

Heavy-fermion systems

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Since the discovery by Steglich *et al.* (1979) of superconductivity in the high-effective-mass ($\sim 200m_e$) electrons in CeCu_2Si_2 , the search for and characterization of such "heavy-fermion" systems has been a rapidly growing field of study. The eight heavy-fermion systems known to date include superconductors (CeCu_2Si_2 , UPt_3 , NpBe_{13}), magnets (NpBe_{13} , U_2Zn_{17} , UCd_{11}), and materials in which no ordering is observed (CeAl_3 , CeCu_6). These f -electron materials have, in comparison to normal metals, enormous specific heat γ values ($450\text{--}1600\text{ mJ/mol K}^2$), large values of the low-temperature magnetic susceptibility χ ($8\text{--}50 \times 10^{-3}\text{ emu/mol G}$), maxima in the resistivity at low temperatures with large ρ_{max} values ($100\text{--}200\ \mu\Omega\text{ cm}$), and unusual temperature dependences of their specific heats below 10 K. The three heavy-fermion superconductors show such unusual behavior that the possibility of p -wave pairing of the superconducting electrons, rather than the usual BCS s -wave pairing, cannot be ruled out. This paper reviews the experimental results to date, to serve both as a status report and as a starting point for future research. Several correlations between properties are pointed out, including the observation that a low value of the Wilson ratio ($\sim \chi/\gamma$) appears to correlate with the occurrence of superconductivity.

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I. INTRODUCTION

Steglich, Aarts, Bredl, Lieke, Meschede, Franz, and Schäfer discovered bulk superconductivity in the highly correlated, high-effective-mass $4f$ electrons in CeCu_2Si_2 in 1979. Based on what was known prior to this work, an f -electron system such as CeCu_2Si_2 , with a specific heat γ of 1100 mJ/mol K^2 , an effective mass $m^* \sim 10^2 m_e$, a large Ce-Ce separation of $4.1\ \text{\AA}$, and a high-temperature Curie-Weiss susceptibility χ with $\mu_{\text{eff}} = 2.68\mu_B$, would be expected to have magnetic interactions that were much too strong to allow superconductivity. The discovery of an enormous specific-heat discontinuity and bulk Meissner effect associated with resistive and inductive indications of superconductivity at 0.5 K was a *tour-de-force* proof that it is indeed the high-mass electrons ("heavy fermions") which are superconducting in CeCu_2Si_2 . This seminal work started an intense search for and study of other such (so-called "heavy-fermion") systems, as well as continued study of CeCu_2Si_2 . Two other heavy-fermion superconductors have since been discovered, UPt_3 (Ott *et al.*, 1983) and NpBe_{13} (Stewart *et al.*, 1984c), as well as three heavy-fermion materials that are magnetic (nonlocal and nonferromagnetic), NpBe_{13} (Stewart *et al.*, 1984b), U_2Zn_{17} (Ott *et al.*, 1984a), and UCd_{11} (Fisk *et al.*, 1984b). In addition, there exist two heavy-fermion systems with no ordering down to 0.050 K: CeAl_3 (Andres *et al.*, 1975), the first known heavy-fermion system, and the recently discovered CeCu_6 (Stewart, Fisk, and Wire, 1984) with low-temperature γ values of 1620 and $\sim 1600\text{ mJ/mol K}^2$, respectively. Thus, according to the arbitrary definition that a "heavy-fermion" system has $\gamma > 400\text{ mJ/f atom mol K}^2$, eight heavy-fermion systems are now known. (The specific heat γ will be normalized to a mole of f atoms throughout this paper to allow a proper comparison between systems with dissimilar formula units, e.g., UCd_{11} and U_2Zn_{17} .)

Because this field is rapidly growing, with new results (it seems) every month, the present paper must become incomplete even before publication. However, precisely because the study of heavy-fermion systems has grown so

rapidly, a review of the large amount of work done to date on these eight materials should be a useful status report, pointing out the consistencies and correlations that already exist, as well as the unanswered questions, in what may seem a welter of unconnected data.

All eight of these systems, which have large γ 's and m^* 's, contain f electrons. They all have f -atom separations greater than 4 Å, significantly greater than 3.25–3.50 Å (the Hill limit), beyond which Hill (1970) pointed out that f - f overlap ceases and, unless f hybridization with s , p , or d electrons prevents it, magnetism occurs. They all have susceptibilities that are large but finite at $T \rightarrow 0$ and obey (reasonably well) a Curie-Weiss law from 100–300 K with large effective moments. However, the sum of these properties is not a sufficient condition for a material to have a $\gamma > 400$ mJ/ f atom mol K². For example, USn₃ has (Buschow and van Daal, 1972; van Maaren *et al.*, 1974) $\chi(T \rightarrow 0) = 10 \times 10^{-3}$ emu/mol, a Curie-Weiss law in χ between 100 and 300 K, with $\mu_{\text{eff}} = 2.5\mu_B$, and $d_{\text{U-U}} = 4.63$ Å, with γ equal to "only" 169 mJ/ f atom mol K². (The χ , γ , and d values for systems with $\gamma > 300$ mJ/ f atom mol K² are summarized later in Table VI.) Obviously, the value of 400 mJ/ f atom mol K² is an arbitrary one. In fact, the term "heavy fermion" has been used to describe systems with significantly lower γ values, including U₂PtC₂, $\gamma = 75$ mJ/ f atom mol K² (Meisner *et al.*, 1984), U₆Fe, $\gamma = 24$ mJ/ f atom mol K² (DeLong *et al.*, 1983), and U₆Co, $\gamma = 21$ mJ/ f atom mol K² (Klaase *et al.*, 1984). In order to maintain a manageable focus, we shall concentrate on the eight systems with $\gamma > 400$ mJ/ f atom mol K². In addition, some discussion of less heavy systems such as YbCuAl ($\gamma = 260$) (units for γ understood from now on to be mJ/ f atom mol K²), NpSn₃ ($\gamma = 242$), and Pu_{0.7}U_{0.3}Al₂ ($\gamma = 260$) will be included to show how materials properties vary in the region between "normal" systems and the "heavy-fermion" systems.

Even with this draconian focus, the field at hand to be discussed remains quite large. The organization of this paper is given in the table of contents. Separation of the discussion into sections on each type of measurement enhances comparison between systems, but hinders a full coherent discussion of all the properties of any given system. For the reader interested in one given system, the order of discussion of the 11 systems in the specific-heat section is followed as closely as possible in succeeding sections, especially for figures and within tables, to facilitate an overview of one material by skipping from section to section. A summary is included after Sec. II (specific heat) to try to maintain a focus on the main points discussed. Figures and tables are made to point out clearly correlations between the systems. Section V includes short discussions of neutron scattering, photoemission, NMR, thermal expansion, and chemical substitution experiments on heavy-fermion systems. Section VI is an experimentalist's attempt to discuss (briefly) the existing theories on heavy-fermion systems as well as on superconductivity therein. This section is merely an attempt at a thorough, up-to-date literature survey. Section VII is a

summary of how the heavy-fermion systems intercompare, and offers suggestions for useful further measurements.

A final observation should be made about the materials parameters discussed herein: where more than one measurement of a given parameter is known to exist, all the results are given. Sample variation plays a crucial role in (at least) CeCu₂Si₂ and CeAl₃, where, for example, γ for superconducting samples of CeCu₂Si₂ varies from 1300 (Bredl *et al.*, 1983) to 730 (Assmus *et al.*, 1984), while nonsuperconducting crystals can have a γ of 400 (Willis *et al.*, 1984b). Similar, although usually smaller, variation occurs in *all* the other parameters where more than one measurement exists (not yet the case for many of the systems), not only due to sample variations (degree of lattice disorder, stoichiometry, amount and kind of second phase), but also possibly due to measurement errors. Sometimes measurements of all the parameters are not available on the same sample, and, in the case of CeCu₂Si₂, there is even an "aging" effect where sample parameters measured by the same group on the same sample show important changes with time (Steglich *et al.*, 1979). The reader should therefore beware of making conclusions of greater precision than is warranted by the data.

II. SPECIFIC HEAT

A short discussion of the interpretation of specific-heat data in general is presented here. For a more thorough treatment, see Gopal (1966). In a usual metal (see Sec. II.H) the specific heat C is given by

$$C = \gamma T + \beta T^3 \quad (1)$$

for temperatures below $\Theta_D/50$ (and depending on the material even up to $\Theta_D/10$), where Θ_D is the Debye temperature and γ is proportional to the electronic density of states. (Neither γ nor Θ_D depends significantly on T in a normal metal in this temperature range.)

Therefore, a C/T vs T^2 plot of a normal metal at sufficiently low temperatures has an intercept of γ and a positive slope of β , where

$$N(0)(1 + \lambda) = 0.4244\gamma/n, \quad (2)$$

$$\beta = (1944 \times 10^3)n/\Theta_D^3,$$

with $N(0)$ the bare, or band-structure electronic density of states at the Fermi energy in units of states/eV atom, λ an interaction (electron-phonon and electron-electron) parameter (see Grimvall, 1968), C in units of mJ/formula unit mol K, and n the number of atoms in a formula unit (e.g., $n = 14$ for UBe₁₃). [To calculate $N(0)$ for d - and f -atom systems, one can presume that the non- d or non- f atoms contribute little to $N(0)$, and use n as the number of d or f atoms in a formula unit to calculate $N(0)$ in states/eV d (or f) atom. This choosing of the normalization basis, or n , may not be done for Θ_D , since all atoms vibrate in a common lattice.]

A. CeCu_2Si_2

1. History; discussion of properties

The history of research on CeCu_2Si_2 (tetragonal structure, $d_{\text{Ce-Ce}}=4.1 \text{ \AA}$; see Rossi *et al.*, 1979) is an interesting one. Rieger and Parthe (1969) showed that EuCu_2Si_2 and YbCu_2Si_2 had abnormal lattice constants (for a discussion, see Lawrence *et al.*, 1981, p. 27) in comparison with other rare-earth (R_E) Cu_2Si_2 compounds. This was taken as a sign of intermediate valence by Sales (1974) and Sales and Viswanathan (1976), who made measurements of susceptibility (χ), resistivity, thermopower, specific heat, and lattice constants as a function of temperature for various $R_E\text{Cu}_2\text{Si}_2$ compounds. The deviation from a Curie-Weiss behavior in χ at lower temperatures, coupled with the apparent nonintegral valence implied from the lattice constants, was taken as a sign of interconfiguration fluctuation of the valence in EuCu_2Si_2 , YbCu_2Si_2 , and CeCu_2Si_2 —although the lattice constants appeared almost normal for the latter. Sales and Viswanathan (1976) measured the specific heat only on the Eu and Yb compounds. Franz, Griessel, Steglich, and Wohlleben (1978) reported on further work on LaCu_2Si_2 and CeCu_2Si_2 , stating that the latter was “quite close” to integral valent, with an uncertainty of 5%. These authors reported in a footnote the measurement of the resistance down to 0.3 K by W. Lieke, who discovered a superconductivity transition, R going to 0, at 0.6 K in CeCu_2Si_2 .

Fifteen months after the manuscript of Franz *et al.* (1978) was submitted, bulk superconductivity in CeCu_2Si_2 (Steglich *et al.*, 1979) had been shown by specific-heat and dc Meissner-effect measurements. The C vs T data are shown in Fig. 1. Data for two other samples (not shown here) also had bulk specific-heat anomalies. Also

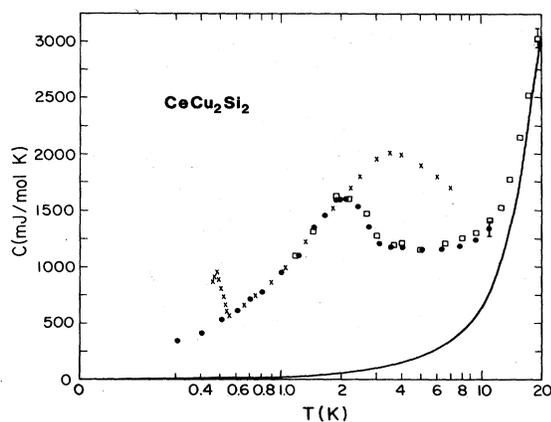


FIG. 1. Specific heat of CeCu_2Si_2 : \times , superconducting polycrystalline sample (Steglich *et al.*, 1979—only a representative subset of the data is shown); \bullet and \square , nonsuperconducting single crystals (Stewart, Fisk, and Willis, 1983). The solid line drawn is an extrapolation of the higher-temperature, “normal” data which follow $C = \gamma T + \beta T^3$. The units for C are per f atom mol, as for all the figures and text.

shown in Fig. 1 are data from a nonsuperconducting single crystal (Stewart, Fisk, and Willis, 1983), in good agreement with the data of Steglich *et al.* between T_c and 2 K. For a better analysis of the normal-state specific heat of CeCu_2Si_2 , the nonsuperconducting crystal data, which extend over a wider temperature range, are replotted as C/T vs T^2 in Fig. 2.

The nonsuperconducting peak in the C vs T data between 2 and 4 K in Fig. 1 at low temperature (to be discussed later) is not visible in Fig. 2 because such a plot (C/T vs T^2) obscures structure in C . What is evident, however, is that nowhere in the range $T < \Theta_D/10$ (Θ_D should be $200 \pm 10 \text{ K}$ for CeCu_2Si_2 , based on the specific-heat data for EuCu_2Si_2 and YbCu_2Si_2 and the relative masses of the $4f$ atoms), is Eq. (1) satisfied for CeCu_2Si_2 . Instead what is seen in Fig. 2 is a rapid increase in C/T below 10 K for CeCu_2Si_2 that is characteristic of five of the eight heavy-fermion systems known, the exceptions being UPt_3 , U_2Zn_{17} , and UCd_{11} . Even over a very restricted temperature range (inset, Fig. 2) there is no region where Eq. (1) is followed. [Implicitly, Eq. (1) has γ not equal to a function of temperature and $\Theta_D > 0$.]

Because γ is not a rapid function of temperature at low temperatures in normal metals, the almost tenfold in-

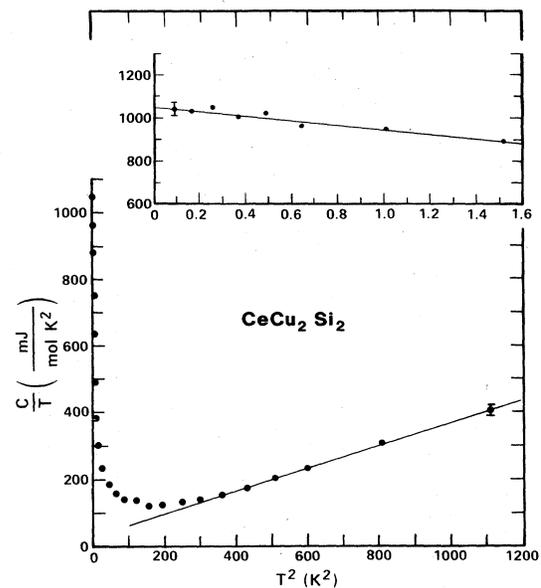


FIG. 2. Specific heat of the nonsuperconducting single crystals of CeCu_2Si_2 shown in Fig. 1, but replotted as C/T vs T^2 . Note that the sizable anomaly peaked at 2 K in Fig. 1 is not visible here when plotted as C/T vs T^2 . The inset shows the very-low-temperature behavior, i.e., γ vs T^2 , where γ clearly rises with decreasing temperature. The high-temperature ($T > 20 \text{ K}$) specific heat, although it follows Eq. (1), $C = \gamma T + \beta T^3$, has a β that gives $\Theta_D = 310 \text{ K}$, which, as discussed in the text, is too high. It is probable that the high-temperature specific-heat data shown contain a contribution from a crystal-field split electronic state which shapes C in this temperature range, giving an apparent simple Debye-law behavior which is in fact fallacious.

crease in C/T within 10 K shown in Fig. 2 would normally be thought to be due to the anomaly seen at 2 K, and not to an extremely temperature-dependent dressed density of states, Eq. (2). However, the second-order superconducting phase transition observed in CeCu_2Si_2 opens a gap in the heavy, $\gamma=1100$, f -electron spectrum, as shown by the large discontinuity ΔC in the specific heat at T_c (see Fig. 1). This discontinuity is proportional to $N(0)(1+\lambda)$, the dressed density of states, through the relation $\Delta C \propto \gamma T_c$, where in BCS theory the proportionality constant is 1.43. It is this discovery of superconductivity in a system with such a huge value of $N(0)(1+\lambda)$ that has caused the enormous interest in heavy-fermion systems.

The ratio of $\Delta C/\gamma T_c$ is an important parameter for a superconductor, not only because it verifies the bulk nature of the superconductivity, but also because it serves as a means of distinguishing between weak ($\Delta C/\gamma T_c \sim 1.43$) and strong ($\Delta C/\gamma T_c > \sim 1.6$) electron-electron coupling. The ratio $\Delta C/\gamma T_c$ for CeCu_2Si_2 is uncertain for three reasons. One of these, sample dependence, will be discussed below. Another is temperature dependence—because γ is temperature dependent, does one use $\gamma(T=0)$ or $\gamma(T=T_c)$? Finally, because the superconducting anomaly in C is broad, does one use the value C_{max} minus the value of C_n extrapolated to this lower temperature, or does one estimate, using entropy arguments, an idealized, sharp ΔC and use this (30–40%) larger value for ΔC ? Figure 3 shows this set of choices graphically. Work to date has consistently used the choices that give the largest value for $\Delta C/\gamma T_c$, i.e., $\gamma(T_c)$ and ΔC (idealized; Steglich *et al.*, 1979, 1982; Lieke *et al.*, 1982; Bredl *et al.*, 1983; Assmus *et al.*, 1984). This is considered further in the discussion of sample dependence below.

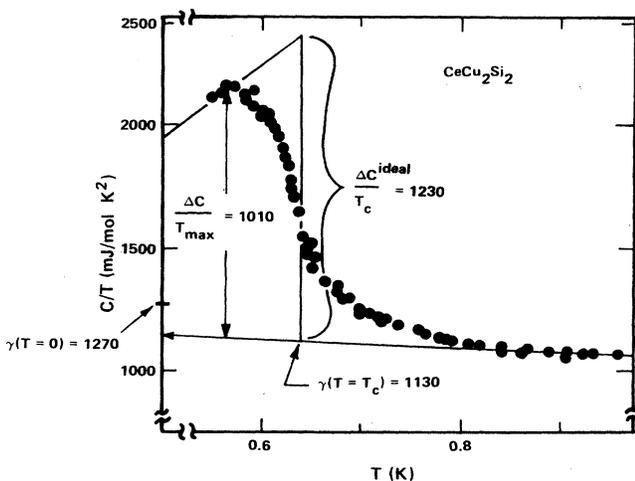


FIG. 3. Data for CeCu_2Si_2 , sample 7, by Lieke *et al.* (1982), which show graphically the various ways to calculate $\Delta C/\gamma T_c$. Note that, although $\Delta C/T_{\text{max}}$ is only 20% less than the $\Delta C^{\text{ideal}}/T_c$ shown, the difference in the respective ΔC values is larger, due to the difference between T_c and T_{max} .

Before the sample dependence of the specific heat is discussed, the anomaly at 3.5 K (see Fig. 1) should be considered. What is this anomaly in the superconducting crystal of Steglich *et al.*? The current description of CeCu_2Si_2 is that it is a “Kondo-lattice system” (Assmus *et al.*, 1984; Steglich *et al.*, 1982), whereas Sales and Viswanathan focused on a “mixed valency” description. “Kondo-lattice system” is the nomenclature used by these and other authors to allow the understanding of dilute Kondo systems, e.g., $\text{La}_{1-x}\text{Ce}_x\text{Al}_2$ (Bader *et al.*, 1975; Bredl *et al.*, 1978) to be used qualitatively in describing concentrated f -atom systems, e.g., CeAl_2 (Bredl and Steglich, 1978), which show behavior reminiscent of dilute systems. For CeCu_2Si_2 the decision as to which description (Kondo or mixed-valent) to use is a difficult one, because CeCu_2Si_2 is “close” to being integral valent (Franz *et al.*, 1978) and also because the two models are phenomenologically quite similar. In a dilute Kondo system, one expects the compensation of the local impurity moments by the conduction electrons to remove the spin-degeneracy entropy $k_B \ln(2s+1)$ associated with each local spin s . For a “Kondo-lattice system,” without any complete theory for guidance, an entropy of $R \ln(2s+1)$ per mole of the moment-bearing atoms associated with an anomaly is taken as implying “Kondo-like” behavior. Since approximately $R \ln 2$ is found for the 3.5-K anomaly for the superconducting crystal shown in Fig. 1, and based on other arguments (e.g., existence of distinct crystal-field levels—Horn *et al.*, 1981), the anomaly is attributed to “Kondo-lattice” behavior (Steglich *et al.*, 1982). Theoretically, as we shall see, the nearness of CeCu_2Si_2 to integral valency can be used as one way to explain the heavy-fermion behavior.

