Specific heat in a magnetic field: A probe of the magnetic ground-state properties of heavy-fermion $Ce(Ru_{2-x}Rh_x)Si_{2-y}Ge_y$

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Specific-heat data up to 12.5 T are presented for $CeRu_2Si_2$, $Ce(Ru_{0.5}Rh_{0.5})_2Si_2$, and $CeRu_2Si_{1.8}Ge_{0.2}$. The behaviors with field of the specific heats are compared, also with susceptibility data. The possibility that the maximum in C/T, or γ , at $H_{\text{metamagnetic}} = 8$ T in $CeRu_2Si_2$ corresponds to an increase in m^* at the metamagnetic transition is discussed. Similar data for CeB_6 are compared.

I. INTRODUCTION

The nearness of heavy-fermion systems (HFS's) to magnetic behavior has long been clear.¹ Inverse magnetic susceptibility versus temperature above 100 K behaves linearly for HFS's, i.e., follows a Curie-Weiss behavior, giving evidence for an $\sim 3\mu_B$ local moment. This moment is, for HFS's, at least partially compensated during the formulation of the high-effective-mass heavy-fermion ground state at lower temperatures. Some HFS's actually order antiferromagnetically, e.g., UCd_{11} (Ref. 2) and U_2Zn_{17} , ³ while UPt₃ displays evidence^{4,5} of (antiferromagnetic) spin fluctuations as does (Ref. 6) CeCu₆. Doping leads to magnetic ordering in both UPt₃ (Refs. 7-9) and CeCu₆ (Refs. 10-12) at low temperatures. This connection between magnetism and the heavy-fermion ground state has been exploited in order to manufacture pseudobinary heavy-fermion systems near in composition to magnetic order, e.g., U(In,Sn)₃.¹³

In all of these systems, the strong nearly magnetic behavior of UPt₃ coupled with its bulk superconductivity⁴ at 0.5 K has remained a focus of interest. UPt₃ can, with good reason, be called the most nearly magnetic of all known HFS's based not only on its spin fluctuation ground state,⁴ its⁵ nonzero frequency ordered magnetic moment of $0.02\mu_B$, and its rapid conversion to a magnetically ordered ground state with doping,⁷⁻⁹ but also based on the metamagnetic transition at ~ 20.5 T applied field. Recently, another system has been discovered with very similar properties, except for superconductivity. $CeRu_2Si_2$, according¹⁴ to neutron scattering measurements, has short-range magnetic correlations that become fully established below 15 K, undergoes¹⁵ a metamagnetic transition at ~8 T, has¹⁶ a large γ at low temperatures of 385 mJ/mol K² (versus 450 mJ/mol K² for UPt₃), and orders antiferromagnetically upon doping on any of the three sites $[Ce_{0.9}La_{0.1}Ru_2Si_2 \text{ orders}^{17} \text{ at } 2.7 K$, $Ce(Ru_{0.4}Rh_{0.6})_2Si_2$ orders¹⁸ at 11 K, and $CeRu_2Si_{1.5}Ge_{0.5}$ orders¹⁹ at 8 K]. We report here measurements of dc magnetic susceptibility in fields up to 5.5 T and, for the first time, low-temperature specific-heat measurements in fields up to 12 T on polycrystalline (where¹⁸ CeRu₂Si₂, $Ce(Ru_{0.5}Rh_{0.5})_2Si_2$ $\gamma = 550$ mJ/mol K²), and CeRu₂Si_{1.8}Ge_{0.2} (where²⁰ $\gamma = 600$ mJ/mol K²) in order to more fully understand this metamagnetic transition, and possibly, the reasons for the occurrence of superconductivity in UPt₃. Comparison of these data will be made with our recent 18 T specific-heat data²¹ on single-crystal UPt₃, which are being extended to 23 T by several groups.

II. EXPERIMENTAL RESULTS AND DISCUSSION

The samples were prepared by arc melting together high-purity starting materials using the highest available purity Ce, that from Ames Laboratory. We have found large differences in sample properties, particularly in the dc magnetic susceptibility, even using Reacton[™] grade Ce from Johnson Matthey. The Ru, Rh, Si, and Ge were all from Johnson Matthey and were 99.99% purity or better.

