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$CeCu_4Al$ and $CeCu_2Zn_2Al$: Very-heavy-fermion systems in high magnetic fields

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$CeCu_4Al$ and $CeCu_2Zn_2Al$ are heavy-fermion systems with extremely enhanced C/T (the specific heat divided by temperature) values of 2.3 and 1.8 J/K^2, respectively, as $T \rightarrow 0$ K. The field dependence of the low-temperature specific heat is also extreme: At 11 T, C of $CeCu_4Al$ is reduced by more than a factor of 5 and at 12.5 T, C of $CeCu_2Zn_2Al$ is suppressed by a factor of about 7. Magnetic-field-caused changes of the specific heat of $CeCu_4Al$ are consistent with a single-ion Kondo model. Magnetic correlations are at least partially responsible for the enhanced low-temperature specific heat of $CeCu_2Zn_2Al$.

The proximity of heavy-fermion and magnetic ground states has been recognized for some time already.[1,2] Many heavy-fermion systems order antiferromagnetically at low temperatures of the order of 1 K. In some systems, e.g., UPt_3, $CeCu_2Si_2$, or $CeAl_3$, magnetic order has very little effect on the low-temperature thermodynamic properties,[3] such as specific heat (C). The linear, low-temperature coefficient of the specific heat, γ (also called the Sommerfeld coefficient), can truly be electronic in nature (as proved by the large discontinuities in the specific heats of UPt_3 and $CeCu_2Si_2$ at their superconducting transition temperatures) and reaches spectacular values of order 1000 mJ/K^2 mol. The ordered moments associated with these itinerant heavy quasiparticles are extremely small; a value of the order of $0.02\mu_B$ has recently been reported[4] for UPt_3.

Another group of systems, e.g., $CeAl_2$, $CeCu_2$, CeB_6, CePdIn, order magnetically with large local moments of the order of $1\mu_B$ before the heavy-fermion ground state is fully established.[5] In this case, the enhanced high-temperature coefficient γ (at temperatures much larger than the ordering temperature), of the order of 100 mJ/K^2 mol, is reduced by the ordering to much lower values as $T \rightarrow 0$ K.

There is, however, a class of heavy-fermion systems that, although they do not undergo magnetic ordering down to the lowest temperatures, have large, low-temperature specific heats clearly not due to heavy electrons but rather associated with magnetic-entropy or crystal-field effects. Examples are $CeCu_{6.5}Al_{6.5}$ (Ref. 6) and $CePd_3B_{0.3}$ (Ref. 7) in which disorder on non-Ce sites causes a variation in the Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions between Ce ions that prevent magnetic ordering. Instead, spin-glass behavior with low freezing temperatures is observed.

One could also speculate on intermediate cases when the low-temperature enhancement of the specific heat arises from a combination of the formation of a heavy-fermion state and an onset of a magnetic phase transition as has been suggested for $CuCeIn_2$ (Ref. 8), and as might be the case for $CeCu_4Ga$.[9]

$CeCu_4Ga$ has recently become a subject of very intense studies due to its extremely large C/T values, of the order of 2 J/K^{2}mol at $T \rightarrow 0$ K. The nature of this enhancement (magnetic versus heavy fermions) still remains a subject of controversy.[9-11]

Two more isostructural systems have lately been synthesized:[12] ($CeCu_4Al$ and $CeCu_2Zn_2Al$) that do not order magnetically down to 150 mK which have C/T values approaching 2.3 J/K^{2}mol in the 0.3–0.4 K temperature range. At still lower temperatures, this value is reduced to about 2 and 1.7 J/K^{2}mol for $CeCu_4Al$ and $CeCu_2Zn_2Al$, respectively. Such shallow maxima have been observed for other heavy-fermion systems (e.g., $CeCu_2Si_2$ and $CeAl_3$) and are usually attributed to the formation of a coherent heavy-fermion state.[13] According to both theoretical[14] and experimental studies,[13] these maxima shift to lower temperatures when magnetic field is applied and vanish for sufficiently large fields (2 T for $CeAl_3$ and 8 T for $CeCu_2Si_2$).