2. Sample dependence

The compound CeCu_2Si_2 has *extremely* sample-dependent properties whose systematics are not yet completely understood. The anomaly shown in Fig. 1 is clearly different for the two different samples. The entropy associated with the anomaly ($T < 20$ K) in the nonsuperconducting single crystal is only about $\frac{2}{3}$ of $R \ln 2$ (Stewart, Fisk, and Willis, 1983). There is also a sample dependence in the size of this anomaly for superconducting samples (Aarts, 1983). Currently, using both superconducting and nonsuperconducting single crystals of CeCu_2Si_2 , a study is underway to correlate this anomaly with $\gamma(T=0)$ and $\chi(T=0)$ (Willis *et al.*, 1984b; Batlogg *et al.*, 1984c).

The variation of γ , T_c , and $\Delta C/\gamma T_c$ between samples for CeCu_2Si_2 is enormous. Ishikawa *et al.* (1983) have shown that T_c for CeCu_2Si_2 drops below 0.07 K for a very slight Cu deficiency ($\text{Cu}_{1.95}$ instead of $\text{Cu}_{2.00}$), and less rapidly for variations in Si composition. This work provided a probable explanation for the results of Hull *et al.* (1981) and Aliev *et al.* (1982), who found no indication of superconductivity above 0.060 K in their samples of CeCu_2Si_2 , in spite of following the preparation and an-

TABLE I. Sample variation of specific-heat parameters for CeCu_2Si_2 . ΔC is defined as $C^{\text{max}}/T - C_n^{\text{extrap}}/T$, where C_n^{extrap}/T is the extrapolated value of C_n/T at the temperature where the maximum in C occurs. Units for γ and C are mJ/mol K^2 , the same as used in the text, since a formula unit CeCu_2Si_2 has 1 Ce atom. Values for T_c are taken from the midpoint of the specific-heat anomaly. Values for $\gamma(T=0)$ and $\Delta C/T_{\text{max}}$ are taken from published graphs and are accurate to $\pm 5\%$. All samples are polycrystalline unless noted otherwise. It is not clear how samples 4 and 7 from Breidl *et al.* (1983) differ from those from Steglich *et al.* (1982) and Lieke *et al.* (1982); other than that they appear not to be the same samples in their properties.

Sample	T_c^{midpoint} (K)	$\gamma(T=0)$	$\Delta C/T_{\text{max}}$	$\Delta C/\gamma(T=0)T_{\text{max}}$	$\Delta C^{\text{ideal}}/\gamma(T=T_c)T_c$
sample 3 ^a	0.53	830	790	0.95	1.40
sample 4 ^a	0.47	1130	1000	0.89	1.36
sample 7 ^b	0.64	1270	1010	0.80	1.09
c	0.51				0.85
same as previous sample, but stored for 2 months in argon and remeasured	0.49				0.76
sample 4 ^d	0.66	900-1000	660	0.69	1.22
sample 7 ^d	0.66				0.6
60% Meissner effect					
sample 10 ^d	0.47				0.1
40% Meissner effect					
sample 11 ^d	0.29				0.0
50-65% Meissner effect					
single crystal ^e	0.69	860	580	0.67	1.27
single crystal ^f	0.050	1050			
single crystal ^g	0.3	675			
grown in excess Cu	[$T_c(\chi)=0.6$]				
single crystal ^h	0.4	400			
grown in In					
single crystal ⁱ	0.4	1100			
grown in Sn					

^aSteglich *et al.*, 1982 (their Fig. 1).

^bLieke *et al.*, 1982 (their Fig. 3).

^cSteglich *et al.*, 1979.

^dBreidl *et al.*, 1983.

^eAssmus *et al.*, 1984.

^fStewart, Fisk, and Willis, 1983.

^gWillis *et al.*, 1984b.

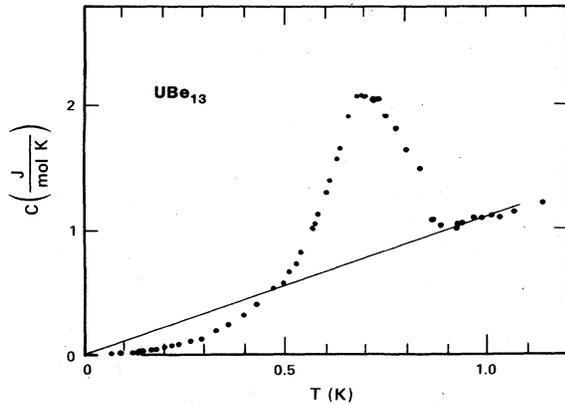


FIG. 4. Low-temperature specific heat of UBe_{13} single crystals by Ott *et al.* (1983).

nealing conditions reported by Steglich and co-workers. A similar study of T_c with composition has been performed by Spille *et al.* (1983), who also studied the alloying dependence of T_c .

For polycrystalline samples, Steglich *et al.* (1982) and Bredl *et al.* (1983) have pointed out a correlation between T_c and C/T at T_c [$\equiv \gamma(T=T_c)$], namely that T_c varies inversely as $\gamma(T=T_c)$. Unfortunately this correlation does not hold for single crystals (Stewart, Fisk, and Willis, 1983; Assmus *et al.*, 1984).

The variation of ΔC has been similarly large, with some samples having a substantial dc Meissner effect (50%) showing *no* ΔC (Bredl *et al.*, 1983), i.e., gapless superconductivity. The variation of the specific-heat parameters for CeCu_2Si_2 is shown in Table I. We see that γ varies from 400 to 1270 and that $\Delta C/\gamma T_c$ also varies widely, from sample to sample and based on how ΔC and γ are chosen, as discussed above.

B. UBe_{13}

Fortunately for researchers in the field, not all heavy-fermion superconductors show the enormous sample variation found in CeCu_2Si_2 , as shown by the discovery of bulk superconductivity in cubic UBe_{13} ($d_{\text{U-U}} = 5.13 \text{ \AA}$; see Pearson, 1958, p. 458) by Ott *et al.* (1983) and subsequent work. The discovery of heavy-fermion superconductivity in UBe_{13} also has an interesting history. Bucher *et al.*

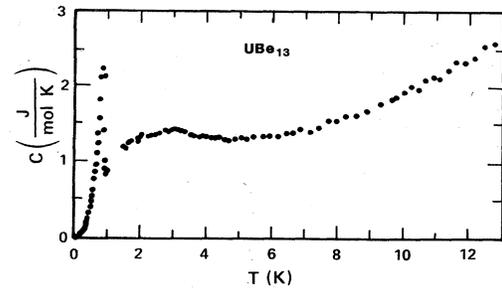


FIG. 5. Specific heat of polycrystalline UBe_{13} between 0 and 13 K by Ott *et al.* (1984c). Note the anomaly in C centered between 2 and 4 K.

(1975) investigated the electronic properties of 17 $M\text{Be}_{13}$ compounds, where M included U. They discovered a sharp superconducting transition in UBe_{13} at 0.97 K, which was only slowly depressed by applied magnetic field. Although powdering the sample did not destroy the ac-susceptibility-measured T_c , precipitated U filaments were speculated as being the cause of the T_c , and not the majority phase. If the specific-heat measurements made by Bucher *et al.* on UBe_{13} down to 1.8 K had been performed to lower temperature in a ^3He refrigerator, the field of heavy-fermion superconductivity would have started five years earlier than it did.

The specific heat at low temperatures reported by Ott *et al.* (1983) is shown in Fig. 4 and has an extrapolated $\gamma(T=0)$ of 1100. This broad superconducting transition has since been considerably improved by Ott *et al.* (1984b), see Table II and Fig. 5.

The specific heat above T_c of UBe_{13} has been measured by a number of workers. This work is summarized in Table II, including a comparison of the results at 12 K, the common point of overlap in the various sets of data. The results of Ott *et al.* (1984c) up to 13 K are shown in Fig. 5. Note the anomaly in C in Fig. 5 centered on 2 K. How does this anomaly for UBe_{13} compare to that for CeCu_2Si_2 discussed above? The large "Kondoesque" anomaly seen in CeCu_2Si_2 (Fig. 1) at 3.5 K by Steglich *et al.* (1979) involved about $R \ln 2$ of entropy, where the height of the peak was 1.85 J/mol K above the extrapolation of the higher-temperature data. Although there is a peak in the data of Fig. 5 for UBe_{13} , centered on 2 K, its height above the extrapolation of the higher-temperature data to $T=0$ is only about 1 J/mol K. Ott *et al.* (1984c)

TABLE II. Variation in specific-heat parameters for UBe_{13} .

Reference	Temperature range (K)	T_c^{onset} (from C) (K)	ΔT_c (K)	$C_n(T_c)$ (mJ/U mol K)	ΔC (at C_{max}) (mJ/U mol K)	C/T at 12 K (mJ/mol K ²)
Ott <i>et al.</i> , 1983	0.06–10	0.88	0.19	1000	1360	
Ott <i>et al.</i> , 1984c	0.06–13	0.92	0.06	810	1680	185–200
Samorukov <i>et al.</i> , 1974	13.5–300					119
Bucher <i>et al.</i> , 1975	1.8–42	0.97				151 ^a
Stewart and Giorgi, 1984	1.4–11					128

^aCorrected for the ThBe_{13} subtraction in their Fig. 4.

make no mention of the entropy under this bump. By measuring points from the published curve and using a reasonable extrapolation of the data from above the anomaly to $T=0$ and from just above T_c to $T=0$, the anomaly has only 1 ± 0.2 J/mol K associated with it, compared to $R \ln 2 = 5.8$ J. It is unclear how important this difference is and what its meaning is for understanding the heavy-fermion behavior of the two systems. The specific heats of the nonsuperconducting crystal CeCu_2Si_2 (Stewart, Fisk, and Willis, 1983, Fig. 2—entropy only $\frac{2}{3}R \ln 2$) and of UBe_{13} are plotted together as C/T vs T^2 in Fig. 6.

As shown by Fig. 6, CeCu_2Si_2 and UBe_{13} have comparable electronic specific heats. Even though there is a difference in the size of the anomalies of the two systems at around 2 K, the $\gamma(T=0)$ values are identical (~ 1100) and the temperature variation of C/T vs T^2 between 1.5 and 8 K is, on the scale of Fig. 5, the same. Above 8 K, the significant difference in lattice stiffness ($\Theta_D = 200$ K for CeCu_2Si_2 vs ~ 620 K for UBe_{13}) causes the C/T for CeCu_2Si_2 to rise more rapidly.

C. UPt_3

The specific heat of UPt_3 (hexagonal structure, $d_{\text{U-U}} = 4.1$ Å, see Pearson, 1958, p. 825) was first measured by Frings *et al.* (1983) on polycrystalline material from 1.4 to 24 K. These data, plotted as C/T vs T^2 , showed an upturn below 10 K, called “rather similar” to (Trainor *et al.*, 1975; Stewart, Giorgi, Brandt, Foner, and

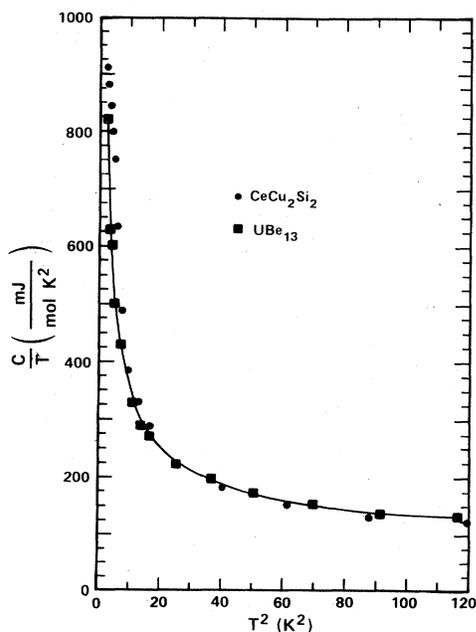


FIG. 6. Specific heat of nonsuperconducting single crystals of CeCu_2Si_2 (●, Stewart, Fisk, and Willis, 1983) and a piece of a superconducting single crystal of UBe_{13} (■, Stewart *et al.*, 1984a). The line through the UBe_{13} serves as a guide to the eye.

Arko, 1983) that observed in the spin fluctuator UAl_2 , with a zero-temperature intercept γ of about 420. Doniach and Engelsberg (1968) and Brinkman and Engelsberg (1968) predicted that, in addition to the usual γT and βT^3 terms in the specific heat, long-range spin fluctuations would produce a $T^3 \ln T$ temperature dependence in C . Such a $T^3 \ln T$ term was first observed in the low-temperature specific heat of ^3He , and was later (Greywall, 1983) found to be valid up to 200 mK for this system; a $T^3 \ln T$ term has been found in the specific heat of UAl_2 (Trainor *et al.*, 1975; Stewart, Giorgi, Brandt, Foner, and Arko, 1983) and of TiBe_2 (Stewart *et al.*, 1982).

The study of UPt_3 by Stewart *et al.* (1984c) was motivated by the possibility that the observed upturn in UPt_3 was a $T^3 \ln T$ or spin-fluctuation term, as well as by the fact that the structure of UPt_3 , hexagonal DO19, was the same as that found in CeAl_3 , the first heavy-fermion system to be discovered. Stewart *et al.* (1984c) found not only a $T^3 \ln T$ term in C , but also resistive, ac susceptibility and specific-heat transitions at 0.54 K into the superconducting state, making UPt_3 the third heavy-fermion superconductor known, and the first-known superconductor with a $T^3 \ln T$ term in C . The specific-heat anomaly observed at T_c is shown in Fig. 7, with $\Delta C^{\text{max}}/\gamma T_c = 0.48$. Although the transition is quite broad, it is a clear indication of bulk superconductivity. An idealized $\Delta C/\gamma T_c$ was estimated by Stewart *et al.* (1984c) to be at least 1.0.

The specific-heat data for $T > T_c$ of Stewart *et al.* on single crystals of UPt_3 are shown in Fig. 8. The temperature dependence of C above T_c does in fact obey a $\delta T^3 \ln T + \gamma T + \beta T^3$ dependence (solid line in Fig. 8). Because C/T in Fig. 8 equals $\gamma(=452) + \beta T^2 + \delta T^2 \ln T$, a plot of $(C/T - 452)/T^2$ vs $\ln T$ should be (and is, see Fig. 9) a straight line for UPt_3 . This $T^3 \ln T$ term, coupled

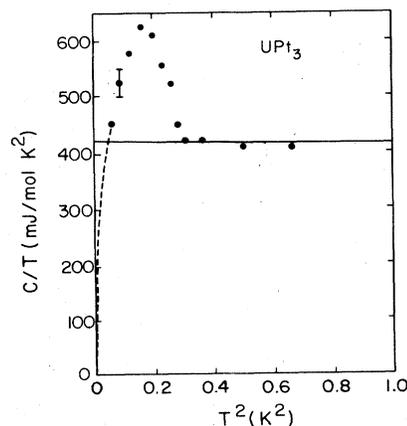


FIG. 7. Superconducting transition in the specific heat of annealed single crystals of UPt_3 (Stewart *et al.*, 1984c). The disagreement between the high temperature extrapolation and the low-temperature one shown here was due to an error in the absolute accuracy of the lower-temperature platform, with the precision of the data stated to be $\pm 2\%$.

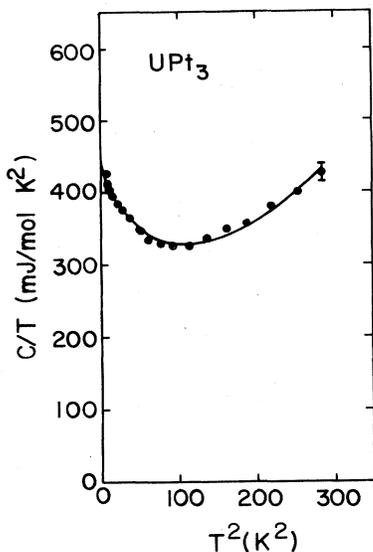


FIG. 8. Specific heat in the normal state of flux-grown single crystals of UPt_3 (Stewart *et al.*, 1984c). The line through the data is a least-squares computer fit of the data to $C = \gamma T + \beta T^3 + \delta T^3 \ln T$.

with resistive and magnetic evidence discussed by Stewart *et al.* (1984c), is strong indication of spin fluctuations in UPt_3 . Thus a major difference between UPt_3 and CeCu_2Si_2 and UPe_{13} (see Fig. 6) is the temperature dependence of C/T , with the latter two compounds having a much more rapid variation of C/T with temperature. This is apparent when comparing the size of the increases in C/T below 10 K of the three systems, i.e., C/T increases less than 50% from 10 K down to T_c in UPt_3 , versus approximately a factor of 10 in CeCu_2Si_2 and UPe_{13} . Moreover, attempts to fit the C data for UPe_{13} or

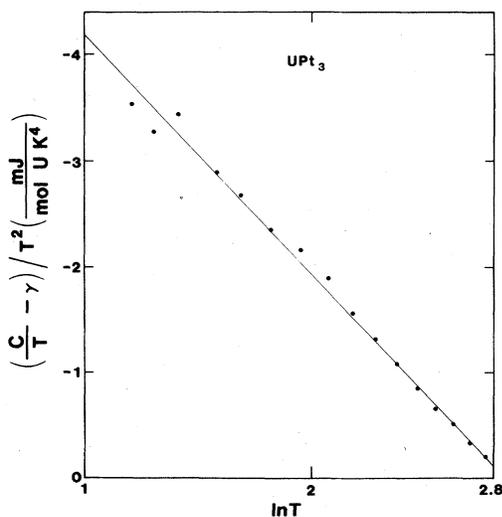


FIG. 9. Specific-heat data for UPt_3 plotted as $(C/T - \gamma)/T^2$ vs $\ln T$, in order to show clearly the $T^3 \ln T$ behavior.

CeCu_2Si_2 to the form $\gamma T + \beta T^3 + \delta T^3 \ln T$ using a least-squares computer fit fail completely (Stewart, 1984), whereas this form fits the UPt_3 data quite well (Figs. 8 and 9).

The apparent coexistence of spin fluctuations and superconductivity in UPt_3 raises the possibility of p -wave pairing of the superconducting electrons (as found in the nuclear spins of superfluid ^3He ; see Leggett, 1975) rather than the alignment of antiparallel spins, s -wave pairing, as derived by the BCS theory for superconductors. Analysis (Varma, 1984a) of critical-field data for T_c in UPt_3 (measured by Willis *et al.*, 1984a and Chen *et al.*, 1984), discussed below in the magnetic properties section, and of ultrasonic attenuation results (Bishop *et al.*, 1984) lend further credence to the speculation that UPt_3 may be the first p -wave superconductor. Benoit *et al.* (1981) have pointed out similarities in the specific heat of CeAl_3 to that of ^3He above the paramagnon region in the latter. Moreover, Ott *et al.* (1984e) use the similarity of the temperature dependence of C for UPe_{13} between 1 and 8 K with that of the specific heat of ^3He between 0.1 and 0.6 K (mostly above the paramagnon region) to argue for p -wave pairing in UPe_{13} . These speculations of p -wave pairing in UPt_3 and UPe_{13} enliven the field of heavy-fermion systems and have given a tremendous impetus to both experimentalists and theorists for further work.

Recently, de Visser *et al.* (1984) have also measured the specific heat down to 0.13 K on an unannealed single crystal of UPt_3 . Although de Visser *et al.* observe a broader transition ($\Delta C^{\text{max}}/\gamma T_{\text{max}} = 0.25$) with $T_c^{\text{onset}} = 0.43$ K, the value of $\gamma(T=0)$, 422, is in reasonable agreement with the measurement of Frings *et al.* and Stewart *et al.* Thus sample variation plays only a slight role in the normal-state specific heat of UPt_3 , although, as was pointed out by Stewart *et al.* (1984c) sample quality plays an important role for superconductivity (discussed further in the context of resistivity, Sec. IV)—another argument for p -wave pairing.

D. Specific heat for $T < T_c$

Now that the specific heats of the three heavy-fermion superconductors have been reviewed for $T > T_c$, the data below T_c should be discussed. Due to the lower T_c and much broader transition in UPt_3 (see Fig. 7), insufficient data exist at this time to include UPt_3 in the discussion.