The question of preferential orientation of these polycrystalline samples is of importance. Previous work²² on single-crystal tetragonal CeRu₂Si₂ shows $\chi(4.2)$ $K_{c} \simeq 35 \times 10^{-3}$ emu/mol for H along the c axis, while $\chi(4.2 \text{ K}) \simeq 2.6 \times 10^{-3} \text{ emu/mol for } \tilde{H}$ perpendicular to the *c* axis. Additionally, there is $\frac{1}{2}$ a slight $([\chi^{\text{peak}} - \chi(4.2 \text{ K})]/\chi^{\text{peak}} \simeq 1\%)$ peak in χ at $\sim 10 \text{ K}$ for H parallel to c, with no such feature for H perpendicular to c. Our polycrystalline CeRu₂Si₂ sample shows some preferential alignment, with a slight peak in χ at 10 K visible for all orientations of the polycrystalline sample and a factor of 2 difference (9 versus 20×10^{-3} emu/mol) in χ at 1.8 K obtainable for different orientations. Preferred orientation is also apparent in our susceptibility measurements on CeRu₂Si_{1.8}Ge_{0.2} and $Ce(Ru_{0.5}Rh_{0.5})_2Si_2$. Thus, where changes in the magnitude of χ or C with field occur, in this work they will be less accentuated than in single-crystal results. In the cases where no change with field occurs, the results of this work are of general applicability. As will be seen later, the magnitude of the change of a property with field (unless there is no change) will not play a central role in understanding these systems.

A. Susceptibility data

In general, $CeRu_2Si_2$, $Ce(Ru_{0.5}Rh_{0.5})_2Si_2$, and $CeRu_2Si_{1.8}Ge_{0.2}$ have already been characterized by oth-

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ers^{15,18-20,22} via dc magnetic susceptibility measurements. Thus, we present susceptibility data here for our samples primarily as an introduction to the central issues addressed by the specific-heat data discussed in the following. One of the motivations for doping $CeRu_2Si_2$ is to vary the metamagnetic transition. Pure $CeRu_2Si_2$ has²² an M versus H behavior that, in the H parallel to c direction, is linear below 6 T and again above 12.5 T, while there is a steep increase in the slope of M versus $H(=\gamma)$ in between whose steepest slope is at 8 T. Magnetoresistance measurements²² show a peak in $[\rho(H) - \rho(0)]/\rho(0)$ also at 8 T. (This may be compared to UPt₃, where the steepest part of the *M* versus *H* transition²³ and the peak in the magnetoresistivity are²⁴ at 20.5 T in one measurement, with some sample dependence, i.e., ρ peak is at 18.5 T, more recently reported.²¹)

Upon doping with Ge, but below the Ge concentration where antiferromagnetism begins to occur, we observe not one but two steps in M versus H (see Fig. 1), with the first transition narrower in field. (Similar results on CeRu₂Si_{2-y}Ge_y have already been reported.²⁰) The fields at which these transitions occur in CeRu₂Si_{2-x}Ge_x are, of course, composition dependent. We have arbitrarily chosen to focus on CeRu₂Si_{1.8}Ge_{0.2}, since at this composition both transitions are just in the range of the upper field (5.5 T) of our Quantum Design dc magnetometer. Upon increasing doping, the transitions occur at lower fields but become less pronounced and broader.

In addition to depressing the field H^* where the metamagnetic transition occurs in pure CeRu₂Si₂, we have found that Ge doping also depresses the temperature where the peak in the magnetic susceptibility occurs (10 K in the parent compound versus 7 K for Ge=0.2), accentuates the peak,

 $[\chi(7 \text{ K}) - \chi(1.8 \text{ K}) / \chi(1.8 \text{ K}) = 32\%$

in our polycrystalline sample, and raises the magnitude of χ in our polycrystalline specimens versus pure CeRu₂Si₂ by a factor of 2 (see Fig. 2). This may be precursor fluctuations setting in before the long-range magnetic order



FIG. 1. Magnetization vs field for $CeRu_2Si_{1.8}Ge_{0.2}$. There are two increases in *M* as a function of *H*, the one at lower field being sharper and easier to discern. The line is a guide to the eye.



FIG. 2. Susceptibility χ vs temperature for polycrystalline CeRu₂Si_{2-y}Ge_y. Note the accentuation of the slight peak at 10 K for y = 0 upon addition of Ge. The magnitude of χ for CeRu₂Si_{1.8}Ge_{0.2} is larger than that of pure CeRu₂Si₂, even allowing for uncertainties in alignment of our polycrystalline sample.

at 7.5 K reported²⁵ for pure CeRu₂Ge₂.