Both $CeCu_4Al$ and $CeCu_2Zn_2Al$, as well as the above-mentioned $CeCu_4Ga$, are derivatives of $CeCu_5$ that undergoes two magnetic phase transitions[15] at 3.6 and 3.8 K, probably indicating a very delicate and complicated magnetic ground state. $CeCu_5$ has the hexagonal $CaCu_5$-type structure containing two inequivalent Cu sites; twofold $2(c)$ ($\bar{6}m2$ symmetry) and threefold $3(g)$ (mmm symmetry) sites. According to the studies performed on RCu_4Al compounds,[16] where R is a rare-earth atom, Al preferentially takes one of the $3(g)$ sites. However, to the best of our knowledge, no definite structural studies have been reported indicating whether Al occupies one of the particular $3(g)$ sites or randomly occupies all of them. There is an even larger uncertainty about a distribution of non-Ce atoms in the unit cell of the second compound, $CeCu_2Zn_2Al$.

A replacement of one or more Cu atoms in $CeCu_5$ with either Al, Zn, or Ga leads to the destruction of the magnetic order in a very abrupt way[12,15,17] and a great enhancement of low-temperature C/T. In order to further elucidate the nature of this enhancement, we have performed specific-heat measurements on $\mathrm{CeCu_4Al}$ and $\mathrm{CeCu_2Zn_2Al}$ in magnetic fields to 12.5 T and temperatures down to 0.35 K. Our zero-field data (Fig. 1) for $\mathrm{CeCu_4Al}$ agree well with already published results;[12] we observe a maximum in C/T of 2320 $\mathrm{J/K^2\,mol}$ between 0.35 and 0.45 K. On the other hand, our lowest-temperature (0.35 K) value of C/T for $\mathrm{CeCu_2Zn_2Al}$ (Fig. 3) is near 1.8 $\mathrm{J/K^2\,mol}$, i.e., about 20% lower than the previously reported value. The above-mentioned values of C/T together with our lowest-temperature (1.8 K) magnetic susceptibilities, 100 memu/mol for $\mathrm{CeCu_4Al}$ and 50 memu/mol for $\mathrm{CeCu_2Zn_2Al}$, have been used to calculate the so-called Wilson ratio $R=\pi^2k^2\chi(T=0)/\mu_{\mathrm{eff}}^2\gamma$, a popular parameter quantifying heavy-fermion systems.[1] We assume that the Ce ions are in a stable trivalent configuration with an effective moment $\mu_{\mathrm{eff}}=2.54\mu_B$, giving Wilson ratios of 1.5 and 0.9 for $\mathrm{CeCu_4Al}$ and $\mathrm{CeCu_2Zn_2Al}$, respectively. Both values are comparable to those of $\mathrm{UCd_{11}}$ (Ref. 1) ($R\sim0.9$), $\mathrm{NpBe_{13}}$ ($R\sim1.8$), and $\mathrm{CeAl_3}$ ($R\sim0.7$), those compounds regarded as heavy fermions exhibiting band magnetism at low temperatures.[18]

The dramatic effects of magnetic field on the low-temperature specific heat of $\mathrm{CeCu_4Al}$ are presented in Fig. 1. In the temperature range of the measurement, 0.35–1.2 K, C/T monotonically decreases with the increasing field. 11 T reduces C/T at the lowest temperature to about 0.45 $\mathrm{J/K^2\,mol}$, i.e., by a factor larger than 5 from the zero-field value. Similar strong sensitivity to the magnetic field has been observed only for the isostructural compound $\mathrm{CeCu_4Ga}$,[10] where a magnetic field of 6.7 T suppresses C/T at 100 mK from about 1.8 to 0.74 $\mathrm{J/K^2\,mol}$. For comparison, the 11-T field reduces C/T ($T\rightarrow0$) of $\mathrm{CeCu_6}$ (Ref. 19) by a factor of 2.