The logarithm of the superconducting electronic specific heat C_s , divided by the normal-state specific heat at T_c , is shown versus T_c/T in Fig. 10 for both CeCu_2Si_2 and UPe_{13} . The line shown is the behavior predicted by BCS. The data for UPe_{13} [$\Delta C^{\text{ideal}}/\gamma(T=T_c)T_c \sim 2.5$] were reported by Ott *et al.* (1984e). The data for CeCu_2Si_2 [sample 4, $\Delta C^{\text{ideal}}/\gamma(T=T_c)T_c = 1.22$] were reported by Bredl *et al.* (1983), who also reported data for sample 7 similar to those shown; data for sample 10 and sample 11 decreased more slowly with increasing T_c/T . Although, near T_c , the data shown in Fig. 10 differ significantly in temperature dependence and magnitude, the

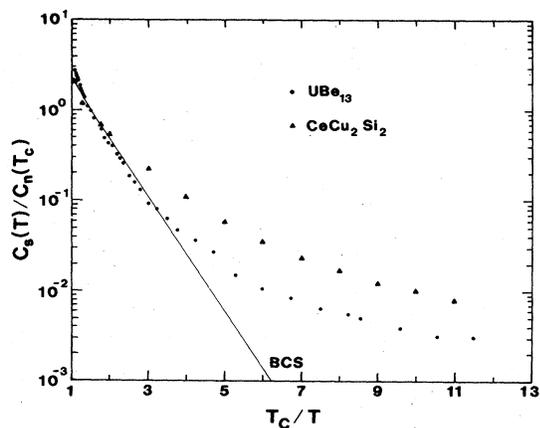


FIG. 10. Logarithm of the low-temperature superconducting electronic specific heat vs T_c/T for UBe_{13} (Ott *et al.*, 1984e) and for CeCu_2Si_2 , "sample 4" (Bredl *et al.*, 1983). The solid line is the predicted BCS behavior. Data for other samples of CeCu_2Si_2 can fall even more slowly with increasing T_c/T . However, for the samples shown, the similarity of the temperature dependence of the data for $T_c/T > 5$ is striking.

latter due to the factor-of-2 difference in $\Delta C/\gamma T_c$, above $T_c/T=5$ the two sets of data appear *parallel*.

However, in describing their respective data, Bredl *et al.* and Ott *et al.* have taken quite different approaches. Bredl *et al.* (1983) ascribe the observed deviation of C_s for their samples from BCS behavior as likely due to gapless superconductivity. Ott *et al.* (1984e) claim their data at low temperatures follow a power law of the form $C_s(T)/C_n(T_c) \sim 2.8(T/T_c)^3$, which they find suggestive of *p*-wave superconductivity. Ott *et al.* find a better qualitative fit of their data to the Anderson-Brinkman-Morel state of ^3He , with strong-coupling corrections important, than to the Balian-Werthamer state of ^3He (see Leggett, 1975, for a discussion of these two states in ^3He). The arguments of Bredl *et al.* (1983) for gapless superconductivity in CeCu_2Si_2 obviously apply equally well to the data of Ott *et al.* on UBe_{13} , as is apparent in Fig. 10. How well do the data for CeCu_2Si_2 follow a T^3 power law? A $\log C_s(T)/C_n(T_c)$ vs $\log(T/T_c)$ plot is shown in Fig. 11 for CeCu_2Si_2 , sample 4 of Bredl *et al.* (1983). The C_s data for CeCu_2Si_2 indeed follow a power law close (but not equal) to T^3 . Although Ott *et al.* (1984e) do not show a plot like Fig. 11, their C_s data appear to follow a T^3 power law more closely ($T^{3.15 \pm 0.15}$) than those for CeCu_2Si_2 for $T_c/T < 7.5$. This is consistent with the more rapid fall in $C_s(T)/C_n(T_c)$ for UBe_{13} nearer T_c in Fig. 10 than seen in CeCu_2Si_2 . For $T_c/T > 7.5$, the C_s data for UBe_{13} appear to deviate from a T^3 power law, with the correct exponent being less than two, due to experimental error due to self-heating from the U.

The question of how significant this non-BCS behavior is remains a subject for discussion. As we have seen for CeCu_2Si_2 (see inset of Fig. 2) γ increases as T decreases. Ott *et al.* (1984e) speculate that γ increases with decreasing T for UBe_{13} as well, to explain their observation that

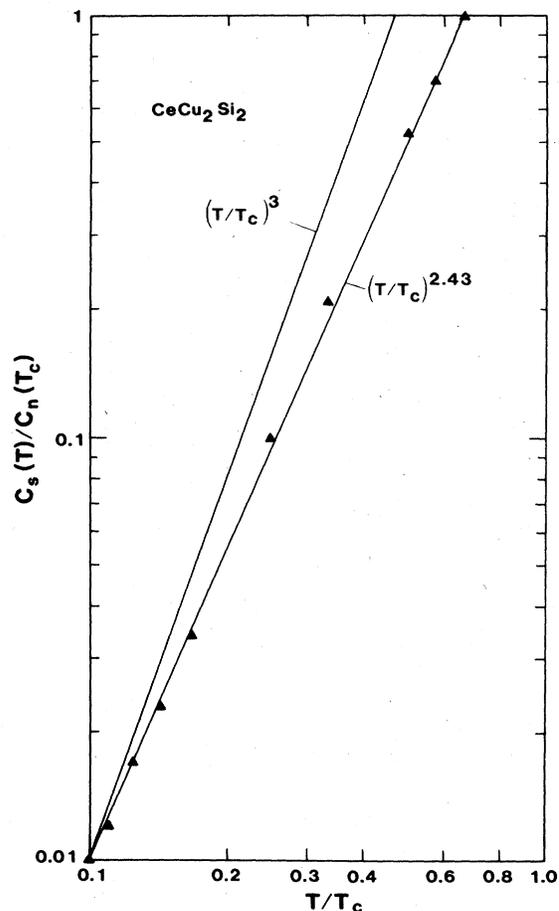


FIG. 11. Logarithm of the C_s data for CeCu_2Si_2 shown in Fig. 10 vs logarithm of T/T_c . The two lines shown correspond to two different power laws.

the entropy under the measured C curve in the superconducting state [$\equiv S_s(T_c) = \int_0^{T_c} C/T dT$] is 10% larger than that obtained in the normal state [$\equiv S_n(T_c)$] by extrapolating C_n below T_c to $T=0$ assuming a constant γ . [For a second-order phase transition, $S_n(T_c) = S_s(T_c)$.] Thus in order to have enough normal-state entropy below T_c in UBe_{13} to match the observed $S_s(T_c)$, γ must increase by about 20% below T_c , i.e., $S_n(T_c) = \int C_n^{\text{extrap}}/T dT$ with $\gamma(T) = 1.2\gamma(T=T_c) - 0.2\gamma(T=T_c)T$ for $T < T_c$.

Taking an increasing γ below T_c as given for both CeCu_2Si_2 and UBe_{13} , consider now what one would predict (ignoring the measurements for the moment) for the superconducting electronic specific heat $C_s(T)$. If $C_s(T)$ followed the BCS behavior, which assumed γ constant below T_c , then too little superconducting-state entropy would occur. Therefore, a positive deviation from the BCS prediction for $C_s(T)$ is to be expected when γ increases below T_c .

In conclusion, the observed sample dependence of $C_s(T)$ in CeCu_2Si_2 may be too large to explain by this qualitative argument. Moreover, the significance of the differing temperature dependences observed in $C_s(T)$ in

UBe₁₃ and CeCu₂Si₂ needs further theoretical work, remembering that the observed power law is only approximately T^3 . As pointed out recently by Volovik and Gor'kov (1984), a T^3 power law for the superconducting electronic specific heat may occur in a singlet, or BCS superconductor, as well as in a p -wave superconductor.

E. NpBe₁₃

Stewart *et al.* (1984b) reported that NpBe₁₃ (same structure as UBe₁₃, $d_{\text{Np-Np}} = 5.13 \text{ \AA}$) was the first (and so far only) magnetic heavy-fermion system to have the factor-of-5 to -10 rise in C/T below 10 K characteristic of CeCu₂Si₂ and UBe₁₃. The specific-heat data for NpBe₁₃, as well as for Np_{0.68}U_{0.32}Be₁₃ and UBe₁₃, are shown in Fig. 12. As Np is added to UBe₁₃, T_c is depressed, approximately 0.3 K/% Np (Smith *et al.*, 1984). As shown in Fig. 12, replacement of $\frac{2}{3}$ of the U by Np increases C/T significantly between 1 and 11 K. If one associates the rapid increase in C/T below 10 K with increasing electron-electron correlations, then at some point this increased interaction will lead to magnetism. There is a sharp increase in C/T for Np_{0.68}U_{0.32}Be₁₃ below 0.8 K (Stewart *et al.*, 1984b) that may be the onset of magnetic order. However, magnetic order is quite clearly present in pure NpBe₁₃ at 3.4 K (see Fig. 12). Although the nature of this magnetic ordering is not yet clear (spin-density wave, itinerant antiferromagnetism?) from the specific-heat, resistivity, and susceptibility results, neutron scattering work is underway.

The entropy associated with the magnetic transition, defined as

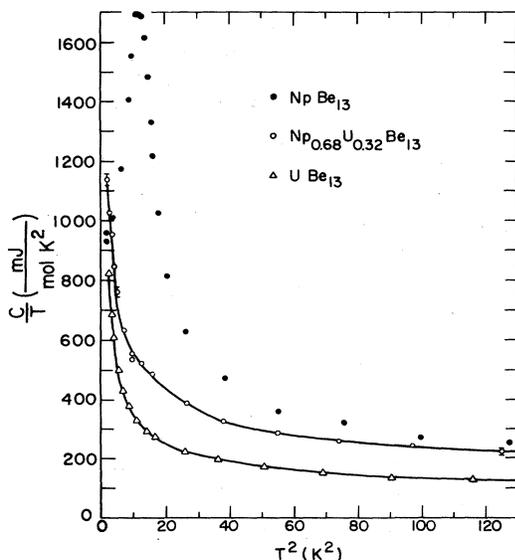


FIG. 12. Low-temperature specific heats of three different compounds: ●, NpBe₁₃; ○, Np_{0.68}U_{0.32}Be₁₃; △, UBe₁₃ (Stewart *et al.*, 1984b). The lines shown are guides to the eye.

$$\int_0^{11} (C^{\text{NpBe}_{13}}/T) dT - \int_0^{11} (1.13 C^{\text{Np}_{0.68}\text{U}_{0.32}\text{Be}_{13}}/T) dT$$

(the factor 1.13 scales the two specific heats to match at 11 K), is 2.3 J, or 40% of $R \ln 2$. Whether the entropy change is really positive depends on the correct background subtraction. It is interesting to speculate what γ would be in NpBe₁₃ without the magnetic order. By scaling low-temperature susceptibility values for NpBe₁₃/UBe₁₃, i.e., assuming χ/γ is constant, one would obtain a $\gamma(T=0)$ for nonmagnetic NpBe₁₃ over 3000. C/T for Np_{0.68}U_{0.32}Be₁₃ is 2100 at 0.4 K, although, as stated, this may be due to the onset of magnetic order. If one presumes that heavy-fermion systems like CeCu₂Si₂, UBe₁₃, and NpBe₁₃ have a generalized temperature dependence to their specific heat (Fig. 6) and differ only by some ratio (see summary, Sec. II.J below), then taking this ratio of C/T at 11 K for NpBe₁₃/UBe₁₃, well above the transition at 3.4 K, gives a ratio of about 2.1, implying that $\gamma(T=0)$ for "nonmagnetic" NpBe₁₃ would be 2300. What is indisputable is that the $\gamma(T=0)$ for hypothetical nonmagnetic NpBe₁₃ is significantly greater than the observed $\gamma(T=0)$ for magnetic NpBe₁₃ of 900, still a very large value. Smith *et al.* (1984) have found a nonmonotonic behavior of T_c with (Th) impurity in UBe₁₃; Ott *et al.* (1984b) have found two bulk superconducting specific-heat transitions in U_{0.9669}Th_{0.0331}Be₁₃; and Giorgi *et al.* (1984) have found a total disappearance of T_c in UBe_{12.94}Cu_{0.06}. Since these findings, the question of impurity effects on the magnetic transition in NpBe₁₃ has become of interest. Perhaps a slight substitution of Cu in the Be site in NpBe₁₃ would depress the magnetic transition, just as T_c is suppressed in UBe_{12.94}Cu_{0.06}. This would enable a determination of $\gamma(T=0)$ for nonmagnetic NpBe₁₃, since Stewart and Giorgi (1984) have shown that the specific heat above 1 K is the same in UBe₁₃ and UBe_{12.94}Cu_{0.06}.

F. U₂Zn₁₇, UCd₁₁, and NpSn₃

Recently, rhombohedral U₂Zn₁₇ (Ott *et al.*, 1984a; U-U distance equal to 4.39 Å; see Pearson, 1967, p. 1284) and UCd₁₁ (Fisk *et al.* 1984b; U-U distance of 6.56 Å; see Elliott, 1965, p. 293) were discovered to have large, temperature-independent γ 's (535 and 840, respectively) above magnetic transitions at 9.7 and 5.0 K, respectively. The electronic specific-heat data for single-crystal UCd₁₁ and polycrystalline U₂Zn₁₇ are shown together in Fig. 13, with the lattice contribution subtracted as discussed in the figure caption. UCd₁₁ has a constant γ to the limit of the data (up to 13 K), as does U₂Zn₁₇ (up to 16.5 K). If these γ values remained constant to higher temperatures, the electronic entropy would be enormous. The question of the temperature dependence of these γ values at higher temperatures is of interest.

The transitions observed in the two systems are different in several ways, in addition to the different magnitudes of ΔC observed: 5.65 J/mol U K for U₂Zn₁₇ and 7.55 J/mol U K for UCd₁₁. Above the magnetic ordering

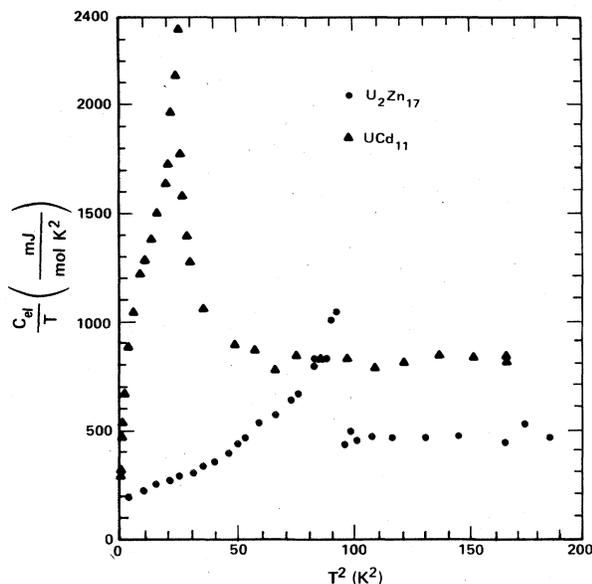


FIG. 13. Low-temperature electronic specific heats: ●, U_2Zn_{17} (Ott *et al.*, 1984a); ▲, UCd_{11} (Fisk *et al.*, 1984b). The lattice contribution subtraction for U_2Zn_{17} was calculated using specific-heat data for Th_2Zn_{17} and the assumption that $C_{lattice}$ is the same for both compounds. Since C for UCd_{11} obeyed Eq. (1) above 8 K, the β thus obtained was used to calculate $C_{lattice}$ for UCd_{11} . The data for U_2Zn_{17} have been normalized per f atom mol, i.e., a mole here contains 1 U atom and 8.5 Zn atoms, times Avogadro's number.

temperature T_m , the specific heat for UCd_{11} shows signs of precursor, or short-range-order, effects, whereas the transition for U_2Zn_{17} is quite sharp, with no apparent ordering above T_m . The possibility of lattice disorder in the low-temperature-grown (400°C) phase of UCd_{11} (cubic $BaHg_{11}$ type) causing this smeared transition cannot be ruled out. The entropy associated with the two transitions is also different, defining the net change in entropy as $\Delta S = \int (C_{measured}/T)dT - \int (C_{extrap}/T)dT$, where C_{extrap} is the extrapolation of the higher-temperature data from above the transition to $T=0$. For U_2Zn_{17} , ΔS is -1.37 J/mol U K, while for UCd_{11} ΔS is $+1.63$ J/mol U K, the latter positive fraction (0.28) of $R \ln 2$ indicating local moment behavior. Although measurements of the specific heat of UCd_{11} in an 11-T applied field to suppress the magnetism show that γ remains relatively (better than 15%) constant to lower temperatures (5 K), the possibility of a slightly temperature-dependent γ below 8 K causing the entropy to balance cannot be ruled out. Moreover, the possibility that the magnetic ordering is of the induced type, as in Pr_3Tl (Andres *et al.*, 1972), could also account for the low entropy. Neutron scattering measurements would clearly be useful.

The last point of comparison between U_2Zn_{17} and UCd_{11} is the value of the $\gamma(T=0)$ measured below T_m . Data below 1 K give $\gamma(T=0)=250$ ($\Delta\gamma=590$) for UCd_{11} and 198 ($\Delta\gamma=337$) for U_2Zn_{17} . The fractional change in

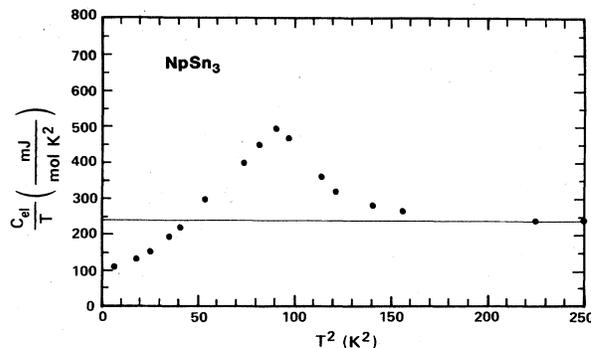


FIG. 14. Specific heat of $NpSn_3$, after Trainor *et al.* (1976a), who had plotted C/T vs T , not T^2 as here.

γ for the two compounds (70% for UCd_{11} and 63% for U_2Zn_{17}), or the amount of Fermi surface removed, is remarkably similar. (For a discussion of Fermi-surface effects, see Trainor *et al.*, 1976a or Herring, 1966.)

It is interesting at this point to compare the specific-heat results for a similar system, $NpSn_3$, although the γ is "only" 242. These data taken by Trainor *et al.* (1976a) are shown in Fig. 14 as $C_{electronic}/T$ vs T^2 with the lattice contribution subtracted away. $T_{max}=9.5$, a value similar to that for U_2Zn_{17} . The value of ΔS associated with the transition is, within the error of their data, stated to be 0, while the stated $\Delta\gamma$, 154, is 64% of γ above the transition, again like UCd_{11} and U_2Zn_{17} . The extremely broad tail above T_{max} for the specific heat of $NpSn_3$ may also be at least partly due to sample inhomogeneity (Trainor *et al.*, 1976a). Qualitatively the specific heats of U_2Zn_{17} , UCd_{11} , and $NpSn_3$ at low temperatures appear similar, despite the factor-of-3.5 difference in their $\gamma(T > T_m)$ values. Therefore, if one is seeking a magnetic analog to superconductivity in UBe_{13} , the behavior of $NpBe_{13}$ seems the best choice for comparison, while U_2Zn_{17} and UCd_{11} seem quite ordinary in comparison, due to their lack of temperature-dependent γ 's and their similarities to the nonheavy system $NpSn_3$.

G. $CeAl_3$ and $CeCu_6$

Until the discovery of $CeCu_2Si_2$, $CeAl_3$ may have been the most studied Ce compound, unfortunately with some of the same sample dependence of the properties as $CeCu_2Si_2$. $CeAl_3$ has the same DO19 hexagonal (Ni_3Sn -type) structure as UPt_3 , with $d_{Ce-Ce}=4.43$ Å. $CeAl_3$ seems clearly to be a classic case of a material for which years of false conclusions concerning its intrinsic properties were caused by lack of sufficient materials characterization in the beginning. In the specific-heat work of Mahoney *et al.* (1974), not one but *three* sizable peaks due to second phases are present below 10 K, leading to incorrect conclusions. The peak at 4 K is due to (Berton *et al.*, 1977) magnetic ordering in a small amount of $CeAl_3$ second phase. This peak can be avoided by proper sample preparation. The two peaks at 2.5 and 6 K are

probably caused by magnetic ordering in small amounts of $\text{Ce}_3\text{Al}_{11}$ second phase (Chouteau *et al.*, 1978.) It is not certain that the properties of CeAl_3 below 1 K are not also affected by contributions from these second phases. However, such low-temperature properties seem relatively reproducible, suggesting that measurements in CeAl_3 below 1 K are of intrinsic properties. A peak in C for CeAl_3 at 25 K (which is probably intrinsic) was identified by Mahoney *et al.* as a Schottky anomaly, with an entropy of approximately $R \ln 3$ associated with it.