The reason that $Ce(Ru_{2-x}Rh_x)Si_2$ is of interest is that (1) the γ has been reported¹⁸ to be quite large, ~600 mJ/mol K^2 , and (2) neutron scattering data¹⁸ imply that $Ce(Ru_{0.4}Rh_{0.6})_2Si_2$ is an ordered antiferromagnet with a local moment of $0.65\mu_B$ and $T_N = 11$ K, while these neumagnetic behavior tron data show no for $Ce(Ru_{0.5}Rh_{0.5})_2Si_2$. (CeRh₂Si₂ is antiferromagnetic at 36 K). Such a large γ near, in "composition space," to magnetism is similar to the behavior¹³ of U(In,Sn)₃, and although our M versus H data show no transition up to 5.5 T, and there is no peak in χ versus T (see Fig. 3), investigating the behavior of the specific heat with field of a large γ system related to CeRu₂Si₂ might shed further light on the magnetic properties of this parent system.

B. Specific heat

Presented here are specific heat divided by temperature (C/T) data for 0 and 12 T for CeRu₂Si₂ (Fig. 4) and for



FIG. 3. χ vs T for Ce(Ru_{0.5}Rh_{0.5})₂Si₂. Rather than a peak in χ vs T, as seen in Figs. 1 and 2, a change in slope occurs at around 7 K.



FIG. 4. Specific heat divided by temperature vs temperature squared for $CeRu_2Si_2$ in 0 (circles) and 12 T (triangles). The zero-field data agree with the literature. The inset shows specific heat as a function of temperature for $CeRu_2Si_2$ (circles) and $CeRu_2Si_2$, with the specific heat of $LaRu_2Si_2$ subtracted (squares).

Ce(Ru_{0.5}Rh_{0.5})₂Si₂ (Fig. 5), with an inset for the zero-field C versus T plot. Figure 6 shows C/T versus T^2 data for several fields between 0 and 10 T for CeRu₂Si_{1.8}Ge_{0.2}, while Fig. 7 shows the same data plotted as C versus T. We will present the behavior of C/T at low temperature (1.5 K) as a function of field with much finer gradations in H. The only specific-heat data already present in the literature on these systems, to our knowledge, for comparison are zero field up to 7.5 T specific-heat data (i.e., to just below the metamagnetic transition) for single-crystal CeRu₂Si₂ (Ref. 26) and zero-field, low-temperature (T < 2 K) data¹⁸ for Ce(Ru_{0.5}Rh_{0.5})₂Si₂. These data are in agreement with that of this work, except for the size of



FIG. 5. Specific heat divided by temperature vs temperature squared for $Ce(Ru_{0.5}Rh_{0.5})_2Si_2$ at 0 (circles) and 12 T (triangles). Note the agreement of the 0 and 12 T data above 5 K, in contrast to both $CeRu_2Si_2$ and $CeRu_2Si_{1.8}Ge_{0.2}$. The insert show C vs T for $Ce(Ru_{0.5}Rh_{0.5})_2Si_2$. Note the anomaly at 11 K.



FIG. 6. Specific heat divided by temperature vs temperature squared of $CeRu_2Si_{1.8}Ge_{0.2}$ as a function of magnetic field. The peak seen in C/T at around 2 K moves up in temperature and broadens with increasing field. Clearly, field has a much stronger effect on C/T for $CeRu_2Si_{1.8}Ge_{0.2}$ than for $CeRu_2Si_2$ and $Ce(Ru_{0.5}Rh_{0.5})_2Si_2$.

the change in γ with field for CeRu₂Si₂ due to preferential orientation effects discussed earlier.

Can these specific-heat results shed light on the cause of the various peaks in magnetic susceptibility discussed above? Clearly, the very slight peak at 10 K in χ for CeRu₂Si₂ does not correspond to magnetic order neutron measurements¹⁴ show nothing, as is the case for the C/T versus T^2 plot of our data in Fig. 4. A C versus T plot, inset to Fig. 4, with the specific heat¹⁵ of LaRu₂Si₂ subtracted, does show a peak that is consistent with the presence of short-range magnetic correlations below 15 K indicated by the neutron scattering experiment.¹⁴ [That



FIG. 7. Specific heat vs temperature as a function of field for $CeRu_2Si_{1.8}Ge_{0.2}$. The shift of the peak at 6 K in C vs T is only slightly downward in temperature with 3 T and only slightly upward in temperature with 5 T, whereas 10 T shifts the peak up to ~10 K.

is, magnetic correlations appear more weakly in the specific heat than magnetic order. Magnetic order contributes an entropy, which requires an anomaly in C/T since $S = \int (C/T) dT$. Magnetic correlations may have an energy of formation (where C = dE/dT) that is temperature dependent as the magnetic exchange overcomes the thermal energy of disorder.]