The strong magnetic field dependence of the specific heat of $\mathrm{CeCu_4Al}$ is of no surprise considering the very large γ and thus very small effective bandwidth of a few K. What is somewhat unexpected, though, is the fact that C/T for $H=11$ T is proportional to T^2 in the whole temperature range of the measurement. Such a temperature dependence is characteristic of normal metals or antiferromagnets at temperatures well below the Néel temperature. In order to further explore this point, additional measurements over a larger temperature range and in higher magnetic fields are planned. It is also difficult to speculate at this point on the origin of the low-temperature maximum in C/T. It is not clear from our data if the low-temperature structure is removed by an application of the magnetic field or is being smeared out and moved to higher temperatures.

A simple analysis of the field depression of the Sommerfeld coefficient γ is offered by a single-ion Kondo model.[10,20] In this model, the low-temperature value of the specific heat is determined solely by the width Γ_0 of the Kondo resonance band ($\Gamma_0\propto T_k$). Magnetic field H broadens the Kondo resonance band (Γ) and decreases γ in the following way:

$$\begin{aligned}&\Gamma^2=\Gamma_0^2+(\mu H)^2\,,\\&\gamma=\frac{\pi k_B R}{3\Gamma}=\frac{\pi k_B R}{3}\,\frac{1}{[\Gamma_0^2+(\mu H)^2]^{1/2}}\,,\end{aligned}\tag{1}$$

where μ is an effective magnetic moment. It has been shown that the above relationship is well satisfied by the dilute Ce Kondo system $\mathrm{Ce_{0.1}La_{0.9}Cu_6}$ (Ref. 20) for magnetic fields as high as 5 T and the concentrated Ce system $\mathrm{CeCu_4Ga}$ up to at least 6.7 T.[10]

In Fig. 2, γ, taken as C/T at 0.35 K, is plotted versus magnetic field H. The solid line is a fit to Eq. (1). This simple formula describes rather well the magnetic field dependence of γ. The best fit has been obtained with the following parameters: $T_K=\Gamma_0/k_B=3.68$ K, $\mu=2.17\mu_B$. For comparison, such an analysis for $\mathrm{CeCu_4Ga}$ yields $T_K=4.4$ K and $\mu=2.58\mu_B$.

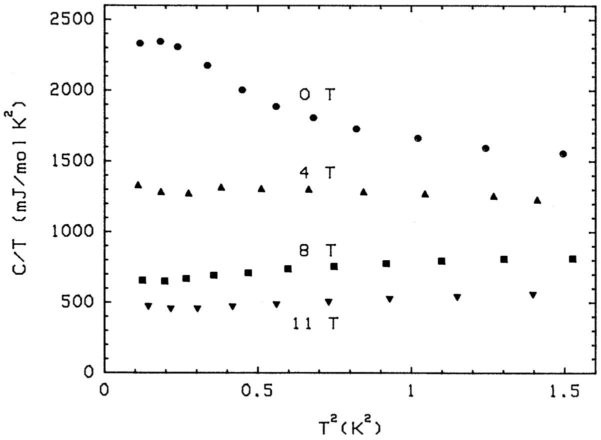

FIG. 1. Specific heat divided by temperature vs temperature squared for $\mathrm{CeCu_4Al}$ between 0.35 and 1.1 K in 0, 4, 8, and 11 T.

