_0 \simeq 1.5$), $CeAl_2$ ($\Delta/T_0 \simeq 1$–2), and $CeCu_2Si_2$ ($\Delta/T_0 \simeq 2$). The position of the peak is correct for $CeRu_2Si_2$, but the peak height is much too high. $CeCu_6$ seems the only material with qualitative agreement in position and a peak height at least not higher than the Schottky.

Desgranges and Rasul have applied[8] their model to $(LaCe)B_6$, assuming a crystal field splitting of $\Delta/T_0 = 0.7$. They get good agreement with the experimental data at low temperatures ($T < 0.2$ K), but the peak is somewhat large in magnitude and above 0.5K the calculated curve drops off too sharply. Although better than that of no crystal-field splitting, the fit is not great, and, in particular, would seem to give too low a value for the limiting high-temperature entropy. Desgranges and Rasul also fit[13] their three-doublet calculations to the case of $CeAl_3$. However, the experimental data they cite shows spurious

magnetic phase transitions due to impurities in the sample. These extraneous peaks occur right at the temperature regime separating Kondo- from Schottky-dominated behavior, making comparison to theory difficult.

In summary, a shoulder-plus-peak structure is indeed seen in Ce compounds, and in this respect there is qualitative agreement with the theory[8,12,13] of Desgranges *et al.* However, the decrease in peak height from a straight Schottky behavior is only seen in a few compounds, and for these the peak position is at too high a temperature. This may be accounted for by uncertainty in the crystal-field splitting. Using the shape of the specific heat to find Δ/T_0 gives qualitatively correct values of T_0. As it stands, the calculations are only applicable to Ce compounds.

Fermi-liquid solid regime $(T \ll T_F^*, T_0)$. At very low temperatures, interactions between the f atoms become noticeable. The clearest evidence for this is the crossover in the electrical resistivity with decreasing temperature from a rising (Kondo-like) resistivity to a dropping resistivity as in ordinary metals. (We expect the resistivity to show interaction effects at a higher temperature than equilibrium properties due to the fact that the resistivity of Bloch states must be zero at zero temperature. This argument hinges on the fact that the resistivity is very sensitive to impurities while the specific heat, susceptibility, and other transport properties are not. Accordingly, a small amount of impurity will destroy the Bloch-coherence at low temperatures and, we expect, strongly effect the resistivity at higher temperatures. In fact, the resistivity typically turns over at T_0.) The principal difference from ordinary metals is that the T^2 coefficient A in $\rho(T) = \rho(0) + AT^2$ is many orders of magnitude[14] larger than in metals such as Pd. Roughly, it appears that $A \propto (1/T_F^*)^2$.

This T^2 dependence suggests a Fermi-liquid description. But if something that simple were true, then it is hard to understand the additional temperature-dependent structure that is seen in various properties for $T/T_F^* < 10^{-2}$. Figure 1 shows a compilation of equilibrium and transport data for $CeAl_3$. A simple Fermi-liquid picture would require that the specific heat, the electronic thermal conductivity, and the thermopower be linear in temperature while the resisitivity would go as T^2. According to Fig. 1, this state of affairs occurs for temperatures below 0.07 K, i.e., for $T/T_F^* < 0.003$. A Fermi-liquid theory of such limited range is not very useful.

What one would want is a theory capable of reproducing the lowest temperature bump in $C(T)/T$ (Ref. 15) and showing the turndown in the thermopower, not to mention the temperature-dependent structure in the Hall effect.[16] A possible model would involve a renormalized band structure of width T_F^* in which a phenomenological interaction between f electrons is added to a conventional band-structure calculation for the remaining electrons. There are preliminary calculations[17,18] for such a model; they show Van Hove–like singularities in the narrow quasiparticle f band crossing the Fermi level. Whether the renormalized band structure, together with model scattering rates, could explain the low-temperature data is still to be tested.
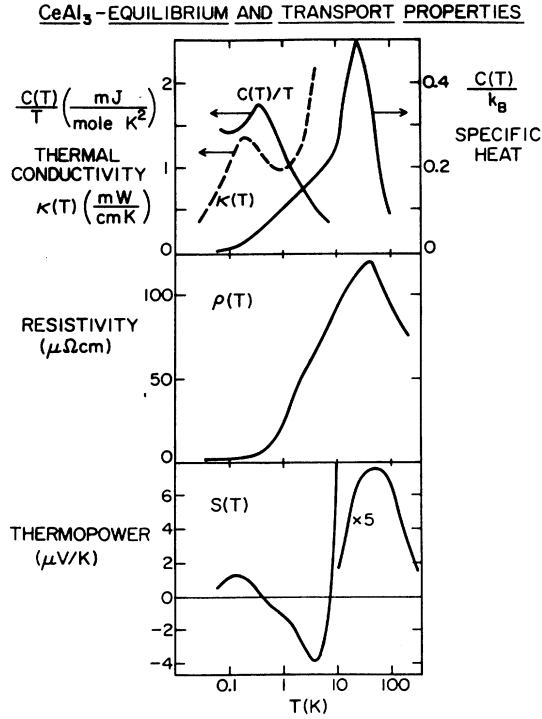
CeAl₃ -EQUILIBRIUM AND TRANSPORT PROPERTIES