The claim that $Ce(Ru_{0.5}Rh_{0.5})_2Si_2$ is nonmagnetic is likely correct (although neutron scattering experiments may not detect small local moment ordering); however, there is a small anomaly in the *C* versus *T* data (inset, Fig. 5) at the same tempearture, 11 K, as where neutron scattering measurements¹⁸ find a local moment of significant size, $0.65\mu_B$, in $Ce(Ru_{0.4}Rh_{0.5})_2Si_2$. [It is not actually clear in Ref. 18 if neutron scattering measurements were performed on the $Ce(Ru_{0.5}Rh_{0.5})_2Si_2$ composition.] This small specific-heat anomaly does not correspond to any peak in the susceptibility of $Ce(Ru_{0.5}Rh_{0.5})_2Si_2$, but instead our χ data (see Fig. 3) show a shoulder beginning at about 8 K.

It remains then to discuss our specific-heat data between 0 and 10 T for $CeRu_2Si_{1.8}Ge_{0.2}$ and to try to present a coherent picture that will link these data to already discussed for CeRu₂Si₂ those and The C/T versus T^2 data for $Ce(Ru_{0.5}Rh_{0.5})_2Si_2$. $CeRu_2Si_{1.8}Ge_{0.2}$ are presented in Fig. 6, while the C versus T data are shown in Fig. 7. Although similiar in magnitude to the specific heat of $Ce(Ru_{0.5}Rh_{0.5})_2Si_2$, the occurrence of a peak in C versus T around 6 K and the decrease of C/T below some temperature in high field in the Ge-doped sample finds similar behavior in the pure $CeRu_2Si_2$. Since the peak in C versus T for $CeRu_2Si_2$ appears to be connected to the magnetic correlations observed via neutron scattering,¹⁴ and since the peak in the specific-heat data, Fig. 7, for the Ge-doped sample is more pronounced than that for the pure CeRu₂Si₂, it is tempting to predict that CeRu₂Si_{1.8}Ge_{0.2} will be found to display stronger magnetic correlations in a similar temperature range when neutron scattering experiments are performed on this material. This is consistent with the lower field needed (~ 1.2 T) to induce a metamagnetic transition in the Ge-doped magnetization data than the ~8 T required in CeRu₂Si₂.

It is thus these stronger magnetic correlations in $CeRu_2Si_{1.8}Ge_{0.2}$ that cause the greater decrease of γ at low temperature with field observed compared with $CeRu_2Si_2$; Figs. 8 and 9: a 54% decrease in the Ge-doped material by 10 T versus only a 17% decrease in $CeRu_2Si_2$ by 12 T and a 29% decrease in $Ce(Ru_{0.5}Rh_{0.5})_2Si_2$, Fig. 10. A concomitant observation is that it is then these magnetic correlations that cause the large γ for $CeRu_2Si_{1.8}Ge_{0.2}$ of 590 mJ/mol K² at low temperature compared with 376 mJ/mol K² for $CeRu_2Si_2$.

Before we discuss the behavior of the low-temperature γ versus *H* through the metamagnetic transition field, what is the explanation of the peak at 2 K in *C*/*T* for the Ge-doped sample, Fig. 6? In previously published zero-field specific-heat data²⁷ for pure CeRu₂Si₂ down to 0.1 K, no sign of any peak was observed. With field, this peak in *C*/*T* in CeRu₂Si_{1.8}Ge_{0.2} moves to higher temper-



FIG. 8. Specific heat divided by temperature at T = 1.5 K vs H for CeRu₂Si_{1.8}Ge_{0.2} Within 2%, our precision in the present type of measurement, no structure in γ is seen until the first (see Fig. 1) field (1.2 T), where a metamagnetic transition occurs, whereupon γ begins to drop sharply with increasing field.

atures and broadens (see Fig. 6). Thus, it is unlikely to be a Fermi-liquid effect of highly correlated Ce 4f electrons (these create¹ a peak in C/T at 0.35 K in CeAl₃ which is not qualitatively nearly so field dependent). A magnetic order explanation is unlikely—no anomaly in C versus T is observed, plus the peak temperature in C/T increases with field, which would be some sort of ferrimagnetic or ferromagnetic coupling. Therefore, some saturation effect of the stronger magnetic-correlation-induced increase in the effective mass present in CeRu₂Si₂ doped with Ge may be a possible explanation.