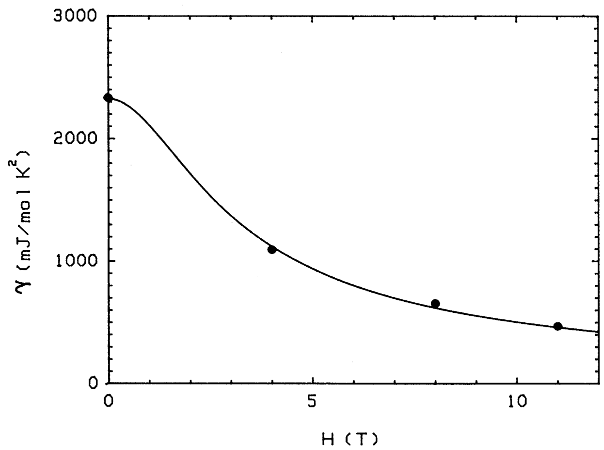

FIG. 2. Specific heat divided by temperature at 0.35 vs magnetic field for $\mathrm{CeCu_4Al}$. The solid line represents the best fit to Eq. (1).

Let us turn to the field response of the specific heat of CeCu$_2$Zn$_2$Al, shown in Fig. 3. The lowest-temperature specific-heat depression is comparable to that of CeCu$_4$Al. A 12.5-T field reduces C/T at 0.35 K by more than seven times. A maximum in C/T at $T \sim 1$ K appears for $H = 9$ T below which C/T falls rapidly with the decrease of temperature. The maximum is shifted to $T \sim 2.5$ K by the 12.5 T field. This behavior is consistent with the low-temperature specific heat of CeCu$_2$Zn$_2$Al being due, in some part at least, to magnetic correlations. The magnetic field raises the temperature below which these magnetic correlations freeze out. The nearby compound in the phase diagram CeZn$_3$Cu$_2$ is antiferromagnetic with $T_N = 6$ K.[21] An analysis similar to that of CeCu$_4$Al (presented in Fig. 2) fails completely for CeCu$_2$Zn$_2$Al. Thus, the magnetic field dependence of C/T at $T = 0.35$ K cannot be described by the single-ion Kondo model.

On the other hand, there are some experimental observations that are difficult to rectify with the magnetic model of the specific-heat enhancement. No apparent phase transition nor spin-glass freezing has been detected in zero field down to temperatures as low as 150 mK.[12] The low-temperature magnetic susceptibility is comparable to that of CeCu$_6$ and CeAl$_3$ and is twice smaller than that of CeCu$_4$Al. The Wilson ratio R is fairly small, much smaller than R of CeCu$_4$Al.

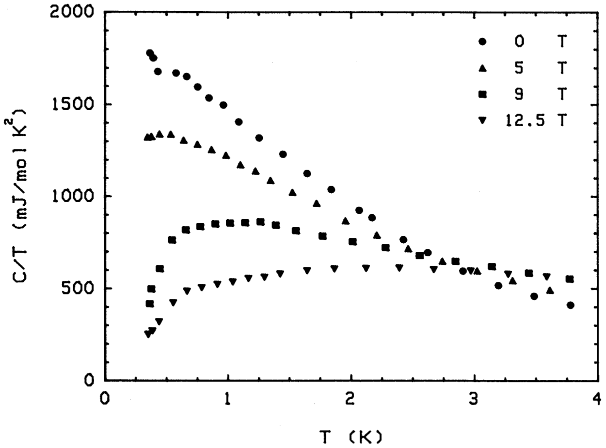

FIG. 3. Specific heat divided by temperature vs temperature for CeCu$_4$Zn$_2$Al between 0.35 and 4 K in 0, 5, 9, and 12.5 T.

In summary, low-temperature specific heats of both compounds are very sensitive to magnetic fields. Despite very similar zero-field temperature dependence, their field dependences are quite different. Different energy scales for antiferromagnetic correlations existing in both compounds can account for their dissimilar behavior in magnetic fields. Although the field dependence of the low-temperature γ of CeCu$_4$Al strongly indicates that dominant interactions in this compound are intrasite Kondo interactions, one can also expect the existence of important antiferromagnetic correlations as observed in most of the Kondo lattices at low temperatures. These intersite magnetic interactions in CeCu$_4$Al would have to be probed in different temperature and field regimes.

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