FIG. 1. Equilibrium and transport properties for CeAl₃ vs log₁₀T from 0.1 to 300 K (Ref. 37). The upper panel contrasts the thermal conductivity (Ref. 38) with the specific heat (Ref. 35) and ΔC(T)/T (both relative to LaAl₃). The second panel shows the resistivity (Ref. 39); note that the crossover into the Fermi-liquid solid regimes occurs quite high (T ~ T₀), while the specific heat in the same temperature range has a Schottky anomaly. The thermopower S(T) (Ref. 40) (lower panel) illustrates a standard Kondo behavior at high temperature and then a complex behavior at low temperature that may reflect competing interactions in the Fermi-liquid solid.

Finally we should point out (1) the considerable sensitivity of the low-temperature properties to pressure [i.e., a peak in $C(T)/T$ for CeAl₃ (Ref. 19)] and magnetic field [e.g., a peak in $C(T)/T$ for CeCu₆ (Ref. 20) and for CeAl₃ (Ref. 21)], (2) the sensitivity of the Hall effect (to impurities) in several heavy-electron metals (UPt₃, UAl₂, CeAl₃, and CeRu₂Si₂),[16] and (3) the extraordinary influence of impurities [e.g., Th in UBe₁₃ (Ref. 22) produces a second transition, whereas Cu in U₂Zn₁₇ (Ref. 23) and Ni in UCu₅ (Ref. 24) remove the magnetic transition].

While we have called these three regimes the "standard model" of heavy electrons (cf. Table IV), we would stress that no theory connects them. In this paper we confine ourselves largely to the high-temperature regime. We present new data for U₂Zn₁₇ and CeCu₆ and examine them for evidence of crystal fields and independent spins, i.e., are the data consistent with the standard model at high temperature?

III. EXPERIMENTAL METHODS

Specific heat measurements were performed using the standard heat-pulse method in a ⁴He cryostat.[25] The samples were produced at the Los Alamos National Laboratory. The masses of the three samples of U₂Zn₁₇ were 0.7750, 0.2933, and 0.2555 g, respectively, those of the Th₂Zn₁₇ samples were 1.6998 and 0.5300 g, and the CeCu₆ and LaCu₆ were 0.5805 g and 1.486 g, respectively. The third U₂Zn₁₇ sample had been measured previously.[26] The thermal sample-to-bath time constants ranged from 60 s at 2.5 K for the smaller Th₂Zn₁₇ sample, to 2000 s at 80 K for the larger samples. The internal time constants were much less than 1 s at 2.5 K and about 10 s at 80 K for the larger samples. Below 10 K, the heat capacity of the addenda was much less than 1% of the total heat capacity, but by 70 K the addenda accounted for almost 10% of the total heat capacity for the smallest samples. This, however, is not a problem because the addenda heat capacity is known to better than 5%.[25] The temperature of the sample was measured with a chip (about 0.5 mg) of a 1200-Ω Allen-Bradley resistor, calibrated on every cooldown against a germanium-resistance thermometer below 40 K, and against a platinum-resistance thermometer above 20 K. The germanium- and platinum-resistance thermometers agree to within 0.1% in the range of overlap; both were calibrated against a Rh:Fe-resistance thermometer with a calibration traceable to the National Physical Laboratory in England.

The dominant source of error in the measurement is the internal time constant, which also includes a sample-holder–to–sample time constant. At the highest temperatures of measurement (80 K), during the first few internal time constants (a few tens of seconds, as stated above) the sample holder is somewhat hotter than the sample. This causes the heat flow into the sample to be overestimated, as some heat is lost to radiation and conduction through the weak link before it reaches the sample. We calculate that this error will cause about a 1% overestimation of the measured heat capacity of the sample. This error, though, should scale with the heat capacity of the sample, and to a large extent will cancel in the subtraction of the specific heat of the non-heavy-electron counterpart. The accuracy of our measurements has been checked by measuring a sample of pure copper, and can also be seen in the reproducibility of the specific heat of the samples of U₂Zn₁₇ of different masses, and of Th₂Zn₁₇ of different masses.

At higher temperature the heat capacity of the phonons exceeds that of the electrons, and the subtraction of the lattice becomes more susceptible to error. For U₂Zn₁₇, at about 17 K the electronic specific heat is half the total, at 36 K it is one-sixth, and at 50 K it is one-tenth. Thus, after subtracting the lattice contribution, assuming a random 1% error, there is about a 10% uncertainty in the electronic specific heat at 50 K, and about a 20% uncertainty at 70 K. The uncertainty is similar for CeCu₆.