Only two measurements of the specific heat of CeAl_3 below 1 K have been published—the work of Andres *et al.* (1975) and the subsequent work of Benoit *et al.* (1981). (See also Flouquet *et al.*, 1982, for discussion of the data by Benoit *et al.*) Andres *et al.* measured the specific heat of CeAl_3 down to ~ 0.015 K and found the first heavy-fermion system, with $\gamma = 1620$ between 0.015 and 0.150 K. The data of Benoit *et al.* are consistent with this result. Coupled with a large but finite susceptibility at low temperatures and the temperature dependence of the resistivity data, this result has led to the description of CeAl_3 as a Fermi liquid. (For a discussion, see Lawrence *et al.*, 1981, p. 32.) Whether CeAl_3 is a “Kondo lattice” or intermediate valent remains in question; see Edelstein and Koon (1983) and Lawrence *et al.*, (1981, p. 9) for both sides of the discussion.

The low-temperature data of Benoit *et al.* and, above 1.5 K, those of Ott *et al.* (1984b) compared to those of Berton *et al.* are shown in Figs. 15 and 16, plotted as C/T vs T^2 . At low temperatures, but above 0.4 K, we see a rising C/T value for CeAl_3 similar to that shown in the inset of Fig. 2 below 1 K for CeCu_2Si_2 . The peak in the specific heat near 0.35 K (Berton *et al.*, 1981) in Fig. 15 correlates with anomalies in other properties as well—magnetoresistance (Edelstein *et al.*, 1977; Remenji *et al.*, 1983), thermal expansion (Ribault *et al.*, 1979), and elastic constants (Niksch *et al.*, 1980). The behavior below this anomaly is then, as mentioned above, taken to be characteristic of a Fermi liquid (see, for example, Niksch *et al.*, 1980). Such a description, discussed more fully in Sec. IIJ, is equally applicable to other systems, including CeCu_2Si_2 .

The peaks (believed to be at least partially due to $\text{Ce}_3\text{Al}_{11}$; Chouteau *et al.*, 1978) in C at 2.5 and 6 K in CeAl_3 are not really evident in the higher-temperature

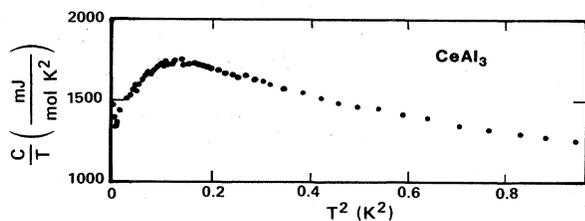


FIG. 15. Low-temperature specific heat of CeAl_3 (Benoit *et al.*, 1981).

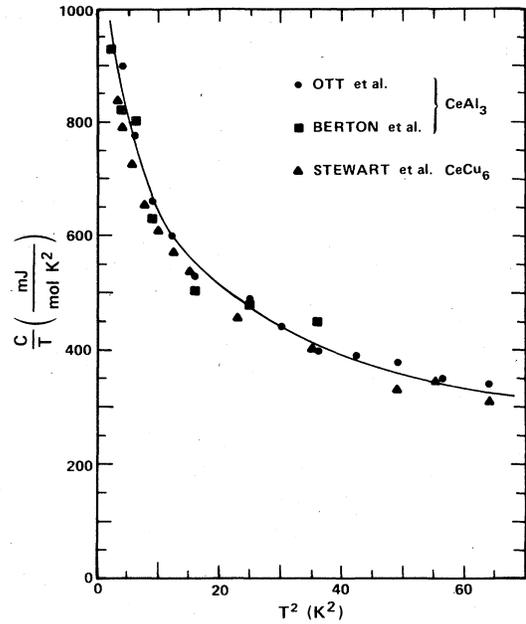


FIG. 16. Higher-temperature specific-heat data: ●, CeAl_3 (Ott *et al.*, 1984b); ■, CeAl_3 (Berton *et al.*, 1977); ▲, CeCu_6 (Stewart, Fisk, and Wire, 1984c). The line is drawn through both sets of CeAl_3 data to serve as a guide to the eye.

data plotted as C/T vs T^2 in Fig. 16, just as the peaks in CeCu_2Si_2 and UBe_{13} are not evident on such a plot. The peak at 2.5 K in the data of Berton *et al.* (1977) is only about 0.3 J/mol K in height, as is their peak at 6 K. Even if these low-temperature peaks are intrinsic, they are not comparable in size to those observed in CeCu_2Si_2 or even in UBe_{13} , nor is the (believed to be) intrinsic peak in C/T observed in Fig. 15 at $T^2 = 0.15 \text{ K}^2$ ($\Delta C \sim 0.1 \text{ J/mol K}$). Thus, while similar models are used to describe CeAl_3 (Niksch *et al.*, 1980) and CeCu_2Si_2 (Bredl *et al.*, 1983), differences (e.g., in ΔC) do exist and should be considered in any unified picture.

In the recently discovered heavy-fermion system CeCu_6 (Stewart, Fisk, and Wire, 1984), with orthorhombic structure, Ce-Ce distance of 4.83 Å (see Cromer *et al.*, 1960), the specific heat does not show spurious second-phase anomalies, but is otherwise quite comparable in magnitude to the data for CeAl_3 (see Fig. 16). When the data are plotted as C vs T , rather than anomalies a slight shoulder is observed below 7 K, with, however, no increase in C as T is lowered, contrary to what is observed in CeCu_2Si_2 and UBe_{13} , Figs. 1 and 5. The slight ($< 10\%$) difference in C/T for CeCu_6 and CeAl_3 may be due to the increased entropy in CeAl_3 from the magnetic second phases. By extrapolation, Stewart, Fisk, and Wire (1984) predicted a $\gamma(T=0)$ for CeCu_6 similar to that of CeAl_3 , approximately equal to 1600. This has since been confirmed by measurement (Ott *et al.* 1984d), with $\gamma(T \rightarrow 0) = 1550$. It is interesting to note that resistivity measurements (Ott

et al., 1984d) in the same temperature range as for CeAl₃ do not find a T^2 term, used to argue for the Fermi-liquid model in CeAl₃. In any case, the comparability of the three Ce heavy-fermion systems is less than perfect, leaving questions of interpretation still open.

H. YbCuAl and Pu_{0.7}U_{0.3}Al₂

These two systems are discussed briefly here solely to illustrate how “low- γ ” systems differ from those heavy-fermion systems which have rapid temperature dependence in their C/T values below 10 K. This is not to deny that both systems are interesting in their own right. YbCuAl is another system thought to be mixed valent (Mattens *et al.*, 1980), but only barely so (Pott *et al.*, 1981). See also Lawrence *et al.* (1981) for a discussion. Pu_{0.7}U_{0.3}Al₂ was characterized in a study (Trainor *et al.*, 1976b) of the spin-fluctuation properties of UAl₂. The compound UAl₂ was found by Trainor *et al.* (1975) to show a $T^3 \ln T$ term in C just as in the case of UPt₃, with a $\gamma(T=0) \simeq 140$. The data for YbCuAl are shown in Fig. 17; the data for Pu_{0.7}U_{0.3}Al₂ are similar, i.e., they show no $T^3 \ln T$ term, with both γ 's equal to 260. Clearly these materials follow Eq. (1) at low temperatures like normal metals, albeit with much larger γ values.

YbCuAl has anomalies in its thermal expansion (at 24 K) and specific heat (at 28 K) which, coupled with other measurements, have been taken to mark the transition from local moment behavior, $T > T_{\text{anomaly}}$, to Fermi-liquid behavior $T < T_{\text{anomaly}}$ (Mattens *et al.*, 1980). If the specific heat γ is taken to scale as T_{anomaly}^{-1} (Lawrence *et al.*, 1981), then the factor-of-5 difference in $\gamma(T=0)$ between YbCuAl and CeCu₂Si₂ is consistent with the ratio of the ΔC anomaly temperatures (28/3.5). Such a comparison between YbCuAl and CeAl₃ fails if the peak in C at 25 K in the latter is used as the comparable feature. More about this scaling by a characteristic temperature will appear in the sections on susceptibility (III.A) and resistivity (IV). Scaling remains at best, how-

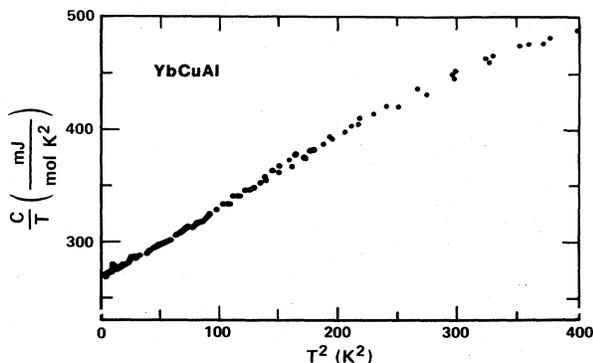


FIG. 17. Low-temperature specific heat of YbCuAl, after Mattens *et al.* (1977). This straight-line behavior of C/T vs T^2 gives γ as the intercept on the ordinate and β as the slope.

ever, a phenomenological means of comparing systems, without explaining the underlying physical processes. By measuring the entropy involved in ΔC , Pott *et al.* (1981) identify the peak in C for YbCuAl as being $R \ln 9$, i.e., due to the “freezing out” of the degrees of freedom for the Hund’s rules ground states of Yb³⁺ ($J = \frac{7}{2}$) and Yb²⁺ ($J = 0$). How this process creates quasiparticles of high effective mass (high γ values) in some systems and not others remains uncertain. (No large valence change is found in YbCuAl below 300 K.) This phenomenological approach does not explain the behavior of the two samples of CeCu₂Si₂ shown in Figs. 1 and 2, where the samples have the same $\gamma(T=0)$, ~ 1100 , but two different entropies and T_{anomaly} 's. Moreover, no large $R \ln(2s+1)$ anomaly is seen in the entropy of CeAl₃ below 10 K. Therefore, simple scaling laws may be appealing, but fail to explain all the observed data.

I. Specific heat in high magnetic field; discussion of models

To discuss the effects of a magnetic field on the specific heat, we need to consider the components and their relative contributions to the large γ values of these systems, i.e., $N(0)$ and $1+\lambda$. If λ is taken as small, then the bandwidths needed to make $N(0)$ large enough are only tens of degrees Kelvin. For example, in UBe₁₃, $\gamma(T=0)$ is 1100; $N(0)(1+\lambda)$ is then 465 states/eV U atom (assuming the Be atoms contribute nothing). In a model one-electron band shaped like a square wave, height (in states/eV atom) is equal to $N(0)$ times width (in eV, 25 meV = 300 K) must equal 1 state/atom. With $\lambda=0$, the width is then 26 K. If an f -electron bandwidth of 0.1 eV is assumed, then $\lambda=46.5$. Under either extremum, or in between, magnetic field is expected to change the specific heat, either by affecting the electron-electron interactions (λ large), or by broadening the very narrow electronic density of states (λ small), or by both mechanisms in the intermediate case. No exact theory exists for the behavior of the specific heat in a high magnetic field in the case of strong electron-electron interactions. If the mass enhancement of the electrons through such a large λ is viewed in the paramagnon model of long-range, short-time-scale fluctuations into a spin-aligned state, then an applied field would be expected to quench these fluctuations.

Since the specific heat of UPt₃ actually fits the temperature dependence ($T^3 \ln T$) proposed for a spin fluctuator (see Figs. 8 and 9), we consider the normal specific heat of this system in a magnetic field first. Data in 0 and 11 T (Stewart *et al.*, 1984d) are shown in Fig. 18. The data above 7 K indeed show a suppression of C with applied field, while the data at lower temperature show an anomalous increase with field. This anomalous behavior is not understood; further experiments in higher fields and with other crystal orientations are needed.

The specific heat of UBe₁₃ in 0 and 11 T (Stewart *et al.*, 1984a) is shown in Fig. 19. There is a sizable in-

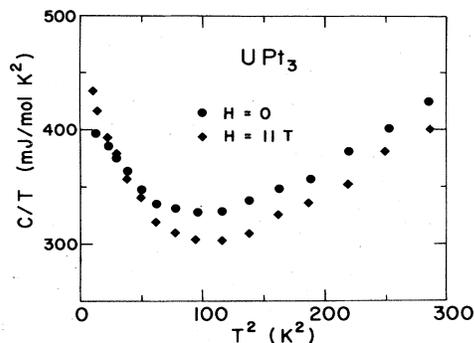


FIG. 18. Specific-heat single crystal for UPt_3 , in zero field (\bullet) and 11 T (\blacklozenge) by Stewart *et al.* (1984d). The crossover at 7 K is not understood at present.

crease with field which has a maximum (+9%) around 3 K, with a decreasing effect below and above 3 K. (Greater than 2% change with field is deemed outside the limits of error.) The authors discussed a simple very-narrow-band model in which λ is small enough to concentrate on the behavior of $N(0)$ with field. Such an heuristic model, with a bandwidth tens of K wide that is broadened by field ($1 \text{ T} \approx 0.7 \text{ K}$ from $kT = g\mu_B H$) and temperature, predicted for UBe_{13} an increase in the specific heat γ above 3.5 K which reaches a maximum at 6 K and falls off at higher temperatures; the model further predicted a net decrease in C/T with 11 T below 3.5 K. Although the details of this model are not obeyed by the data, certain broad features are the following: increase in C/T with H to a maximum at 3 K; a decrease in the observed $\Delta C(H)/T$ going to zero at higher temperatures (which, due to entropy considerations, must be true for any model); and a lessening of the increase also below the maximum.

The specific heat of CeCu_6 in a magnetic field (Stewart, Fisk, and Wire, 1984) has similarities to that of UBe_{13}

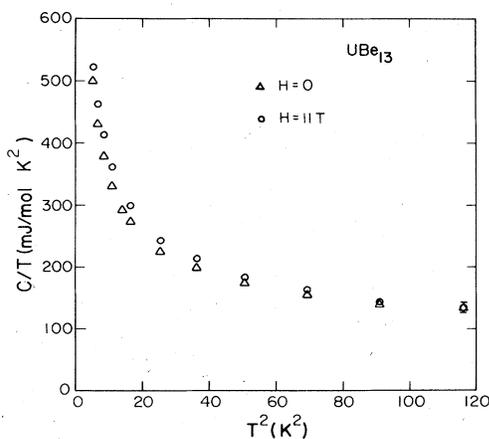


FIG. 19. Specific heat of single-crystal UBe_{13} in zero field (\triangle) and in a field of 11 T (\circ) by Stewart *et al.* (1984a).

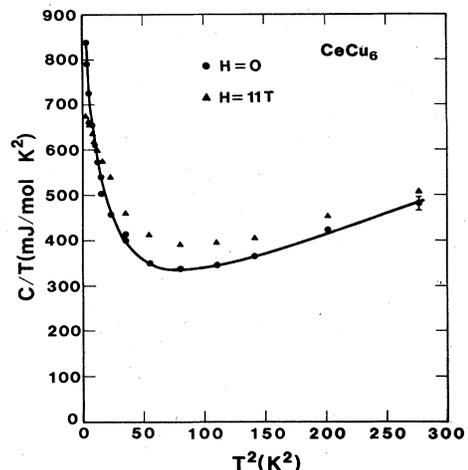


FIG. 20. Specific heat for CeCu_6 in zero field (\bullet) and 11 T (\blacktriangle). The line drawn through the zero-field data is a guide to the eye.

(see Fig. 20), with a crossover around 3.5 K and a decrease in C with 11 T of 19% at 1.8 K. As a final comparison, consider the specific heat of a nonsuperconducting single crystal of CeCu_2Si_2 in 0 and 10 T, Fig. 21 (Stewart, Fisk, and Willis, 1983). The behavior is remarkably like that observed for CeCu_6 —a crossover at 3 K, a 31% decrease in C at 1.5 K in 10 T, and an increase in C/T with field that decreases at higher temperatures. All of these field data behave qualitatively like the predictions

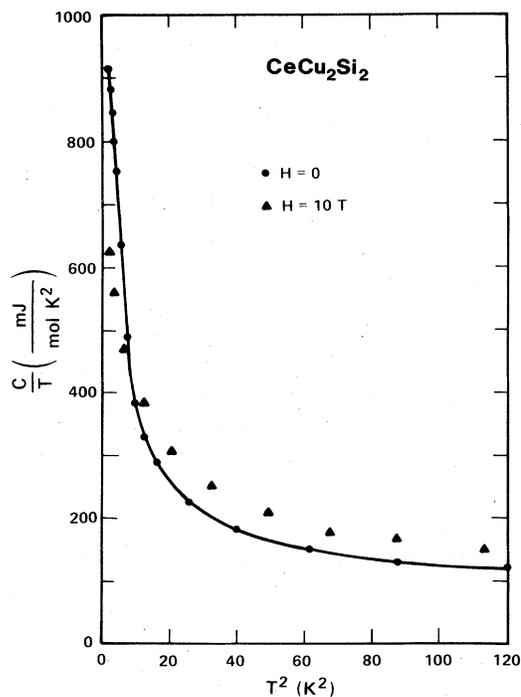


FIG. 21. Specific heat of nonsuperconducting single crystals of CeCu_2Si_2 in zero field (\bullet) and 10 T (\blacktriangle) by Stewart, Fisk, and Willis (1983).

of the very-narrow-band model proposed for UBe_{13} . Whether this behavior of the specific heat with field could also be explained by a theory based on large λ remains in question. (For discussion, see Stewart *et al.*, 1984a and Vollhardt, 1984.) Any discussion of the changes of the low-temperature specific heat with field needs to include an explanation of the low-temperature anomaly in C in these systems, visible when C is plotted versus T (Figs. 1 and 5). We have seen that this anomaly varies between materials and even samples (i.e., in CeCu_2Si_2). How should field affect it? In the narrow-band model for UBe_{13} of Stewart *et al.* (1984a), since the parameters chosen give the correct behavior of γ in zero field, an "anomaly" similar to that experimentally observed in UBe_{13} (Fig. 5) comes simply from the model as increasing temperature broadens the very narrow structure at the Fermi energy (Mueller, 1983).

Thus, as a phenomenological starting point, a narrow-band model has three virtues: it gives a zero-field specific-heat anomaly, it qualitatively predicts the behavior of specific heat with field, and it is conceptually simple.

However, one could also view CeCu_2Si_2 , CeCu_6 , and UBe_{13} as "Kondo lattices" and say that the variation from sample to sample and system to system of the entropy under the anomaly (seen when plotting C vs T) is some complication, as yet not understood, of the Kondo effect present in concentrated systems. If these systems are viewed as Kondo systems, then observations can be made about the behavior of their specific heat in a magnetic field by analogy with more dilute Kondo systems. In the rather thoroughly studied $\text{La}_{1-x}\text{Ce}_x\text{Al}_2$ system, specific-heat results in a field exist for a number of compositions. For $\text{La}_{0.9936}\text{Ce}_{0.0064}\text{Al}_2$, Bader *et al.* (1975) showed that the peak in ΔC ($C_{\text{La}_{1-x}\text{Ce}_x\text{Al}_2} - C_{\text{LaAl}_2}$) moves from ~ 0.15 K upwards monotonically with field to ~ 1.3 K in 3.8 T, and also grows in size. This latter increase in ΔC_{max} is due to removal of the Zeeman degeneracy of the ground-state doublet of the Ce ions by the applied field. Thus the peak in ΔC in zero field becomes quite similar in field to a Schottky peak for a two-level system (see Steglich, 1976 for a complete discussion). Below a certain temperature that increases with field (~ 0.5 K in 3.8 T), the change in ΔC with field in $\text{La}_{0.9936}\text{Ce}_{0.0064}\text{Al}_2$ is negative. This behavior of the specific heat with field in a dilute Kondo system is not qualitatively dissimilar to that observed for CeCu_2Si_2 , UBe_{13} , and CeCu_6 . In the Ce systems a decrease in C at lower temperatures with field is observed, while at higher temperatures an increase is observed. For UBe_{13} only an increase is seen. To carry the analogy further, samples of CeCu_2Si_2 with *less* entropy under the anomaly at 2 K should show a *greater* increase in field, since more Zeeman degeneracy remains to be removed. Taken to sufficiently high temperatures, such measurements should produce $R \ln 2$ of entropy under the anomaly in field. The crossover observed in Fig. 21 around 3 K for the sign of the change of C with field in CeCu_2Si_2 should move upward in higher field.