A separate determination of the phonon density of states would certainly be a useful check on the lattice-subtraction procedure used here. Unfortunately, there is

very little neutron-scattering data at all, and what there is has limited usefulness. In the case of UBe_{13} there have been two measurements. One triple-axis-spectrometer measurement[27] of the lower-lying acoustic modes at room temperature and at 10 K concludes that they are consistent with scaling from pure beryllium (by replacing the Be mass with the mass of $BeU_{1/13}$). Time-of-flight measurements[28,29] at 300 K and 8 K were also able to determine an optic mode at 13 meV. The lattice-vibration spectra of $ThBe_{13}$ and UBe_{13} are very similar, suggesting that using $ThBe_{13}$ as a guide to all the phonons in UBe_{13} is reasonable. Finally, a time-of-flight measurement[30] is available for UPt_3 at 300 K and preliminary measurements at 8 K suggest no dramatic changes in the acoustic and optic modes.

IV. DESCRIPTION OF RESULTS

Shown in Fig. 2 are the measured specific heats for U_2Zn_{17}, Th_2Zn_{17}, $CeCu_6$, and $LaCu_6$, plotted log-log versus temperature. The dominance of the electronic specific heat at the lower temperatures and of the phonon specific heat at high temperatures is clear; the non-heavy-electron counterpart specific heat is orders of magnitude smaller than the heavy-electron specific heat at the lowest temperatures, but only about 10% smaller at 70 K. We have plotted in Fig. 3 the difference, $\Delta C(T)$, between the total specific heat and the phonon specific heat (as estimated using the specific heat of the non-heavy-electron counterparts), divided by the temperature T. The specific heats of U_2Zn_{17} and Th_2Zn_{17} have been measured before[26] up to 17 K. The specific heat of $CeCu_6$ has been measured twice before, by Steward, Fisk and Wire,[31] and by Fujita, Satoh, Onuki, and Komatsubara.[20] Several observations can be made immediately.

For U_2Zn_{17}, the anti-ferromagnetic transition at 9.7 K is very sharp. The specific heat at the transition follows a typical λ curve with a smooth high-temperature tail. There does appear to be an entropy balance at the transition; that is, the total entropy through the transition, i.e., the area under the $C(T)/T$ curve, does appear to be the same as the area under the curve generated by extra-
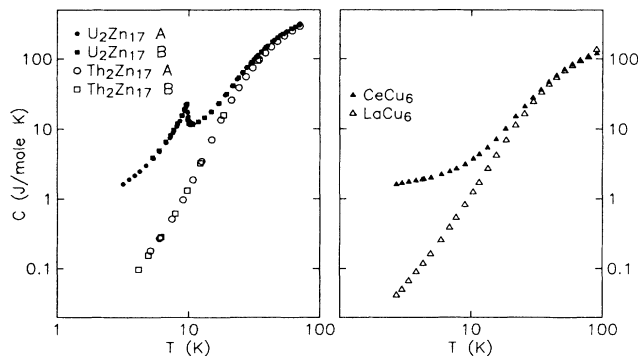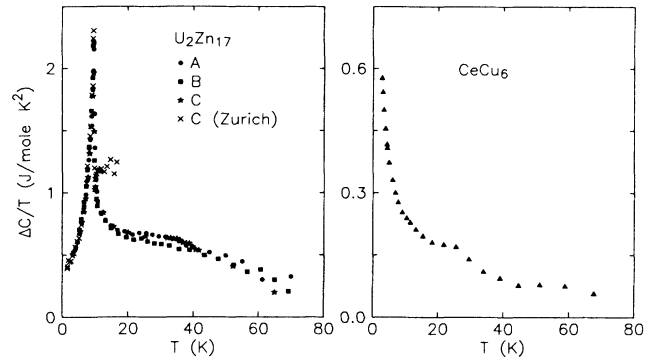

FIG. 3. $\Delta C(T)/T$ for U_2Zn_{17} and $CeCu_6$. Sample C was also measured by Ott et al. (Ref. 26), shown as the ×'s, and is discussed in the text.

polating $C(T)/T$ to $T=0$ from above the transition. The anomalously high electronic specific heat extends to the highest temperature of measurement, 70 K. Below the transition at 9.7 K, our measurement of the specific heat of U_2Zn_{17} and that reported earlier[26] are indistinguishable. However, the electronic specific heat above 9.7 K is quite different, both in the shape of the transition, and in magnitude, by a factor of about 50%. The previous researchers also found that the transition had absolutely no high-temperature tail, and entropy balance was not satisfied. Because of this discrepancy, another sample (sample B) was produced at Los Alamos, and measured at Cornell, and its specific heat was found to agree with that of our first sample (A). In order to determine whether the disagreement between our measurements and those reported earlier was due to a strange sample dependence or to experimental error, we asked H. R. Ott for his sample, which he kindly supplied. Our measurements on his sample agreed with our other samples, and we conclude there are no unusual sample dependences between samples prepared more than 1 yr apart. The sample has been sent back to Zurich, where it will be remeasured as a final check.

Below about 20 K, there are no detectable sample dependences at all. Between 20K and 50 K, though, there is a small difference in the electronic specific heat of the samples which we cannot attribute to measurement uncertainties. Above 50 K the data start to show the scatter that is associated with the uncertain subtraction of the phonon specific heat. Differences above 50 K thus cannot be attributed to sample dependences, but may simply be due to small random errors in the data.

The $CeCu_6$ specific heat clearly displays the large γ characteristic of heavy-electron materials. The $CeCu_6$ electronic specific heat shows no sharp transitions as it is neither superconducting nor magnetic. There is, however, a broad bump in C_{el}/T centered near 26 K, and another near 60 K. The latter, though, is not experimentally significant, as it is smaller than the experimental uncertainty due to the lattice subtraction. The broad bump at 20 K will be discussed later and is attributed to the CEF levels. The measurement of $CeCu_6$ by Fujita,

FIG. 2. Specific-heat data for U_2Zn_{17}, Th_2Zn_{17}, $CeCu_6$, and $LaCu_6$. Note that these data and those in subsequent figures are given per mole of U, Th, Ce, or La, respectively, and not per mole of formula unit (e.g., not per mole of U_2Zn_{17}).

et al.[20] extended to 70 K, high enough to deduce the electronic entropy, but above about 50 K the specific heat showed an anomalous upturn, as it would if radiation difficulties in the measurement were present. Our measurements do not show this upturn at high temperature, but do agree with the previous measurements below 40 K.[32] Our electronic specific heat agrees reasonably well with that reported by Fujita *et al.*, even in the temperature range of their anomalous upturn. This supports our explanation that their upturn was systematic and thus, to a large extent, cancelled upon lattice subtraction.

The subtraction of the lattice heat capacity using the non-heavy-electron counterpart leads to uncertainty in the electronic specific heat, not only for the above experimental reasons, but for more fundamental reasons as well. It is not true that the density, elastic constants, and lattice constants are identical for the counterpart material. See Table V for the densities, crystal structures, and lattice constants of several heavy-electron materials and their nonheavy counterparts. For the case of U_2Zn_{17} the crystal structure is the Th_2Zn_{17} structure, but the lattice constants are between 0.5% and 1.5% smaller for the Th_2Zn_{17}. The density of U_2Zn_{17} is about 2% higher than that of Th_2Zn_{17}. We do not know the difference in the elastic constants. The Debye formalism for the lattice specific heat predicts that the low-temperature molar specific heat has the following dependence:

$$C_V \sim V \left[\frac{\rho}{E} \right]^{3/2} .$$

Here, ρ is the density, V is the volume of a unit cell, and E represents an average elastic constant. Thus, assuming the elastic constants for the two compounds are the same (not true, but we do not know the elastic constants), we estimate from the lattice and density differences that the U_2Zn_{17} should have about a 1% higher Debye lattice specific heat than the Th_2Zn_{17}. For the case of $CeCu_6$, the crystal structure is the same as $LaCu_6$ (orthorhombic), and the lattice constants of $CeCu_6$ are about 0.7%–0.9% smaller than those of $LaCu_6$. The density of $CeCu_6$ is about 2.5% higher than the density of $LaCu_6$. Thus, by using the specific heat of $LaCu_6$, we have slightly underestimated the Debye lattice specific heat of $CeCu_6$ by about 1.4%, based only on the differences in the density and the lattice constants. Again, for $CeCu_6$ and $LaCu_6$ we do not have elastic constants so we cannot estimate their effects.

An even more fundamental objection to the subtraction is that the heavy-electron character could have a non-negligible effect on the phonons directly, so that the lattice specific heat would be substantially different from

TABLE V. Crystal structure, density, and lattice information for some heavy-electron and non-heavy-electron pairs.