Although both the analogy with dilute Kondo systems and the narrow-band model predict this crossover to move higher in temperature with field, differences do exist which may serve to provide direction for further work. In any case, neither picture serves as more than a phenomenological starting point for furthering understanding, since both are too simplistic at the present time.

J. Summary

We have seen three types of temperature dependence in the specific heat of heavy-fermion systems.

(1) In UPt_3 , C has the normal temperature dependence of a metal, $\gamma T + \beta T^3$ [Eq. (1)] plus the spin-fluctuation term $T^3 \ln T$. This dependence is found also in UAl_2 (Trainor *et al.*, 1975; Stewart, Giorgi, Brandt, Foner, and Arko, 1983), where $\gamma(T=0) = 142$, and in TiBe_2 (Stewart *et al.*, 1982), where $\gamma(T=0) = 56$ mJ/mol Ti K². Thus the superconductivity in UPt_3 (Stewart *et al.*, 1984c) in combination with the spin fluctuations is unusual, as well as the large γ , but the temperature dependence is not a characteristic unique to heavy-fermion systems.

(2) Similarly U_2Zn_{17} and UCd_{11} have a normal-metal temperature dependence above their magnetic ordering temperature, and are distinguishable from nonheavy systems, e.g., NpSn_3 , solely by their large γ values, 535 and 840, respectively.

(3) The remaining heavy-fermion systems, CeCu_2Si_2 , UBe_{13} , NpBe_{13} , CeAl_3 , and CeCu_6 , all have in common a rapid rise in C/T below 10 K. Due to the observance of large ΔC anomalies at the superconducting T_c in CeCu_2Si_2 and UBe_{13} , we presume this temperature-dependent C/T is really $\gamma \propto N(0)(1 + \lambda)$ for all these five systems. The temperature dependence of C/T , i.e., γ , for two representative systems, UBe_{13} and CeCu_6 , is shown in Fig. 22 by plotting $\ln C'/T$ vs $\ln T$, where the high-temperature ($\gamma T + \beta T^3$) behavior has been subtracted. We see that for UBe_{13} (and similarly for CeCu_2Si_2 ; see Fig. 6) $C/T (= \gamma)$ goes as $T^{-1.6}$ below 8 K, while for CeCu_6 (and CeAl_3 ; see Fig. 16) it varies from $T^{-0.88}$ to $T^{-1.8}$ over the same temperature range. This huge increase in γ at low temperatures is quite remarkable and remains to be explained.

As has been pointed out (Stewart *et al.*, 1984a), both the zero field and (qualitatively) 11-T C/T data for UBe_{13} obey the behavior below 10 K expected from a simple noninteracting narrow band of electrons, with width tens of K, which is broadened by increasing either temperature or field. This observation, also roughly valid for CeCu_2Si_2 , CeAl_3 , and CeCu_6 in 0 and 11 T, as discussed in Sec. II.I, does not address the mechanism by which this "Fermi liquid" of heavy electrons is formed. Andres *et al.* (1975) explained the low-temperature behavior of CeAl_3 as due to coherence among $4f$ virtual bound states, forming "a band of collective states with an extremely high density. . . to which the Fermi energy is pinned at low temperatures." The width of these virtual $4f$ states

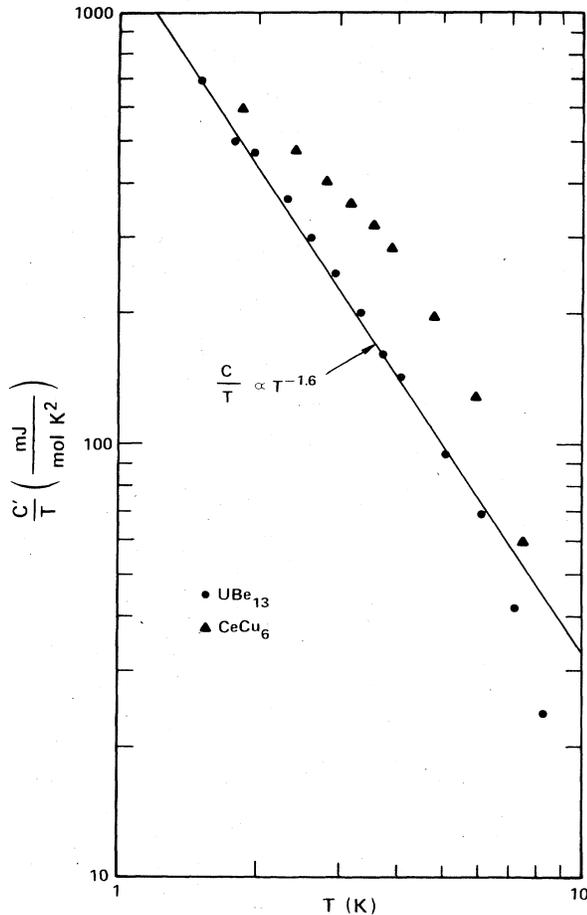


FIG. 22. Log-log plot of the difference between the measured specific heat at low temperatures minus the extrapolation of the high-temperature data vs T , i.e., $\log C_{\text{upturn}}/T$ vs $\log T$: ●, UBe_{13} (Stewart *et al.*, 1984a); ▲, CeCu_6 (Stewart, Fisk, and Wire, 1984). The upturn in C/T in UBe_{13} has a temperature dependence given by $C/T \sim T^{-1.6}$ over a fairly wide temperature range, while the upturn in CeCu_6 has a varying temperature dependence.

was calculated to be tens of degrees K (Andres *et al.*, 1975).

Another possible approach is suggested by the unresolved question of valency, or number of f electrons in several of these systems. For example, CeCu_2Si_2 (Franz *et al.*, 1978) and CeAl_3 (Lawrence *et al.*, 1981) are both thought to be almost but not exactly, integral valent. Brinkman and Rice (1970) predict a large enhancement in the effective mass m^* [$m^*/m = 1 + \lambda$ in Eq. (2)] just near integral valency. This has been used by Ott *et al.* (1984e) as a possible explanation of the heavy-electron Fermi liquid that forms in UBe_{13} at low temperature.

A third possible explanation involves the Kondo effect and was suggested to explain the huge single-ion value of γ ($=1740$) for pure CeAl_2 extrapolated in the $\text{La}_{1-x}\text{Ce}_x\text{Al}_2$ system (Bredl *et al.*, 1978). Bredl *et al.* ex-

plain the high γ (both observed and extrapolated) in $\text{La}_{1-x}\text{Ce}_x\text{Al}_2$ as a "Fermi-liquid effect, i.e., the conduction electrons try to [compensate] the spin (Kondo effect) which brings up their heat capacity." Presumably, some such model could be applied to explain the specific heats of CeCu_2Si_2 , UBe_{13} , NpBe_{13} , CeAl_3 , and CeCu_6 , which all have anomalies (admittedly varying in size) in C between 2 and 7 K (except NpBe_{13} , where the anomaly may be obscured by the magnetic transition at 3.4 K) called "Kondo-like" in the case of CeCu_2Si_2 .

The question of why CeCu_2Si_2 , UBe_{13} , and UPt_3 are superconductors, and CeAl_3 , CeCu_6 , and UAl_2 are not, while NpBe_{13} is magnetic, seems inexplicable based on comparisons of the specific heat at low temperatures.

III. MAGNETIC PROPERTIES

A. Susceptibility

All of the eight heavy-fermion systems have important similarities in their magnetic susceptibilities. These similarities include a large temperature dependence which, near room temperature, obeys qualitatively the Curie-Weiss law $\chi \propto (T + \Theta)^{-1}$, $\Theta < 0$; large ($> 2\mu_B$) effective moments deduced from the Curie-Weiss behavior; and quite large values of χ at low temperatures. These values [$\chi(T=0)$, μ_{eff} , and Θ] are given in Table III; the susceptibility data for each of the eight heavy-fermion systems from which these values were derived are shown in Figs. 23–33, with the ordering of the systems in the table and figures the same as in Sec. II. As can be seen in the fig-

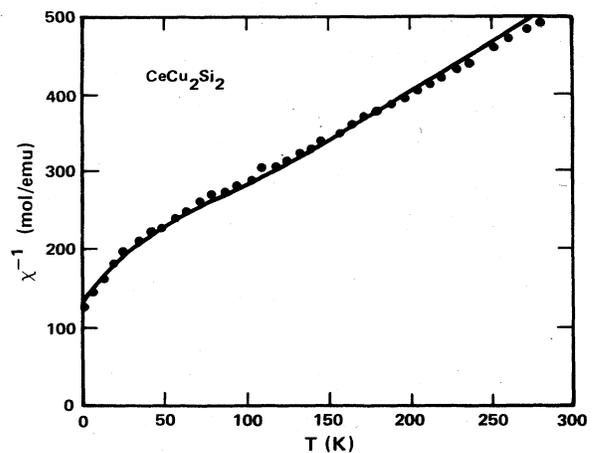


FIG. 23. Inverse susceptibility for CeCu_2Si_2 by Sales and Viswanathan (1976). The solid line shown was a crystal-field model to fit the data, with parameters in disagreement with later neutron work by Horn *et al.* (1981). Specifically, Horn *et al.* assigned a splitting of 140 and 354 K to the two excited doublets, while Sales and Viswanathan used splittings of 13 and 310 K to fit their susceptibility data. Thus assigning crystal-field level schemes without neutron data is seen to be potentially inaccurate.

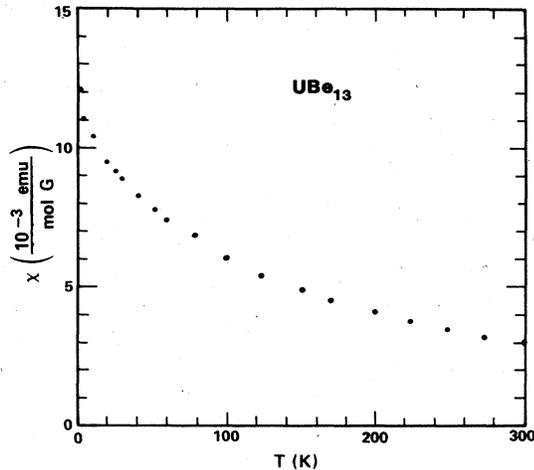


FIG. 24. Magnetic susceptibility vs temperature for UBe_{13} (Brodsky and Friddle, 1974). These data are similar in their temperature dependence, but not their magnitude, to data for NpBe_{13} .

ures, all eight systems follow a Curie-Weiss law (χ^{-1} linear in T) at higher temperatures, with effective moments over $2\mu_B$ and a negative intercept on the temperature axis ($\equiv \Theta$). Let us now concentrate on the parameters in Table III.

1. Sample dependence

The reported susceptibility values for a given compound vary widely, as can be seen in Table III. Just as in Sec. II, CeCu_2Si_2 has the largest sample dependence (at least a factor of 2) in its susceptibility. For the other

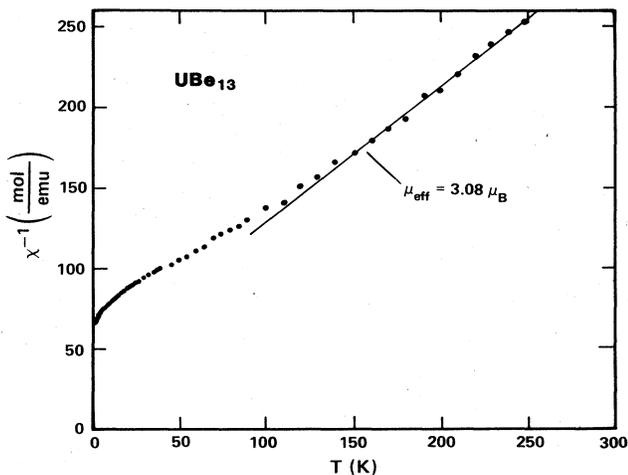


FIG. 25. Inverse magnetic susceptibility for UBe_{13} (Ott *et al.*, 1984c), with the straight line showing the Curie-Weiss behavior at higher temperatures.

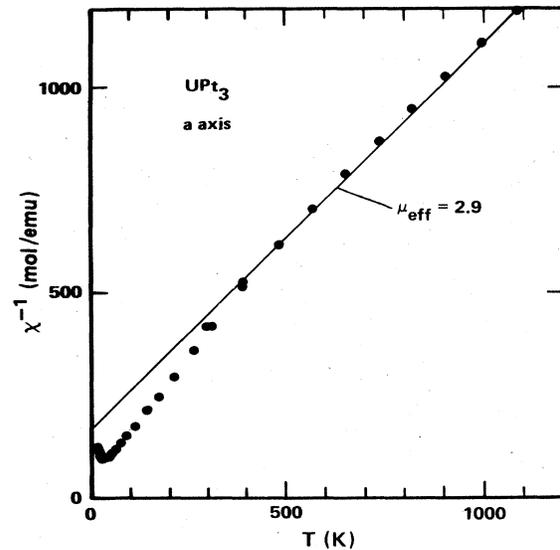


FIG. 26. Inverse susceptibility along the a axis for UPt_3 by Frings *et al.* (1983). Note that data up to only 300 K would give an incorrect μ_{eff} of $2.4\mu_B$.

compounds, sample-to-sample variation in $\chi(T=0)$ is limited to 30%. The variation in the values of μ_{eff} and Θ is also considerable, owing at least partly to crystal-field-caused curvature in the χ^{-1} vs T data that is not finished by 300 K, i.e., the higher-temperature derived values should normally be closer to the correct value. It is not clear, however, in the case of UBe_{13} which value to use for μ_{eff} and Θ .

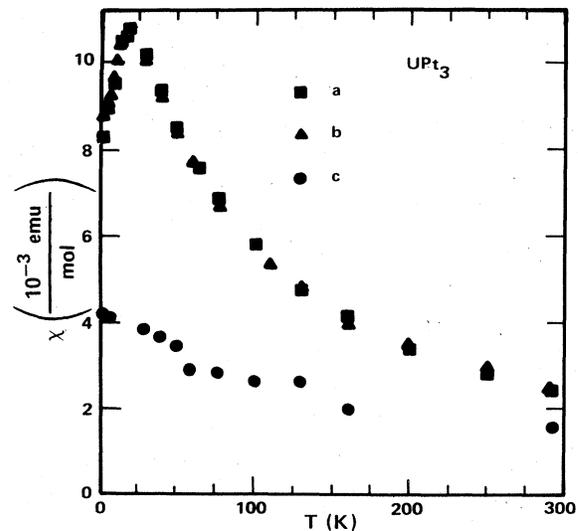


FIG. 27. Susceptibility data on a single crystal of UPt_3 by Frings *et al.* (1983). Note the large anisotropy between the basal plane (a, b axes) and the c axis, and the peak in the susceptibility at 19 K.

TABLE III. Magnetic parameters for the heavy-fermion systems. μ_{eff} for an f^1 state is $2.54\mu_B$, for f^2 , $3.58\mu_B$, and f^3 is $3.26\mu_B$. S.C. means superconducting.

System	μ_{eff}	$\chi(T=0)$ (10^{-3} emu/molG)	Curie-Weiss Θ (K)	Reference
CeCu ₂ Si ₂	2.62	8	-164	Sales and Viswanathan, 1976
CeCu ₂ Si ₂	2.61	6.8 ($T=1.4$)	-175	Batlogg, 1984a
Cu flux in basal plane ⊥ basal plane	2.54	12.5	-50	
In flux in basal plane ⊥ basal plane	2.75 2.54	11.8 19.8	-175 0	Batlogg, 1984a
S.C. crystal in basal plane crystal ⊥ basal plane	4.2 8.4	4.2 8.4		Assmus <i>et al.</i> , 1984 for T at 3.5 K where there is a rather flat peak
Non-S.C. crystal in plane ⊥ plane		~10 ~20		
CeCu ₂ Si ₂	2.68	6.5 ($H \geq 15$ T) at 4.2 K 8.7 (4.2 K, low H) 12.7 (low H , $T \sim 1.5$ K)	-140	Lieke <i>et al.</i> , 1982, sample 7
UBe ₁₃	3.4 2.99 3.52 2.6 ^a 3.08 ^a	13.5 12.1 ~16.6 (at 1.4 T) 15 (at T_c^+) 15	-70 -68 -98 -53 -53	Troc <i>et al.</i> , 1971, to 1000 K Brodsky and Friddle, 1974, to 300 K Bucher <i>et al.</i> , 1975, to 60 K Ott <i>et al.</i> , 1983 Ott <i>et al.</i> , 1984c Schneider and Laubschat, 1981
UPt ₃	2.61 (between 110 and 210 K) 2.9	6.9	?	Frings <i>et al.</i> , 1983
basal plane ⊥ basal plane		8.3 4.2		
UPt ₃ poly	3.0±0.1	8.2	~-200	Frane, 1983 and Frings <i>et al.</i> , 1983
NpBe ₁₃	2.76	43.2 (2 K) 56 (0-2 K)	-42	Brodsky and Friddle, 1974, to 300 K Stewart <i>et al.</i> , 1984b
U ₂ Zn ₁₇	4.5 ^b	12.5 ^b (at χ_{max} , $T \sim 12$ K) 9 ^b ($T \rightarrow 0$) 12.4 ($T = 4.2$ K)	~-250	Ott <i>et al.</i> , 1984a (200-300 K)
UCd ₁₁	3.45	3.84 (constant below 5 K) 45 (4.2 K)	-23	Misuik <i>et al.</i> , 1973 Fisk <i>et al.</i> , 1984b (85-300 K) Misunik <i>et al.</i> , 1973

TABLE III. (Continued).

System	μ_{eff}	$\chi(T=0)$ (10^{-3} emu/mol G)	Curie-Weiss Θ (K)	Reference
CeAl ₃		26 ($T=4.2$ K)		Edelstein <i>et al.</i> , 1974b
	2.63	30 ($T=0.5$ K)	-46	Buschow and Fast, 1966
	2.60	36 ($T=4.2$ K)	-39	Mader and Swift, 1968
	2.63	26 ($T=4.2$ K)		Andres <i>et al.</i> , 1975
CeCu ₆	2.69	36 ($T<0.1$ K)	-45	Stewart, Fisk, and Wire, 1984
		27 (1.4 K)		(40-110 K)
YbCuAl	2.67		-130	Pop <i>et al.</i> , 1977, data above 77 K
	4.35	25.5	-34	Mattens <i>et al.</i> , 1977
Pu _{0.7} U _{0.3} Al ₂		4.7		Arko <i>et al.</i> , 1973

^aThis difference is due to an error, with 3.08 being correct.

^bPer mole U atoms.

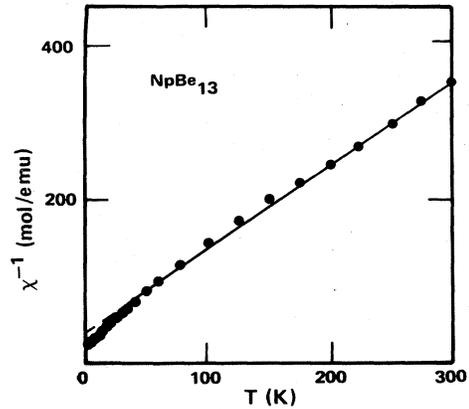


FIG. 28. Inverse susceptibility for NpBe₁₃ by Brodsky and Friddle (1974). The effective moment for the Curie-Weiss line shown is $2.76\mu_B$.

2. Discussion of properties

a. $\chi(T=0)$

The values for the low-temperature magnetic susceptibility shown in Table III are truly enormous. To put them in perspective, the *d*-electron element with the closest approach to being magnetic is Pd (Tsiang *et al.*, 1981), with $\chi(T=0) \approx 0.75 \times 10^{-3}$ emu/mol G, while the *f*-electron atom closest to being magnetic is Pu (Smith and Fisk, 1982), with $\chi(T=0) \approx 0.5 \times 10^{-3}$ emu/mol G. Thus the value of $\chi(T=0)$ in Table III for CeCu₂Si₂, 8×10^{-3} emu/mol G, although smaller than all the other heavy-fermion systems except UPt₃, is a factor of 10 larger than that for the nearly magnetic elements Pd and Pu, while $\chi(T=0)$ for CeAl₃ or NpBe₁₃ is almost a factor of 100 larger.

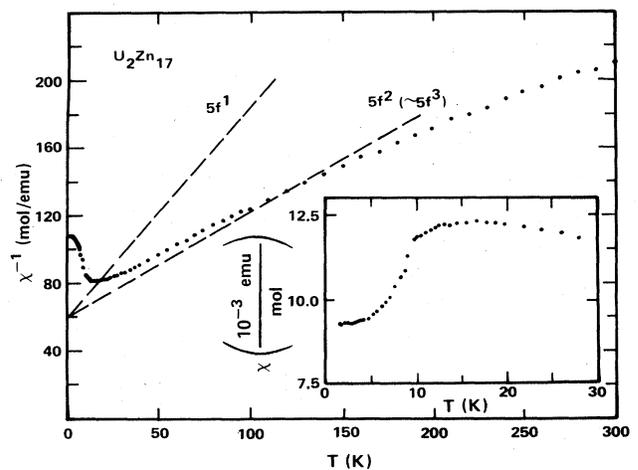


FIG. 29. Inverse susceptibility of U₂Zn₁₇ by Ott *et al.* (1984a), where a mole as defined here has only 1 U atom and 8.5 Zn atoms times Avogadro's number. The lines drawn show the slope corresponding to $5f^1$ and $5f^2$ ($\sim 5f^3$) states, i.e., the measured μ_{eff} does not correspond to any of these.

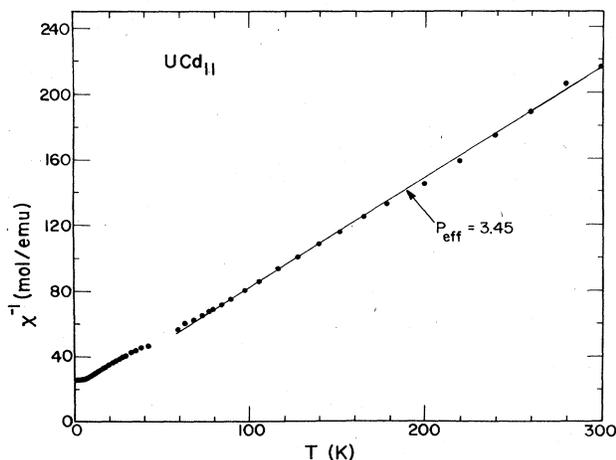


FIG. 30. Inverse susceptibility of UCd_{11} by Fisk *et al.* (1984b).

The enormous values of $\chi(T=0)$ for these heavy-fermion systems are equivalent to their huge γ as the feature which sets them apart from normal systems. As discussed previously, this large $\chi(T=0)$ has led to the description of these systems as a Fermi liquid of strongly renormalized quasiparticles. The source of the huge χ values has been postulated as a mechanism that, instead of allowing long-range ordering of the local magnetic moments inferred from the high-temperature Curie-Weiss law, destroys the local moment.

This mechanism has been identified either as the Kondo effect (see, for example, Bredl *et al.*, 1983), in which the local moments are quenched ("compensated") by the conduction electrons, or as valence fluctuations (Sales and Viswanathan, 1976) between two equivalent-energy f -electron ground states, one magnetic and the other not.

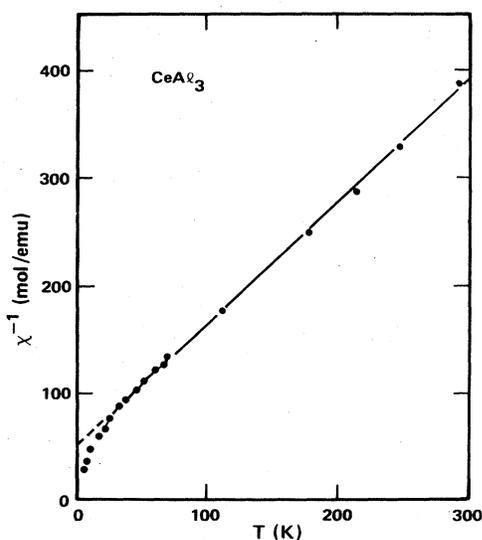


FIG. 31. Inverse susceptibility of CeAl_3 by Buschow and Fast (1966).

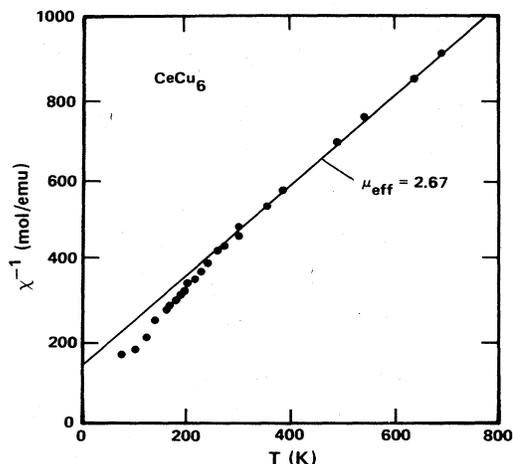


FIG. 32. Higher-temperature inverse susceptibility data for CeCu_6 by Pop *et al.* (1974).

The "nonmagnetic" state (excluding NpBe_{13} , U_2Zn_{17} , and UCd_{11}) then starts to form where the χ^{-1} vs T data start to deviate from the Curie-Weiss law at lower temperatures (~ 100 K for UBe_{13} ; Fig. 25.) It is interesting to note that the behavior of χ^{-1} after this deviation from linear behavior begins at lower temperatures is such that χ^{-1} increases above the extrapolation of the higher-temperature data for UBe_{13} (see Fig. 25) and UCd_{11} (Fig. 30), but decreases below this extrapolation for the other six heavy-fermion systems, Figs. 23, 26–29, and 31–33. What this implies for the moment compensation mechanism remains to be explained.

b. μ_{eff}

The chief use to which the values for μ_{eff} have been put is to determine what valence state (e.g., f^1 , f^2 , or f^3) is present in a given system, with the uniform result that the observed μ_{eff} is typically not assignable to any one of

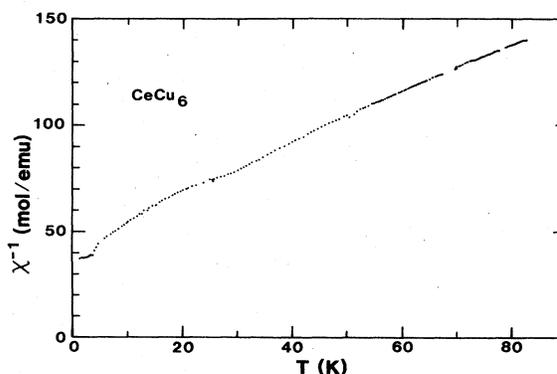


FIG. 33. Low-temperature inverse susceptibility data for CeCu_6 by Stewart, Fisk, and Wire (1984). The feature at 30 K corresponds to a slight anomaly in the specific heat.

them (see caption to Table III). This is taken to mean that the behavior is more complicated than that of simple local, independent f spins, *not* that the compounds are strongly mixed valent.

3. Magnetism

As discussed in the specific-heat section above, three of the eight heavy-fermion systems appear to undergo magnetic order at low temperatures: NpBe₁₃ ($T_{\text{mag}}=3.4$ K), U₂Zn₁₇ ($T_{\text{mag}}=9.7$ K), and UCd₁₁ ($T_{\text{mag}}=5.0$ K), Figs. 28–30, respectively. As seen in the χ^{-1} plots for these systems, neither NpBe₁₃ nor UCd₁₁ has a large anomaly in its susceptibilities at T_{mag} , while there is a peak in χ at about 15 K in U₂Zn₁₇, with $[\chi_{\text{max}}-\chi(T=0)]/\chi(T=0)=0.4$. Coupled with the specific-heat data, these anomalies imply that some type of weak, itinerant magnetic order is present. At higher temperatures, the susceptibilities of these magnetic systems are indistinguishable from those of the other systems (see Table III and the figures).

4. Field dependence of magnetization

As one way to look for magnetic behavior, the magnetization (χ/H) versus field of several of the heavy-fermion systems has been measured. There is a low-temperature contribution to χ in CeCu₂Si₂ that is field dependent below 15 T. Lieke *et al.* (1983) have taken this as a sign of impurities. Ott *et al.* (1984c) have shown that the magnetization at 4.2 K for UBe₁₃ is constant, to 2%, with field up to 10 T. The field dependence of the magnetization of UPt₃ has been measured on a single crystal by Frings *et al.* up to 35 T. In the basal plane, where there is a peak in χ at 19 K, M is linear in H until 15–16 T, where M begins to rise faster than linearly. At 30 T, M is enhanced over the linear extrapolation by 30%, reminiscent of the “metamagnetic” transition in the spin fluctuator TiBe₂ (Acker *et al.*, 1981; Monod *et al.*, 1981). Frings *et al.* see no field dependence of the magnetization up to 35 T perpendicular to the basal plane in UPt₃, where χ is monotonic with temperature. Magnetization measurements on CeAl₃ up to 11 T (Berton *et al.*, 1977) show signs of an impurity contribution (like that in CeCu₂Si₂) which saturates around 7 T. This is not surprising, considering the discussion of magnetic second phases in CeAl₃ in Sec. II.G.

5. Anisotropy

Anisotropy in the susceptibility has been measured for CeCu₂Si₂ (Assmus *et al.*, 1984; Batlogg *et al.*, 1984a) and UPt₃ (see Fig. 27, data of Frings *et al.*, 1983). In both cases, the observed anisotropy is quite large, with $\chi(T=0)$ varying by a factor of 2 in the different crystal directions. Although such anisotropy will not exist in the cubic systems UBe₁₃, NpBe₁₃, and UCd₁₁, except for pos-

sible effects below T_{mag} in the latter two, single crystals of U₂Zn₁₇ and CeCu₆ do exist and should be studied for possible anisotropy in χ .

It should be noted (see Table III) that the susceptibility of YbCuAl seems quite like that of the heavy-fermion systems, especially when one considers that μ_{eff} for an f^{13} Yb state is $4.54\mu_B$. Thus these susceptibility properties, which are common to heavy-fermion ($\gamma > 400$) systems, are not unique to them.

6. Correlations

One possible relation *between* C and χ that may be useful in correlating properties of heavy-fermion systems is the so-called “Wilson ratio” R , where

$$R = \frac{\pi^2 k^2 \chi(T=0)}{g^2 \mu_B^2 \gamma J(J+1)} \quad (3)$$

Wilson has shown that $R=2$ in the spin- $\frac{1}{2}$ Kondo problem (Wilson, 1975). Other discussions of R can be found in Krishna-murthy *et al.* (1980), Lustfeld and Bringer (1978), and Newns and Hewson (1980). Recently, this “Wilson ratio” has become a popular parameter for quantifying the heavy-fermion systems. One of the problems with this is determining what shall be used in Eq. (3) for $g^2 \mu_B^2 J(J+1)$ ($=\mu_{\text{eff}}^2$). As we have seen, the high-temperature susceptibility does not follow the behavior expected from simple f^1 , f^2 , or f^3 (in the case of U) states; therefore calculating $g^2 J(J+1)$ is not necessarily the correct procedure. Moreover, at lower temperatures it is obvious that some mechanism (not just crystal-field splitting) alters the Curie-Weiss behavior of χ . The crystal-field splitting of the ground-state f -electron state, and the corresponding μ_{eff} , have been approximated for CeCu₂Si₂ (Lieke *et al.*, 1982— $\mu_{\text{eff}}=1.65\mu_B$ as $T \rightarrow 0$). Even with neutron determination of the crystal-field split levels, done for CeCu₂Si₂ by Horn *et al.*, 1981, such a determination is rather uncertain and does not determine all the effects on μ_{eff} anyway. (For a discussion of this question for CeAl₃, see Edelstein *et al.*, 1974b.) Table IV presents a variety of ways of calculating R via Eq. (3), i.e., using assumed f^1 , f^2 , and f^3 (the latter only for U systems) states to calculate $g^2 J(J+1)$ (2.54^2 , 3.58^2 , and $3.62^2 \mu_B^2$, respectively), as well as the measured Curie-Weiss μ_{eff} from the higher-temperature χ data. The question of crystal-field splittings as a way to determine μ_{eff} is ignored here (except for CeCu₂Si₂), since in most cases the uncertainty introduced in the Wilson ratio would be larger than the change in μ_{eff} from the values used. Table IV is meant only for comparison between the systems in a consistent fashion, not as a source of definitive Wilson ratios. Fortunately, Lieke *et al.* (1982) and Assmus *et al.* (1984) each report both χ and γ on the same CeCu₂Si₂ sample, allowing a more accurate R value. In all cases, χ and γ are taken at $T=0$. For the interested reader, Eq. (3) reduces to $R=218.7\chi/(\gamma\mu_{\text{eff}}^2)$ when the units for χ are 10^{-3} emu/mol G, those for γ are mJ/mol K², and μ_{eff} is some dimensionless number of Bohr magnetons.

TABLE IV. Wilson ratio for the heavy-fermion systems. High- T experimental μ_{eff} is from Table III.

System	μ_{eff}			High T Curie-Weiss
	2.54(f^1)	3.58(f^2)	3.62(f^3)	
CeCu ₂ Si ₂	0.17	0.087		0.16
Sample 7, χ corrected for impurities, Lieke <i>et al.</i> , 1982				
sample 7, χ unadjusted	0.34	0.17		0.30
superconducting crystal, Assmus <i>et al.</i> , 1984,	0.22	0.11		0.52 ^a
uncorrected χ				
nonsuperconducting crystal grown in Cu, ^b using $\chi = (2\chi_{11} + \chi_1)/3$	0.44	0.22		
UBe ₁₃	0.46	0.23	0.23	0.31
UPt ₃	0.52–0.66	0.26–0.33	0.25–0.32	0.37–0.47
NpBe ₁₃	2.1	1.06	1.04	1.79
γ above T_{mag} unknown, use $\gamma(T=0)=900$ and χ from Stewart <i>et al.</i> , 1984b				
U ₂ Zn ₁₇	0.79	0.40	0.39	0.25
use $\gamma=535$				
UCd ₁₁	1.55–1.82	0.78–0.91	0.76–0.89	0.84–0.98
use $\gamma=840$				
CeAl ₃	0.75	0.38		0.70
use data of Andres <i>et al.</i> , 1975				
CeCu ₆	0.56	0.28		0.59
assume γ is the same as for CeAl ₃ , use $\mu_{\text{eff}}=2.48$ for Curie-Weiss value				
YbCuAl	1.04 ^c			
Pu _{0.7} U _{0.3} Al ₂				2.02 ^d

^aUsing $\mu_{\text{eff}}=1.65$, the low-temperature crystal-field-derived moment for Ce³⁺, see Lieke *et al.*, 1982.

^b χ data from Batlogg *et al.*, 1984a, C data from Willis *et al.*, 1984b. The sample shows inductive signs of superconductivity, but no bulk anomaly.

^cFor YbCuAl, use μ_{eff} for the f^{13} state of 4.54.

^dUsing $\chi(T=0)$ and μ_{eff} from the higher-temperature Curie-Weiss law from Arko *et al.*, 1973.

The values of γ used for the magnetically ordered systems are as stated. If $\gamma(T=0)$ were used for U₂Zn₁₇ and UCd₁₁, the values of R would be about three times larger than shown.

Although the values for R suffer from uncertainty in what to use for μ_{eff} , within a given column the numbers shown may be compared. For example, in a given column, a factor-of-2 increase in R is seen between the superconducting and nonsuperconducting single crystals of CeCu₂Si₂. Similarly, in a given column the R values for the first three systems listed, the superconductors, appear uniformly lower than the magnetic systems and the nonmagnetic, but nonsuperconducting, systems. The nonmagnetic CeAl₃ and CeCu₆ also have lower R values than the magnetic systems.

This observed correlation is an important one and may be fundamental. The mechanism (e.g., the Kondo effect) which prevents the high-temperature local moments (inferred from the Curie-Weiss behavior of χ) from undergo-

ing long-range order at low temperature, and which causes χ to remain finite (but large) and γ to be large, enhances both χ and γ . The trend suggested by the correlation in R values is that, as the magnetic (χ) enhancement varies in proportion to the specific-heat enhancement, the systems vary from magnetic (using column 1; Table IV) ($R \sim 0.8$ – 2.1) to nonmagnetic ($R \sim 0.56$ – 0.75) to superconducting ($R < 0.52$). It is interesting to note that CeCu₆ appears close in its Wilson ratio to UPt₃ and UBe₁₃, suggesting that CeCu₆ is almost a heavy-fermion superconductor.

B. Superconducting critical field

When bulk superconductivity in tetragonal CeCu₂Si₂ was first discovered, the slope of the critical field, dH_{c2}/dT ($T=T_c$), was reported (Steglich *et al.*, 1979) to be approximately -10 T/K, an anomalously large value. A later, more thorough study of H'_{c2} at T_c for CeCu₂Si₂

(Rauchschwalbe *et al.*, 1982) showed sample-dependent values up to -16.8 T/K. Recently, Assmus *et al.* (1984) found H'_{c2} at T_c to -23 T/K in both crystal directions for a single-crystal sample, with $H_{c2}(0)=2.4$ (2.0) T for field perpendicular (parallel) to the Ce planes (see Fig. 34). When cubic UBe_{13} was first found to be superconducting (mistakenly attributed to a second phase), Bucher *et al.* (1975) were able to suppress T_c by only 0.3 K using 6 T, i.e., $H'_{c2} \sim -20$ T/K. Ott *et al.* (1983) in their discovery of bulk superconductivity in UBe_{13} reported H'_{c2} at T_c to be -25.7 T/K. This value has increased with improving measurement technique, with the latest value (Maple *et al.* 1984a) up to -44 T/K, with $H_{c2}(0.37\text{ K})=6$ T. (An even larger value for H'_{c2} at T_c , -400 T/K, reported earlier for UBe_{13} by Maple *et al.*, 1984b, was in error.) When superconductivity was discovered in hexagonal UPt_3 (Stewart *et al.*, 1984c), the critical field was measured resistively with the field perpendicular to the current along the c axis, giving H'_{c2} at T_c of about -2 T/K, clearly the smallest value of the three heavy-fermion superconductors. Recently, two groups (Chen *et al.*, 1984; Willis *et al.*, 1984a) have measured H_{c2} vs T/T_c in single-crystal UPt_3 for H perpendicular and parallel to I , with the latter work having I in both crystal directions, i.e., four sets of data were taken. The data of Chen *et al.* are shown in Fig. 35. UPt_3 shows marked anisotropy in its H_{c2} vs T/T_c behavior, with H'_{c2} at T_c for H parallel to I equal to about -1 or 2 T/K, while for $H \perp I$, H'_{c2} at T_c is -6.3 T/K (Chen *et al.*, 1984). This anisotropy gets much smaller at high fields, and the two H_{c2} vs T curves almost cross at the lowest temperature of measurement, 0.12 K, with $H_{c2}(0) \simeq 2.3$ T (Chen *et al.*, 1984). This temperature dependence of the anisotropy in H_{c2} is the opposite of that found by Assmus *et al.* (1984) in $CeCu_2Si_2$. The temperature dependence of the anisotropy in H_{c2} in UPt_3 has been used by Varma (1984a) to argue that UPt_3 is a p -wave superconductor, in agreement with arguments for p -wave superconductivity in UPt_3 put forward by Stewart *et al.*, 1984c, when they discovered

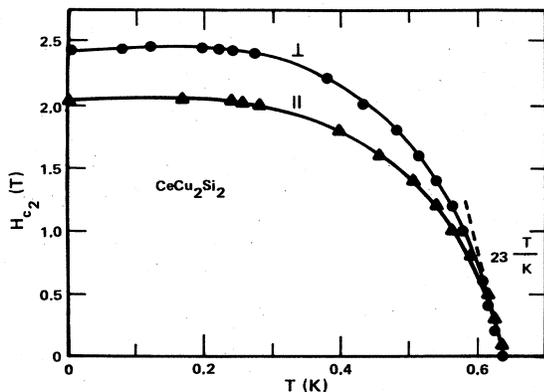


FIG. 34. Critical-field data for a single crystal of $CeCu_2Si_2$ by Assmus *et al.* (1984). Note that the results for the two crystal directions in tetragonal $CeCu_2Si_2$, parallel and perpendicular to the Ce planes, show no anisotropy at T_c , and large anisotropy at lower temperatures.