Compound	Crystal structure	Density (g/cm^3)	Lattice parameters a (Å)	b (Å)	c (Å)	Volume (Å^3)	Units (per cell)
CeAl$_3$[a]	Ni$_3$Sn (hex)	2.521	8.54		4.61	291.2	2
LaAl$_3$[a]	Ni$_3$Sn (hex)	2.438	8.66		4.61	299.4	2
CeCu$_2$Si$_2$[b]	ThCr$_2$Si$_2$ (tet)	6.415	4.10		9.96	167.4	2
LaCu$_2$Si$_2$[b]	ThCr$_2$Si$_2$ (tet)	6.286	4.143		9.914	170.2	2
CeCu$_6$[c]	CeCu$_6$ (orth)	8.23	8.112	5.102	10.162	420.8	4
LaCu$_6$[d]	CeCu$_6$ (orth)	8.03	8.165	5.148	10.23	430.0	4
UBe$_{13}$[e]	(cub)	4.38	10.25			1076.9	8
ThBe$_{13}$[e]	(cub)	4.11	10.41			1128.1	8
UPt$_3$[f]	Ni$_3$Sn (hex)	19.53	5.75		4.89	140.0	2
ThPt$_3$	not the same structure						
U$_2$Zn$_{17}$[g]	Th$_2$Zn$_{17}$ (rhom)[h]	8.60	8.983		13.16	919.6	3
Th$_2$Zn$_{17}$[g]	Th$_2$Zn$_{17}$ (rhom)[h]	8.34	9.060		13.234	940.8	3
CeB$_6$[i]	CaB$_6$ (cub)	4.793	4.141			71.01	1
LaB$_6$[i]	CaB$_6$ (cub)	4.709	4.157			71.84	1
UCu$_5$[j]	UNi$_5$ (cub)	10.60	7.036			348.3	4
THCu$_5$	not the same structure						

[a]Reference 65.
[b]Reference 66.
[c]Reference 67.
[d]Reference 68.
[e]Reference 69.
[f]Reference 70.
[g]Reference 71.
[h]Crystal parameters, volume of cell, and formula units per unit cell are reported for the hexagonal structure formed of two rhombohedral unit cells.
[i]Reference 72.
[j]Reference 73.

the Debye prediction. This we cannot estimate without a direct measurement of the density of states of the phonons. However, at temperatures around 10 K, the phonon contribution from the non-heavy-electron material is almost an order of magnitude smaller than the total specific heat of the heavy-fermion compound. Even if substantially altered, the heavy-electron-phonon part will probably still be quite small compared to the electronic contributions, and will not affect the specific heat. At higher temperatures the phonon contribution becomes substantial, as does the (subtraction-based) error, and the assumption here is that the purely electronic specific heat will still be within the range of uncertainty.

Plotted in Fig. 4 are the normalized integrated entropy and associated number of degrees of freedom (N_{dof}) for U_2Zn_{17} and $CeCu_6$. The entropy is simply the area under the curves in Fig. 3, and the number of degrees of freedom is then $\exp(S/R)$, where R is the molar gas constant. Error in the lattice specific heat subtracted will clearly have an effect on the integrated entropy of the electrons. For U_2Zn_{17}, roughly, a 1% underestimation of the lattice specific heat to be subtracted will cause the calculated number of degrees of freedom to be overestimated by 1 at 70 K. Thus, if the lattice specific heat of U_2Zn_{17} is larger than Th_2Zn_{17} by 1%, as we have estimated above from the density and lattice constants, we should report the number of degrees of freedom at 70 K to be 11 and not 12. For $CeCu_6$, roughly, a 1% underestimation of the lattice specific heat will cause the calculated number of degrees of freedom to be overestimated by about $\frac{1}{3}$ of a degree of freedom at 80 K. Therefore, if the $CeCu_6$ does have a 1.4% larger lattice specific heat than $LaCu_6$, as we have estimated from the differences in the densities and lattice constants, then we should report the number of degrees of freedom as 3.9 and not 4.3 at 80 K. This analysis is meant only as a guide to the size of the uncertainty. Without the precise phonon density of states, or at least the elastic constants, a more precise analysis cannot be done.
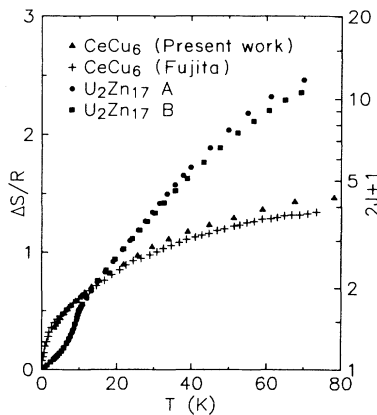

FIG. 4. The entropy difference $\Delta S(T)$ of U_2Zn_{17} relative to Th_2Zn_{17}, and $CeCu_6$ relative to $LaCu_6$, vs T. The right-hand vertical axis, $\exp[\Delta S(T)/R]$, shows the number of degrees of freedom associated with the f electrons. Previous measurements (Ref. 20) are shown as the crosses and are discussed in the text.

V. DISCUSSION

We want to focus on two aspects of the high-temperature $(T > T_0)$ part of the standard model: (1) how many degrees of freedom are seen and (2) what the evidence for CEF levels is. The first provides a crude test of the standard model and the second a more sophisticated one. In the case of both Ce and U compounds we assume we are in the LS-coupling regime; this is probably a reasonably good approximation since the spin-orbit splitting is 0.2 eV for Ce and ~0.8 eV for U. (The relevant comparison is the ratio of the spin-orbit coupling to the F^2 integral deduced from the spectra or Hartree-Fock calculations. These ratios are 0.001 and 0.086 for Ce and U, respectively.) Nonetheless, since it will turn out that we see too many degrees of freedom for U, there is a possibility that these materials are in an intermediate-coupling regime in which the number of degrees of freedom might be larger than $2J + 1$.

A. Entropy $\Delta S(T)$

Figure 4 shows the entropy of U_2Zn_{17} relative to Th_2Zn_{17} and of $CeCu_6$ relative to $LaCu_6$. Under the assumption that the only contribution to this difference is due to the f electrons, then we would expect to recover the full number of degrees of freedom associated with the f electrons at a temperature large compared to T_0 and Δ.

$CeCu_6$: The evidence for the standard model is fairly clear for $CeCu_6$. $\Delta S/R$ exceeds ln4 near 80 K, which is larger than $T_0 \sim 25$ K and, as discussed next, smaller than the estimated CEF splittings. The peak in $\Delta C(T)$ occurs at 26, suggesting a CEF level at 60 K. For an f^1 configuration, $J = \frac{5}{2}$, i.e., the number of degrees of freedom is 6. Thus, $\Delta S \geq$ ln4 is consistent with the standard model; a result which is also the case for the data of $CeAl_3$ and $CeCu_2Si_2$ (cf. Table IV).

U_2Zn_{17}: The situation is much less clear for U_2Zn_{17}. At 70 K, $\Delta S/R$ exceeds ln10 per U atom. This very large number poses two problems for the standard model:

(1) The first problem is that a value of ln10 is already the largest number of degrees of freedom we can readily imagine for independent U atoms. The two most likely configurations for U are f^2 with a ground state $J = 4$ and f^3 with $J = \frac{9}{2}$. These two ground-state terms have 9 and 10 degrees of freedom, respectively. Nonetheless, $\Delta S/R > $ ln10 at 70 K. This fact has caused us to reexamine how the lattice and non-f-electron contributions should be subtracted. Still, as indicated in Section IV, there seems to be no obvious way to substantially reduce $\Delta C(T)$ or $\Delta S(T)$.

(2) The second problem in understanding the large value of $\Delta S(T)$ is that it occurs at too low a temperature compared to T_0 and Δ. Both of these energy scales are difficult to estimate. χ^{-1} is only roughly linear in T between $0.6T_0$ and $3T_0$. Accordingly $T_0 = 65$ K has an uncertainty, but it is unlikely to be smaller by an order of magnitude.[33] Any observable CEF levels must be larger than 350 K according to neutron-scattering exper-

iments.[34] If we use the peak temperature in $\Delta C(T)$, then $\Delta \sim 120$ K, which is much larger than the temperature (below 70 K) at which $\ln 10$ is exceeded. Moreover, as noted earlier, some CEF fits involve two sets of four levels lying above the doublet. In this case Δ is a factor of 3.0 larger than the peak temperature. In summary, even if some CEF levels are found in the (50–100)-K range, there is still too much entropy at too low a temperature for the "standard model" to apply.

B. Specific heat $\Delta C(T)$

The specific heat provides a more detailed test of the standard model than $\Delta S(T)$ provided. As a point of departure we note that the previous attempts to fit the electronic specific heats of CeAl$_3$ (Ref. 35), CeCu$_{22}$Si$_2$ (Ref. 36), CeCu$_6$ (Ref. 20) [for $\Delta S(T)$ only], and UBe$_{13}$ (Ref. 3) are based on the addition of two peaked structures, one due to the Kondo specific heat of the low-lying doublet and the second due to the Schottky contribution of the next CEF doublet. This approach is a little simplistic since the two contributions do not add so simply. But, in the absence of a detailed theory it is a reasonable starting point.

Fitting procedures. We have used both a CEF-level scheme and, in the case of U$_2$Zn$_{17}$, a density-of-states scheme to fit the measured specific heat. In the crystal-electric-field scheme we use the following pattern of levels: for f^1 in CeCu$_6$ we have three doublets whose two spacings can be varied to achieve a fit. In U$_2$Zn$_{17}$ there is a choice of two configurations, both of which we have tried: for f^2 there are three doublets and three singlets, while for f^3 there are five doublets. Then one just uses a generalization of the Schottky specific-heat formula (see Introduction) and adjusts the splittings for the best fit. While the whole process could be computerized, the process goes quite quickly on a trial and error basis.