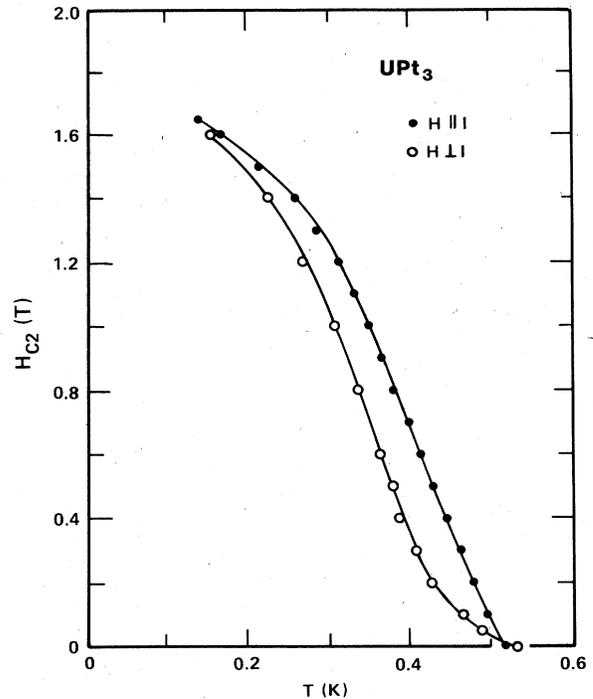


FIG. 35. Critical field for two field directions in UPt_3 , with the current I along the c axis (Chen *et al.*, 1984). The lines drawn are guides to the eye. Note the extreme anisotropy in the slope at T_c . (T_c onset is different because different crystals were used, with slightly differing T_c 's.)

superconductivity in a material where there was strong evidence (the $T^3 \ln T$ term in C) for spin fluctuations.

Whether the large H'_{c2} values at T_c for UBe_{13} and $CeCu_2Si_2$ [which both have no anisotropy in $H_{c2}(T_c)$] may be used as arguments for non-BCS superconductivity is still under discussion (Machida, 1983; Varma, 1984a; Assmus *et al.*, 1984.) Beyond any doubt, however, these very high H'_{c2} values are higher than any observed in other superconductors and signify something unusual in the superconducting state. The comparison of H'_{c2} values at T_c for the three heavy-fermion superconductors makes it clear that $CeCu_2Si_2$ and UBe_{13} are similar, while UPt_3 is different from the other two. The comparison between $CeCu_2Si_2$ and UBe_{13} is made even better when the sample quality issue is considered for $CeCu_2Si_2$, i.e., H'_{c2} may increase even more than the 50% seen by Assmus for the first single-crystal measurement (versus polycrystalline samples) when even better material is prepared.

IV. RESISTIVITY

A. Zero field

The resistivity R between 1 and 300 K for $CeCu_2Si_2$, UBe_{13} , and UPt_3 is shown in Fig. 36; for $NpBe_{13}$, U_2Zn_{17} , and UCd_{11} it is shown in Fig. 37; and for $CeAl_3$ and

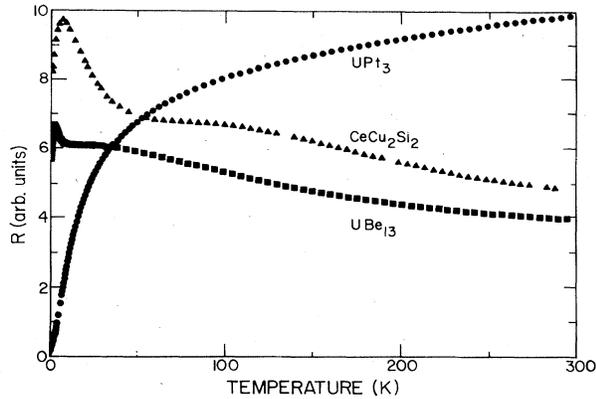


FIG. 36. Resistivity vs temperature: \blacktriangle , CeCu_2Si_2 (Stewart, Fisk, and Willis, 1983); \blacksquare , UBe_{13} (Ott *et al.*, 1983); \bullet , UPt_3 (Stewart *et al.*, 1984c). The relative magnitudes of the three sets of data are arbitrary.

CeCu_6 it is shown in Fig. 38.

Although there is some sample dependence of R in CeCu_2Si_2 , the broad comparison made in Fig. 36 holds true— CeCu_2Si_2 and UBe_{13} are similar, with peaks in R at low T [at 7 K (sample dependent) and 2 K, respectively] and shoulders in R vs T at higher T , while R for UPt_3 looks more like that of a normal metal. (For a review of resistivity in metals, see Fisk and Webb, 1981.) Similarly, Fig. 37 shows that the three magnetic systems (NpBe_{13} , U_2Zn_{17} , and UCd_{11}) have in common with CeCu_2Si_2 and UBe_{13} a rather flat temperature dependence in resistivity above 100 K. Below a certain temperature, the resistivities of UCd_{11} and U_2Zn_{17} fall precipitously (factors of 2 and 20, respectively, by 1.4 K), consistent with a decrease in spin-disorder scattering below T_{mag} . Figure 38 shows that the resistivities of CeAl_3 and CeCu_6 both have maxima at low temperatures with quite similar shapes, as well as rather little temperature dependence above 100 K.

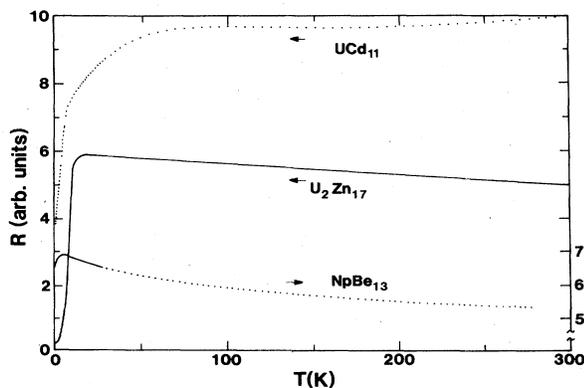


FIG. 37. Resistivity vs temperature for NpBe_{13} (Stewart *et al.*, 1984b), U_2Zn_{17} (Ott *et al.*, 1984a), and UCd_{11} (Fisk *et al.*, 1984b). The relative magnitudes (see Table V) of the three sets of data are arbitrary.

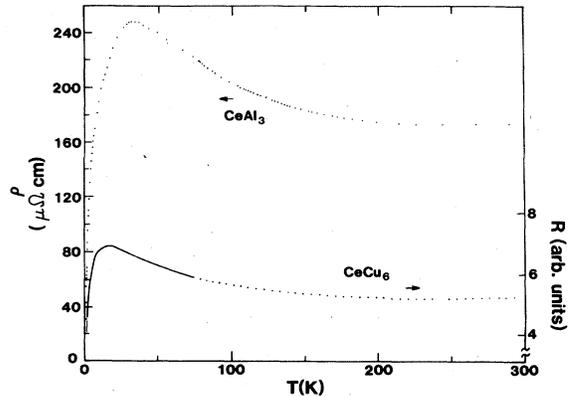


FIG. 38. Resistivity vs temperature of CeAl_3 (Ott *et al.*, 1984b) and CeCu_6 (Wire, Fisk, and Smith, 1984). The relative magnitudes of the two sets of data are arbitrary.

Thus, in terms of temperature dependence of resistivity, UPt_3 is anomalous compared to the other seven heavy-fermion systems, with no region of negative dR/dT at low temperatures. [A slight peak in the resistivity of UPt_3 at 800 K is thought to be due to a transformation (Wire, Thompson, and Fisk, 1984).]

The appropriate parameters [ρ_{max} , at T_{max} , $\rho(T=0)$] are gathered for all the heavy-fermion systems in Table V. Absolute numbers are not available for NpBe_{13} or UCd_{11} . The value of ρ_{max} is quite large in all of the systems (CeCu_2Si_2 , UBe_{13} , U_2Zn_{17} , CeAl_3 , and CeCu_6) where values are known, and presumably would be similar in NpBe_{13} and UCd_{11} . The larger values of ρ_{max} and the limited decreases observed for $T < T_{\text{max}}$ do not appear to be sufficiently dependent on sample quality ever to approach the behavior of UPt_3 . Therefore, the difference in the resistivity ratio [$R(300\text{ K})/R(4\text{ K})$] in UPt_3 and the other heavy-fermion systems is likely intrinsic.

1. Sample variation

The variation in ρ_{max} in CeCu_2Si_2 is a function of temperature cycling and has been attributed to microcracks in the samples (Franz *et al.*, 1978). No explanation for the disagreement for ρ_{max} in CeAl_3 has been proposed. For UPt_3 , Stewart *et al.* (1984c) found large sample variation in the resistivity ratio of their samples that correlated strongly with T_c . When $R(300)/R(T_c^+)$ decreased from 145 to 43 from one sample to another, T_c decreased from 0.54 to 0.27 K. This was taken as another indication of p -wave superconductivity—strong dependence of T_c on defects. (See Balian and Werthamer, 1963, for a discussion of the role of defects in a p -wave superconductor.) A similar correlation between resistivity ratio and T_c was not found by Assmus *et al.* (1984) for CeCu_2Si_2 . Recently, a sample of UPt_3 with a resistivity ratio of 180 and a T_c of 0.486 K has been reported (Willis *et al.*, 1984a), contradicting the earlier reported correlation.

TABLE V. Resistivity parameters for heavy-fermion systems.

System	ρ_{\max} ($\mu\Omega$ cm)	T of the max. (K)	$\rho(T=0)$ ($\mu\Omega$ cm)	Reference and comments	
CeCu ₂ Si ₂	180	6.5		Stewart, Fisk, and Willis, 1983, nonsuperconducting single crystal	
	220	~24	3.5 (extrap)	Lieke <i>et al.</i> , 1983, sample 7	
	220–330	21	5	41 ($T=0.3$ K, $H=3$ T)	Franz <i>et al.</i> , 1978
					130
	155 ($\parallel a$ axis)	20	20	50	Assmus <i>et al.</i> , 1984, superconducting single crystals
140 (\parallel)	20	20	4.5	Schneider <i>et al.</i> , 1983, nonsuperconducting crystal	
					120 (\perp)
162 (\parallel)					
45 (\perp) ^a					
UBe ₁₃	234	2.35	42 (extrap from H'_{c2})	Ott <i>et al.</i> , 1983	
UPt ₃			1 (at T_c^+) 0.5 (extrap)		
U ₂ Zn ₁₇	~130	18	1.6 (at 1.2 K)	Ott <i>et al.</i> , 1984a	
UCd ₁₁		~100		Fisk <i>et al.</i> , 1984b	
CeAl ₃	120	35	32 (at 1.3 K)	van Daal <i>et al.</i> , 1971	
			0.77 (at 0.015 K)	Andres <i>et al.</i> , 1975	
			5 (at 0.050 K)	Remenji <i>et al.</i> , 1983	
250	33			Ott <i>et al.</i> , 1984b	
CeCu ₆	140	14	60 (at 1.4 K)	Stewart, Fisk, and Wire, 1984; Wire, Fisk, and Smith, 1984	

^aNo peak in (001) direction, only a shoulder.

2. T_{\max}

For CeCu₂Si₂, the superconducting transition temperature has been found to scale monotonically with T_{\max} (see column 2 of Table V; Aliev *et al.*, 1982; Bredl *et al.*, 1983; Stewart, Fisk, and Willis, 1983). This low-temperature maximum has been identified as due to "Kondo-lattice" scattering (Aliev *et al.*, 1982), while the shoulder at 100 K (see Fig. 36) is thought to be due to crystal-field effects (Horn *et al.*, 1981). Aliev *et al.* showed that pressure increased both T_{\max} and T_c together in CeCu₂Si₂, with T_{\max} increasing at ~0.4 K/kbar. Percheron *et al.* (1973) found that T_{\max} in CeAl₃ increased monotonically from 34 to 70 K as pressure increased from 0 to 16.8 kbars. The peak in the resistivity of CeAl₃ has been interpreted in various ways, e.g., as Kondo sidebands (van Daal *et al.*, 1971) or virtual bound-state scattering (Andres *et al.*, 1975).

3. T^2 dependence

In Fermi-liquid theory, $\rho \propto AT^2$ at low temperatures. Unfortunately, not only can this temperature dependence come from other model approaches (e.g., paramagnon

theory; Doniach, 1968), but experimentally the temperature range where ρ behaves as T^2 is quite limited. In CeAl₃, ρ ($\mu\Omega$ cm) ~ 31 T (K) between 0.5 and 1.2 K (Remenji *et al.*, 1983). Andres *et al.* (1975) show that ρ ($\mu\Omega$ cm) ~ 35 T² (K²) between 0.015 and 0.1 K, and state that the same behavior holds up to 0.3 K. Lieke *et al.* (1982) claim ρ ($\mu\Omega$ cm) ~ 10 T² (K²) in applied field below T_c in CeCu₂Si₂. Whether such data strengthen a particular theoretical model is a matter of judgment.

4. Effective mass

Since a large effective mass m^* of the conduction electrons is one of the parameters commonly referred to when discussing heavy-fermion systems, a short discussion is given here. Three differing approaches have been taken to calculating m^* for these systems. Rauchschalbe *et al.* (1982) utilize an equation for $H'_{c2}(T_c)$ that involves the Fermi wave number k_F , γ , and $\rho(T=0)$ to calculate $m^* = 220m_e$ for CeCu₂Si₂. Ott *et al.* (1983) use a relation between ρ_{\max} , γ , and k_F to deduce $m^* = 192m_e$ for UBe₁₃. Chen *et al.* (1984) use yet a third equation to calculate $m^* = 187m_e$ for UPt₃. All three approaches assume a spherical Fermi surface, which makes the results of dubi-

ous quantitative accuracy in systems with proven strong f -electron character.

B. Magnetoresistance

Just as specific-heat measurements in a field proved a useful probe of the electronic properties, so do magnetoresistance measurements. Resistivity in 11 T for CeCu_2Si_2 was measured by Stewart, Fisk, and Willis (1983). The resultant negative magnetoresistance was 4.5% at 2 K and decreasing in size with increasing temperature. Batlogg *et al.* (1984b) found a large negative magnetoresistance (-40% in 8 T) (sample-dependent in size) for H parallel to the c axis in CeCu_2Si_2 and a smaller change with field for $H \perp c$ that changed sign (+ to -) when heating through 4 K.

Maple *et al.* (1984a) have reported resistance data for UBe_{13} in fields up to 6 T and below 1 K. Stewart *et al.* (1984a) have measured the resistance of UBe_{13} as a function of field up to 11 T. These data are shown in Fig. 39 and show a large negative magnetoresistance that increases in size with decreasing temperature and increasing field (-42% at 1.2 K and 11 T). A correlation found in that work was that, if $\Delta R/R$ was plotted versus H , the resultant family of curves, one for each temperature (see Fig. 40), could be mapped onto one another by plotting instead $\Delta R/R$ vs $H/(T+T^*)$, with $T^*=1$ K. Another interesting correlation of the magnetoresistance data of Stewart *et al.* (1984a), overlooked at the time, is presented in Fig. 41. If $\Delta R/R$ (in percent) for a given field is normalized at one temperature to the specific heat C/T (in mJ/mol K^2) measured by Stewart *et al.* (1984a), then the $-\Delta R/R$ vs T^2 data at a given field coincide with the C/T vs T^2 curve at zero field, for each given field. This realization that the temperature dependence of the magnetoresistance $-\Delta R/R$ is the same as the temperature dependence of C/T suggests that the mechanism which causes the increase in γ as a function of decreasing temperature causes the increase in $-\Delta R/R$ as the same func-

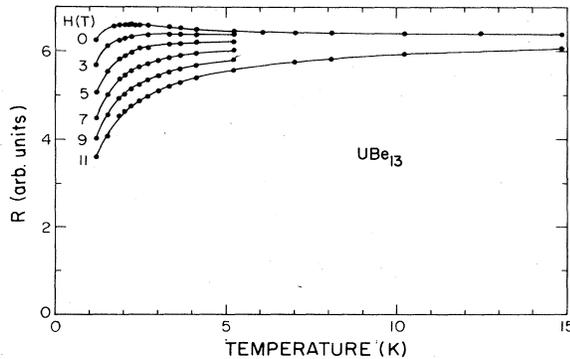


FIG. 39. Resistivity of UBe_{13} as a function of applied field (0, 3, 5, 7, 9, and 11 T). Above 5 K, only the 0- and 11-T data are shown; at each temperature, the higher the field, the lower the resistivity.

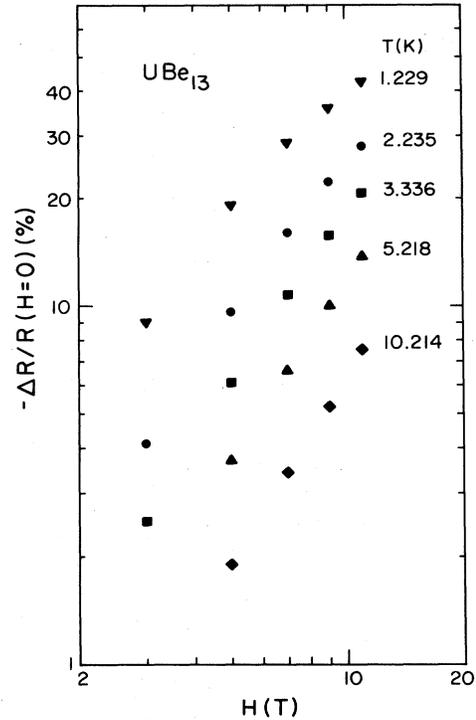


FIG. 40. Magnetoresistance vs field at various temperatures for UBe_{13} (Stewart *et al.*, 1984a).

tion of decreasing temperature.

The positive magnetoresistance of UPt_3 in 11 T has been reported by Stewart *et al.* (1984d). At 1.2 K, the increase in R is 40%. The power law followed at 1.2 K is $\Delta R/R \propto H^{1.25}$.

The magnetoresistance of CeAl_3 has been reported by several workers. For $T > 1$ K, the magnetoresistance is negative, with $-\Delta R/R = 13\%$ at 6 T and 4.2 K (Remenji *et al.*, 1983). Below 1 K, $\Delta R/R$ is initially positive with field, followed by negative $\Delta R/R$ above ~ 2 T. At fixed

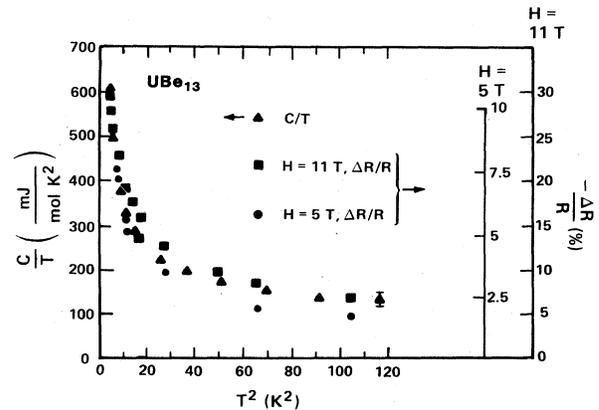


FIG. 41. Here the zero-field C/T data of UBe_{13} (\blacktriangle) are shown plotted vs T^2 along with the magnetoresistance data in fields of 11 T (\blacksquare) and 5 T (\bullet) from data taken by Stewart *et al.* (1984a), also shown vs T^2 . Although the vertical scales for the three sets of points are different, the temperature variation of the C/T and $-\Delta R/R$ data is seen to be quite similar.

(4.05 T) field as a function of temperature, $\Delta R/R$ for CeAl₃ changes from negative below 0.5 K to positive above. Zlatić (1981) has advanced a model to explain the magnetoresistance data for CeAl₃.

Recently, Onuki *et al.* (1984) reported an anisotropic negative magnetoresistance in fields up to 7.5 T for CeCu₆.

V. OTHER MEASUREMENTS

A. Neutron scattering

Horn *et al.* (1981) have performed inelastic neutron scattering on CeCu₂Si₂. They assign a crystal-field splitting scheme (three split doublets) for CeCu₂Si₂ based on two inelastic lines observed at 135 and 360 K in energy. In addition they measure the halfwidth $\Gamma/2$ of the quasi-elastic line to be temperature dependent, with $\Gamma/2(T \rightarrow 0) \simeq 10$ K. This number may be identified as a characteristic spin-fluctuation ("Kondo") temperature (Horn *et al.*, 1981) by analogy with dilute Kondo systems; thus phenomenologically $\chi(T=0) \propto [\Gamma(T \rightarrow 0)]^{-1}$.