U$_2$Zn$_{17}$: Here we also tried fitting the specific heat with a distribution of levels described by a density of states. In particular, we approximate the density of states of the many-body conduction band with a sum of several Lorentzians. The band has a total weight of nine or ten states per U atom (for f^2 or f^3, respectively), but the (temperature-dependent) chemical potential is set to give a constant filling of 1. That is, in analogy with the crystal-field levels, each of which corresponds to a two- or three-f-electron state, the many-body band in its ground state has a unique energy level associated with a many-body (two- or three-electron) state. But in contrast, while the CEF scheme has Boltzmann statistics, the band scheme has Fermi statistics.

We will show that the entropy of this strongly correlated density-of-states model is very similar to what we get for the CEF model, but with a small addition (≤ 1) which increases the entropy sufficiently to account for the experimentally large entropies. For simplicity we consider the case of f^3 where the total degeneracy N is 10. The entropy per U atom has the form

$$\frac{\Delta S}{R} = -N \sum_\epsilon [f_\epsilon \ln(f_\epsilon) + (1 - f_\epsilon) \ln(1 - f_\epsilon)] \ ,$$

where $f_\epsilon = (\exp\{[\epsilon - \mu(T)]/k_B T\} + 1)^{-1}$. The chemical potential $\mu(T)$ is adjusted to maintain the fractional occupancy of the band, which in the case of f^3 is $1/N = 1/10$ (and would be $\frac{1}{9}$ for f^2). At temperatures large compared to the bandwidth, $(\exp[-\mu(T)/k_B T] + 1)^{-1}$ is 0.1. From this we can deduce that the entropy at high temperatures reduces to $\ln 10 + 9 \ln(\frac{10}{9})$ [i.e. $\sim \ln(26)$], which is somewhat larger (by 0.9) than the CEF model, which gives $\ln 10$ for the f^3 case. In general, for large N the degeneracy for the Fermi statistics model is a factor of e more than that of CEF (Boltzmann).

We should stress that this is a very unusual band model, one in which there are very strong correlations. In fact, there is no actual model which contains the features of "quasi-itinerant" f^3 states. On the contrary, the phrase "band model" usually implies a set of largely independent electrons. Such a model leads to large entropies. For example, two independent electrons in a band with six states (as if a crystal-field splitting had moved the rest of band to very high energy) would have a high-temperature entropy of $2 \ln 3 + 4 \ln(\frac{3}{2}) \sim \ln(46)$ $= 3.8$, whereas if the band held ten electrons the entropy would be $\sim \ln(149)$, which is almost twice the largest entropy seen so far (5.0). It is numbers such as these that makes it difficult to accept any over-simple renormalized band model. As the temperature increases, the band must broaden rapidly to prevent the entropy from reaching its "ionized" atom limit at room temperature.

CeCu$_6$: From the CeCu$_6$ specific-heat data we can only deduce the possible presence of one higher-lying doublet. (The data cannot be used to extract a second crystal-field level.) Figure 5 [$\Delta C(T)$ versus $\ln T$] shows a reasonable fit to the overall pattern of the data, but the theory is higher and narrower than the data. We could improve the fit by broadening the high-lying CEF levels, but defer using that approach until analyzing the U$_2$Zn$_{17}$ data.
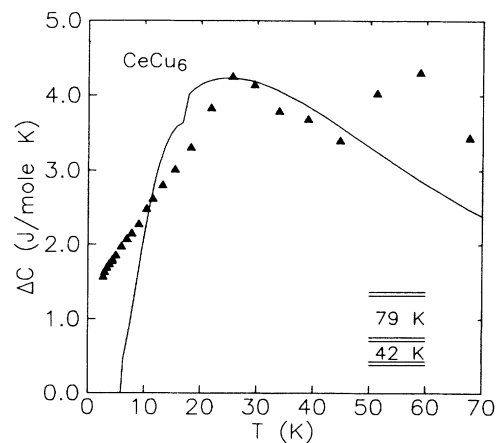

FIG. 5. Fit of the CEF model (solid line) to $\Delta C(T)$ data (triangles) for CeCu$_6$ relative to LaCu$_6$. The experimental uncertainty at higher temperatures (> 30 K) is about the same size as the scatter in the points. The inset shows the level scheme of three doublets separated by 42 and 79 K.
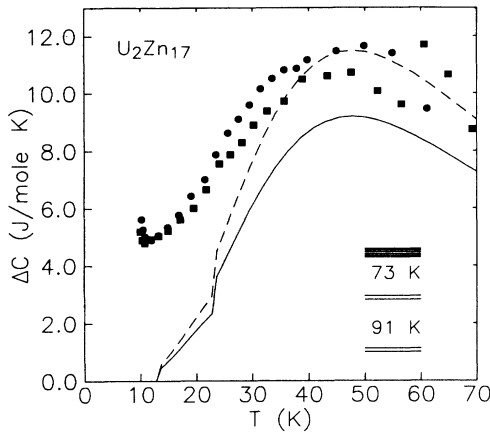
FIG. 6. Fit of CEF model (solid line) to the $\Delta C(T)$ data (circles, sample A; squares, sample B) for U_2Zn_{17} relative to Th_2Zn_{17}. The inset shows the level scheme for f^3 with two doublets separated by 91 K and the remaining six levels some 73 K higher. Note that to get anything approaching agreement we have scaled the calculation by 1.25 to produce the dashed curve.

FIG. 8. Fit of density of states model (solid line) to $\Delta C(T)$ (triangles) for $CeCu_6$ relative to $LaCu_6$. The dashed lines indicate the sensitivity of the fit to only $\pm 4\%$ changes in the width of the lower peak in the model density of states. The inset shows the density-of-states model used in the fit, scaled by 100 as in Fig. 7. The peaks are more narrow than for U_2Zn_{17}, but are separated again by ~ 100 K (91 K). The lower peak is almost directly at the Fermi level.

U_2Zn_{17}: In Fig. 6 we see the fit for a CEF pattern for U_2Zn_{17}. Note that in order to give any agreement whatsoever we must scale the calculated curve (solid line) by a factor of 1.5 (dotted line). This illustrates the point made in the preceding section: there are more degrees of freedom in the data than are consistent with a CEF model.

In contrast, Fig. 7 shows the good fit that results from using the band density of states shown in the inset of the figure. Two Lorentzian peaks were found necessary to fit the data. Similarly, Fig. 8 shows the fit for $CeCu_6$ ob-
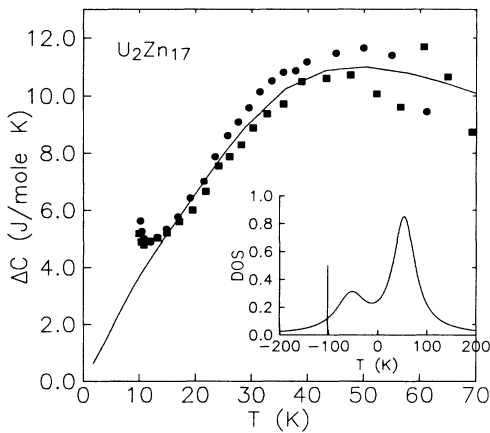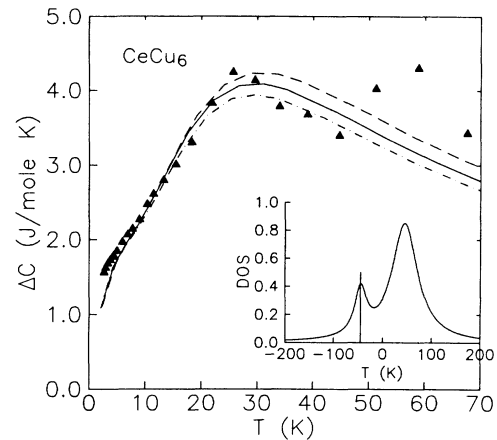
tained by using a two-peak structure in the density of states (inset). (Note that the experimental uncertainty for temperatures greater than about 30 K is roughly the size of the scatter of the points.) For both materials, a second peak is necessary to account for the low-temperature shoulder seen in the data. These densities of states should be viewed as only illustrative since any such renormalized band would vary strongly as the temperature increases through T_0. We include them merely to stress the difficulties faced by any theory attempting to explain the full temperature dependence of any property.

VI. CONCLUSIONS

The specific heats and entropies of the heavy-electron metals at high temperatures $(T \geq T_0)$ suggest the following.

(1) In Ce-based materials a crystal-electric-field-split f^1 state is consistent with the data. However, the lack of any solid observation of a CEF level in the needed range, less than 100 K, (with the exceptions of $CeAl_2$ and $CeAl_3$), suggests that if there are CEF levels present, the effect of the Kondo coupling is so strong as to make them unobservable by neutron scattering. The low-temperature shoulder in the data further indicates a broadening of the levels.

(2) In U-based materials no CEF levels have been directly observed, although in two materials (UBe_{13} and U_2Zn_{17}) the specific heat is roughly consistent with their existence. However, especially in the case of U_2Zn_{17}, the entropy is too large for traditional CEF even up to degeneracy 10. A model band density of states with a splitting of ~ 100 K gives a good fit to the data and suggests that the additional entropy may be a sign of

FIG. 7. Fit of density of states model (solid line) to $\Delta C(T)$ (circles, sample A; squares, sample B) for U_2Zn_{17} relative to Th_2Zn_{17}. The inset shows the density-of-states model used in the fit; note the 108-K splitting between the two peaks. The density of states, in K^{-1}, has been scaled by a factor of 100. The vertical line marks the Fermi energy.

greater correlations in these materials, with perhaps an effective hopping between sites giving greater degeneracy and accounting for the different statistics. Clearly, a more ambitious study of crystal-electric-field levels is warranted, especially for U compounds.

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