Ott *et al.* (1984c) report preliminary neutron scattering results of Kjems *et al.* (1984) for UBe₁₃ which are interpreted as ruling out a crystal-field splitting explanation of the low-temperature peak in the resistivity, as used for the resistivity data in CeAl₃. Other neutron scattering work, both elastic single-crystal work and inelastic work, is in progress.

Buyers (1984) has reported neutron scattering evidence for local spin fluctuations in UPt₃ at room temperature. In the spin-fluctuation compound UAl₂, Loewenhaupt *et al.* (1979) found $\Gamma/2(T \rightarrow 0) \sim 300$ K in energy, with a slight increase of this quasielastic linewidth with temperature. There is an important difference between local spin fluctuations, as seen by neutron scattering, and long-range spin fluctuations ("paramagnons"), as seen in the $T^3 \ln T$ term in C in UPt₃ and UAl₂ only at low temperatures.

Neutron scattering determination of the type of magnetic ordering in NpBe₁₃, U₂Zn₁₇, and UCd₁₁ would be of interest.

Several neutron scattering experiments have been performed on CeAl₃ (Edelstein *et al.*, 1974; Murani *et al.*, 1977, 1980). The latter work reports a temperature-dependent $\Gamma/2$ which is about 6 K at $T=0$. The $T^{1/2}$ temperature dependence observed for the quasielastic linewidth above a constant behavior below 1 K is used by Murani *et al.* (1980) to argue that CeAl₃ is a Fermi liquid below 1 K, with some inconsistencies. Murani *et al.* (1977) found two inelastic lines in their neutron scattering spectrum at 60 and 90 K, which were interpreted to imply a crystal-field splitting scheme of the Ce³⁺ $J = \frac{5}{2}$ state.

For intermediate valent compounds, $\Gamma/2$ is found to be mostly temperature independent, with $\Gamma/2(T \rightarrow 0)$ equal to some large value, for example, 120 K in YbCuAl (Mattens *et al.*, 1980). This different behavior in $\Gamma/2$ is one of the main experimental differences used at present in distinguishing "Kondo lattice" systems from intermediate

valent systems, which have similar physical properties. For a discussion, see Lawrence *et al.* (1981).

B. Photoemission

All the heavy-fermion systems, with their huge γ 's and $\chi(T=0)$'s, have quite small characteristic energies, of the order of tens of K, as seen by, for example, field measurements of C and the values of T_{\max} in the resistivity measurements. A useful discussion of the strengths and weaknesses of photoemission as a probe of such sharp electronic structure is given by Parks *et al.* (1983), who measured resonant photoemission in CeCu₂Si₂ and related systems. Problems of resolution (0.15 eV = 180 K in energy), initial versus final state in the photoemission process, and measurement at room temperature prohibit the measurement of sharp structure near the Fermi energy implied from the low-temperature measurements. Parks *et al.* (1983) do find significant $4f$ character near (0.25 eV) to the Fermi energy E_F , similar to that seen in Ce.

Landgren *et al.* (1984) have studied UBe₁₃ using resonant photoemission and find $5f$ character at E_F , similar to that found in pure U. Wuilloud *et al.* (1984) measured x-ray photoemission and bremsstrahlung isochromat spectroscopy (XPS and BIS) for UBe₁₃ and could find, using a resolution of ~ 0.5 eV, no sharp structure at the Fermi energy. The question of oxygen contamination of the surface in UBe₁₃ and CeCu₂Si₂ has been raised in a recent work by Parks *et al.* (1984), who find their resonant photoemission results and implied strong hybridization between the f electrons and neighbor atom orbitals in contradiction to earlier work.

Schneider and Laubschat (1981) have studied UPt₃ using x-ray photoemission and find significant $5f$ character at E_F .

Ott *et al.* (1981) looked for an excessively high density of states in CeAl₃, using room temperature XPS and BIS with resolutions of 0.25 and 0.45 eV, respectively. No structure was found possibly due both to resolution and the fact that evidence for a narrow band (large C/T) exists only below ~ 10 K. Baer *et al.* (1981) have also measured XPS and BIS in CeAl₃.

C. Nuclear magnetic resonance

Aarts *et al.* (1983) have measured the Si NMR in CeCu₂Si₂, and MacLaughlin *et al.* (1984a) have measured the nuclear quadrupole resonance of Cu in CeCu₂Si₂. It is interesting to compare the results of MacLaughlin *et al.* (1984a) with those of Clark *et al.* (1984) for UBe₁₃. MacLaughlin *et al.* find that the spin-lattice relaxation rate $1/T_1$ below T_c in CeCu₂Si₂ has the temperature dependence found in ordinary superconductors, with some quantitative deviation from theory. Clark *et al.* find that the expected peak in $1/T_1$, observed in CeCu₂Si₂ near T_c , is anomalously below T_c in UBe₁₃, below $T/T_c = 0.65$. The data of MacLaughlin show an anomaly in $1/T_1$ at

1.2 K not seen in UBe_{13} by Clark *et al.* Moreover, $1/T_1$ does not vary as T between T_c and 1.3 K in CeCu_2Si_2 , contrary to expectation from a Fermi-liquid description. Clark *et al.* observe an anomalous increase in $1/T_2$, the transverse relaxation rate, below T_c not seen in CeCu_2Si_2 and call this increase evidence for very slow magnetic fluctuations in the superconducting state. Further investigation of this effect is of interest as regards p -wave superconductivity. Recently, MacLaughlin *et al.* (1984b) have found that $1/T_1$ in UBe_{13} varies as T^3 below T_c , consistent with p -wave superconductivity.

Pop *et al.* (1974) reported NMR on CeCu_6 above 77 K.

D. Thermal expansion

The thermal expansion of CeAl_3 has attracted considerable interest, both experimentally (Andres *et al.*, 1975; Ribault *et al.*, 1979) and theoretically (Edelstein and Koon, 1983). Starting at $T=0$, the thermal expansion coefficient decreases linearly with increasing temperature to about $-1.2 \times 10^{-5} \text{ K}^{-1}$ (Ribault *et al.*, 1979), then increases to a positive maximum of about $1.3 \times 10^{-5} \text{ K}^{-1}$ at 2 K, and falls off at higher temperatures (Andres *et al.*, 1975). Between 4.2 and 300 K, Andres *et al.* found a volume change equivalent to a few percent of that which would come from a 3^+ to 4^+ valence change.

The compound YbCuAl has a negative peak in its thermal expansion coefficient at 24 K of about $-0.7 \times 10^{-6} \text{ K}^{-1}$. The behavior of the thermal expansion between 0 and 300 K was interpreted by Pott *et al.* (1981) as a change of valency of 3.5%.

Thermal expansion has also been measured on UCd_{11} (Fisk *et al.*, 1984b), with a peak observed at $T_m = 5 \text{ K}$ of $2 \times 10^{-6} \text{ K}^{-1}$, stated to be similar to results for U_2Zn_{17} . The broad falloff of the thermal expansion above the peak at 5 K in UCd_{11} is consistent with the broad tail to higher temperatures observed in the specific heat.

E. Chemical substitution

For the materials scientist, the substitution of one element for another in a compound, and the subsequent observation of the change in properties, is one of his most powerful tools. A number of such studies have been made and are mentioned briefly here.

In CeCu_2Si_2 , Ishikawa *et al.* (1983) found that decreasing the Cu content to $\text{CeCu}_{1.95}\text{Si}_2$ depresses superconductivity to below 0.070 K, while Steglich *et al.* (1983) found the disappearance of T_c below 0.040 K between $\text{CeCu}_{1.95}\text{Si}_2$ and $\text{CeCu}_{1.90}\text{Si}_2$. Both works also investigated the less rapid variation of T_c with Ce and Si stoichiometry. Ishikawa *et al.* found no superconducting or magnetic transitions between 0.07 and 20 K in CeT_2Si_2 ($T = \text{Co}, \text{Pd}, \text{Rh}, \text{or Ru}$) and CeT_2Ge_2 ($T = \text{Co or Ni}$). This result is contrary to work by Murgai *et al.* (1982) and Grier *et al.* (1984), who find that CePd_2Si_2 is antiferromagnetic at 10 K. A large number of materials form in

the CeCu_2Si_2 structure (see Rossi *et al.*, 1979, for examples), and many are currently under investigation to further understanding of heavy-fermion systems in general and CeCu_2Si_2 in particular.

Steglich *et al.* (1983) performed substitutions in CeCu_2Si_2 and found that Gd substituted for Ce depressed T_c twice as fast as nonmagnetic substitutions. This may be used as an argument against p -wave superconductivity in CeCu_2Si_2 , since nonmagnetic substitutions are supposed to be equally destructive to T_c in a p -wave superconductor.

Smith *et al.* (1984) studied the effect on T_c of substitutions for the U in UBe_{13} . There appears to be a non-monotonic decrease of T_c in $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$, with a T_c ($\sim 0.6 \text{ K}$) apparently independent of x between two and three percent. Ott *et al.* (1984b) found two sharp specific-heat transitions in $\text{U}_{0.967}\text{Th}_{0.033}\text{Be}_{13}$, with as yet no explanation. Giorgi *et al.* (1984) found that small substitutions of B or Cu for Be in UBe_{13} dramatically suppressed T_c ; e.g., $\text{UBe}_{12.94}\text{Cu}_{0.06}$ has $T_c < 0.015 \text{ K}$. Stewart and Giorgi (1984) have shown that no change in the specific heat above 1.5 K in UBe_{13} is caused by this minimal Cu substitution, indicating that the heavy-fermion specific heat is not a sufficient condition for the superconductivity in UBe_{13} .

A large number of substitutions for the Ce in CeAl_3 have been performed, including Th and La (Buschow and van Daal, 1970; van Maaren *et al.*, 1971; van Daal *et al.*, 1971), as well as Y (van Maaren *et al.*, 1971; van Daal *et al.*, 1971).

VI. THEORY

The discovery of heavy-fermion systems (HFS) and superconductivity therein has caused a great deal of interest not only in the experimental community, but also among the theorists. Some theoretical works are briefly mentioned here for completeness.

There is a rather thorough review by Leggett (1975) of the theory of the superfluid A and B phases of ^3He . As we shall see, some recent theories have focused on the similarities of the not-yet-understood properties of HFS to the already understood properties of ^3He . A more recent review on ^3He is that by Vollhardt. Nozières (1974) gives a Fermi-liquid description of Kondo systems. Newns and Hewson (1980,1981) have two discussions of local Fermi-liquid theory applied to intermediate valent systems. Varma (1976) and Lawrence *et al.* (1981) have written reviews of intermediate valent systems.

Razafimandimby *et al.* (1984) have investigated electron-phonon mechanisms in Kondo lattices, and conclude that they are strong enough to explain superconductivity in CeCu_2Si_2 . Lee and Bowen (1984) predict a new, nonphonon, attractive interaction between electrons in the Kondo regime to explain superconductivity in HFS. Tachiki and Maekawa (1984) predict superconductivity in the Kondo-lattice systems CeCu_2Si_2 and UBe_{13} , using a

phenomenological attractive interaction which might or might not be of phonon-mediated origin. The possibility of p -wave superconductivity in HFS was raised early by Varma (1984b,1984c). Varma (1984a) uses the critical-field anisotropy at T_c measured by Chen *et al.* (1984) and Willis *et al.* (1984a) in UPt_3 to argue for p -wave superconductivity in this material. Anderson (1984) concludes that electrons in HFS superconductors, in analogy to ^3He , have attractive parallel-spin (p -wave) interactions. Ueda and Rice (1984) and Ott *et al.* (1984e), in analogy with ^3He and using the Brinkman-Rice theory (1970) of a strongly correlated Fermi liquid, have argued that superconductivity in UBe_{13} is p wave. Bedell and Quader (1984) use a Fermi-liquid model to argue that p -wave pairing occurs in HFS.

VII. CONCLUSIONS

A. Correlations

The usual methodology in a new, as-yet-not-understood field of research is to search for correlations that provide a clue to the underlying mechanisms. In this paper we have seen that some correlations exist, and some that are appealingly simple do not work at all. In order to end on a positive note, let us first consider the correlations that do not appear to be valid.

(1) There is no good agreement between the characteristic temperatures or the entropies associated with the specific-heat anomalies observed for CeCu_2Si_2 (C_{max} at 2–3.5 K, $\Delta S = 4\text{--}5.8$ J), UBe_{13} (C_{max} at 2–3 K, $\Delta S = 1$ J), CeAl_3 [C_{max} at 2.5, 4, 6 K due to impurities (?), $\Delta S < 1$ J; C_{max} at 25 K, $\Delta S = 9.1$ J] and CeCu_6 (no maximum, shoulder between 2 and 7 K). Therefore, using a phenomenological scaling for γ or $\chi(T=0)$ based on a characteristic anomaly temperature [$\gamma, \chi(0) \propto T_{\text{anomaly}}^{-1}$] may appear to work on a crude scale, if one says that one of the low-temperature anomalies in CeAl_3 is intrinsic. However, since the entropies associated with these anomalies are so different, it is not clear that such a scaling law is not accidental. Moreover, within a given system (CeCu_2Si_2) T_{anomaly} varies by 75%, and γ is unchanged.

(2) The appealing scaling law (see Sec. II.A) of Bredl *et al.* (1983) for CeCu_2Si_2 , that γ varies inversely with T_c , fails if single-crystal results are included (Stewart, Fisk, and Willis, 1983; Assmus *et al.*, 1984).

(3) The inverse scaling of T_c with the temperature of ρ_{max} in CeCu_2Si_2 appears to work within a given lab, but not for comparing results between different groups, since Aliev *et al.* find $T_c = 0.5$ K for $T(\rho_{\text{max}}) = 7$ K, while Stewart, Fisk, and Willis (1983) find no T_c down to 0.050 K for $T(\rho_{\text{max}}) = 7$ K.

(4) Another appealing correlation (that fails) is between observed properties, e.g., γ or $\chi(0)$, and the f -atom separation in these compounds. Hill pointed out that f -atom systems may have magnetic order above a certain separation (~ 3.4 Å for U and Ce compounds). In the

heavy-fermions systems, where the f -atom separation is so much larger, surely the properties scale with the f -atom separation? Not so, as shown in Table VI. In fact, as shown by some nonheavy systems at the end of the table, a large f -atom separation, although a necessary condition, is not sufficient for a large γ or $\chi(0)$.

As for the more promising correlations, these include the following.

(1) One correlation that appears positive, but on limited data, is that magnetic field appears to affect in a similar way the low-temperature specific heat of the “temperature-dependent- γ ” heavy-fermion systems, i.e., CeCu_2Si_2 , UBe_{13} , CeCu_6 , with no data on CeAl_3 at high field. Although models have been proposed to explain these data, whether these results in magnetic field will elucidate the low-temperature electronic state of these systems depends on further experimental and theoretical work.

(2) Another correlation shown in this paper that may be fruitful for further study is the similarity in the temperature dependence of the magnetoresistance and zero-field γ in UBe_{13} .

(3) The rather constant percentage decrease ($\sim 66\%$) found in γ below T_{mag} in U_2Zn_{17} , UCd_{11} , and NpSn_3 is an intriguing correlation. If this correlation is not fortui-

TABLE VI. Noncorrelation of properties with f -atom separation. As in the text, a mol means mole f atoms. Values used are representative only. The parameter d is the shortest distance between f atoms. No other systems are presently known which have $\gamma > 300$ mJ/ f (or d) mol K². Systems with γ values above 200 include YbCuAl (260), $\text{Pu}_{0.7}\text{U}_{0.3}\text{Al}_2$ (260), and NpSn_3 (242) mentioned in the text, as well as UCu_5 (250; van Daal *et al.*, 1975), $\text{U}_{0.85}\text{La}_{0.15}\text{Al}_2$ (238; Wire *et al.*, 1983), UAu_3 (260, Fisk *et al.*, 1984a), and NpOs_2 (200; Brodsky, 1977). Probably over 20 compounds exist with γ between 100 and 200, including UAl_2 (142) mentioned in the text, USn_3 mentioned here, CeBe_{13} (115; Cooper *et al.*, 1971), YbCu_2Si_2 (135; Sales and Viswanathan, 1976), SmS under pressure (145; Bader *et al.*, 1973), and CeAl_2 (135; Bredl and Steglich, 1978). For d -electron compounds, $\text{V}_{1.97}\text{O}_3$ ($\gamma = 54$ mJ/mol V K²; McWhan *et al.*, 1973), TiBe_2 ($\gamma = 56$ mJ/mol Ti K²; Stewart *et al.*, 1982), and V_3Ga ($\gamma = 33$ mJ/mol V K²; Cort *et al.*, 1981) have among the highest γ values.

	d (Å)	$\chi(0)$ (10^{-3} emu/mol)	$\gamma(0)$ (mJ/mol K ²)
CeCu_2Si_2	4.1	7–?	1100
UBe_{13}	5.13	15	1100
UPt_3	4.1	7	450
NpBe_{13}	5.13	56	900–?
U_2Zn_{17}	4.39	12.5	535
UCd_{11}	6.56	38	840
CeAl_3	4.43	36	1600
CeCu_6	4.83	27	~ 1600
USn_3^a	4.63	10	169
UGe_3^a	4.21	1.3	20.4
UPt_3^b	5.25	5.7	85

^avan Maaren *et al.*, 1974.

^bFrings *et al.*, 1983.

tous, one could obtain $\gamma(T > T_{\text{mag}})$ by simply knowing $\gamma(T=0)$. This gives yet another estimate for γ above T_{mag} in NpBe_{13} , i.e., 2700.

(4) The comparison of the properties of the three heavy-fermion superconductors made it clear that there was no correlation between CeCu_2Si_2 and UBe_{13} on the one hand, and UPt_3 on the other, in temperature dependence of C , χ , or R , size or anisotropy of $H'_{c2}(T_c)$, or dependence of T_c on residual resistivity—where CeCu_2Si_2 does not have $T_c \propto \rho(T=0)^{-1}$ and UPt_3 has a very strong T_c dependence on $\rho(0)^{-1}$. The correlation that did show up was that all three heavy-fermion superconductors had relatively low Wilson ratios, $R [\propto \chi(0)/\gamma(0)]$, whereas the other heavy-fermion systems had larger R values, with the magnetic systems having the largest R 's. Although this correlation does not obviate the need to look for T_c down to dilution refrigerator temperatures (0.050 K) in new candidate systems, it does suggest that one should not be too hopeful in looking for T_c in systems with $R > 1$.

B. Unanswered questions; further work

Probably the most interesting question remains: do any (or all, or some) of the heavy-fermion superconductors have p -wave pairing? Evidence for anisotropy in the superconducting properties, such as the already observed $H'_{c2}(T_c)$ anisotropy in UPt_3 , needs to be looked for, along with other distinguishing properties (e.g., sensitivity of T_c to damage and magnetic impurities) of p -wave superconductors. Many such experiments are underway; many are quite difficult and are in the way of negative experiments—if they do not work, they prove nothing. An example of this is the attempt to tunnel from a BCS superconductor into one of the three heavy-fermion superconductors—no supercurrent can mean either p -wave pairing or poor junction contact.

The T^3 dependence of $1/T_1$ below T_c in the nuclear resonance measurements in UBe_{13} needs to be further investigated, as well as the differences in $1/T_1$ in CeCu_2Si_2 and UBe_{13} . The non-BCS-like behavior of the electronic superconducting specific heat C_{es} in UBe_{13} , used by Ott *et al.* (1984e) to argue for p -wave superconductivity, needs to be investigated and compared with CeCu_2Si_2 , where “gapless” superconductivity (i.e., extreme non-BCS-like C_{es} behavior) has been observed (Bredl *et al.*, 1983). Especially, the question of how a γ that is a function of T affects the expected shape of C_{es} should be considered in UBe_{13} , where the superconducting and normal-state entropies do not agree by 10% (Ott *et al.*, 1984e), a quite large discrepancy.

It would truly be surprising if CeCu_2Si_2 and UBe_{13} were not both the same type of superconductor, due to their many similarities, whereas UPt_3 seems clearly different.

In the nonsuperconducting heavy-fermion systems, neutron scattering and Mössbauer measurements in NpBe_{13} , the magnetic analog of superconducting UBe_{13} ,

would be useful to determine the type of magnetic order. Such measurements would also be useful in UCd_{11} and U_2Zn_{17} . The behavior of γ as a function of temperature to higher temperatures in UCd_{11} and U_2Zn_{17} needs to be investigated.

Finally, more large f -atom separation compounds need to be investigated (and not just Ce and U systems) to try to find further clues as to what creates a heavy-fermion system, and why, and why some have a lower Wilson ratio and become superconducting.

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