 γ versus H. Figures 8, 9, and 10 detail the behavior of C/T at 1.5 K for each sample as a function of field.



FIG. 9. Specific heat divided by temperature at T=1.5 K vs H for CeRu₂Si₂. The 6% rise in γ between 0-8 T would be (Ref. 26) more pronounced in a single crystal with H in the C direction. The fall off of γ in CeRu₂Si₂ above 8 T is reported here for the first time.



FIG. 10. Specific heat divided by temperature at T=1.5 K vs H for Ce(Ru_{0.5}Rh_{0.5})₂Si₂. Note that, although no anomaly in M vs H is observed for this alloy, again two regions of behavior are observed: at low H, γ is \sim constant, while above 5 T γ falls sharply. Although the low temperature C/T values for Ce(Ru_{0.5}Rh_{0.5})₂Si₂ and CeRu₂Si_{1.8}Ge_{0.2} are comparable, the rate of the fall off of C/T with H is approximately a factor of 2 different.

These field data (γ versus H) in related systems that have variously one ($CeRu_2Si_2$), two ($CeRu_2Si_{1.8}Ge_{0.2}$), or no $[Ce(Ru_{0.5}Rh_{0.5})_2Si_2]$ metamagnetic transitions up to 5.5 T were the primary goal of this work. [The higher fields (>20 T) required to examine γ versus H through such a transition in UPt₃ have thus far²¹ (coupled with the vagaries of reliability of such very high-field normal-state magnets) prevented us from such a study]. We see immediately from Figs. 8-10 several correlations. At the metamagnetic transitions at 8 T in CeRu₂Si₂ (Fig. 9) and at 1.2 T in CeRu₂Si_{1.8}Ge_{0.2} (Fig. 8), a sharp drop in γ at higher fields is observed. The larger and sharper the anomaly in M versus H, the greater is the change in behavior of γ through the transition. Thus, referring to Figs. 1 and 8, we see that the broad anomaly in M versus H at 3.7 T for $CeRu_2Si_{1.8}Ge_{0.2}$ does not produce a (visible) corresponding γ versus H anomaly. Further, the much more distinct M versus H anomaly in CeRu₂Si₂ actually corresponds to a distinct peak in γ versus H at the same field (Fig. 9). This peak, whose onset up to 7.5 T has been reported,²⁶ is evidence of the complexity of this system.

If one adopts the simple antiferromagnetic correlation picture as being the important cause of magnetic field effects [and also of the peak in the specific heat of CeRu₂Si₂ (Fig. 4, inset) at ~8 K], then the explanation of the γ versus *H* data in Fig. 9 is straightforward. The entropy due to these antiferromagnetic correlations is suppressed in temperature with increasing field. As the anomaly and its associated entropy pass (however broadened) through 1.5 K, " γ " ($\equiv C/T$) is increased. At higher fields, the antiferromagnetic correlations are totally suppressed, and γ falls precipitously. Thus, by this model γ at low temperatures is not really tracking m^* , the electron effective mass, since a broadened anomaly in the specific heat due to the entropy of antiferromagnetic correlations is the (at least partial) source of the variation in C/T. The analog of this behavior is the peak in C versus T at 2 K in UBe_{13} . The entropy under this as yet not understood anomaly is substantial (Ref. 1), ~ 1 J/mol K. This peak moves²⁸ upon doping the U site; thus, measuring C/T in a heavy-fermion system at some low temperature may not give a clean determination of m^* ($\propto \gamma_{\text{electronic}}$) due to the admixture of an additional magneticspecific-heat contribution. Since the correlation entropy observed in CeRu₂Si₂ at $T \sim 9$ K (see inset, Fig. 4) can, under suppression by magnetic field, have a finite contribution to the specific heat at arbitrarily low temperature, it is premature to conclude that m^* , as would be measured by dHvA, first increases and then decreases as H is varied through a metamagnetic transition. Since dHvA measurements are difficult to achieve at this low (8 T) a field, the question of the real behavior of m^* through the metamagnetic transition in CeRu₂Si₂ is likely to remain moot.

This view, that m^* versus H may be difficult to determine from the specific heat of a heavy-fermion system due to non- m^* -producing specific-heat contributions which mask the real γ value, is not universally held. The cause of the creation of the large m^* in heavy-fermion systems is still an open question. Magnetic fluctuations have been experimentally proven to^{5,6} exist in heavyfermion systems and, indeed, in (Ref. 14) CeRu₂Si₂, via neutron scattering experiments, and are visible in other measurements as well, including specific heat and susceptibility. Theoretically, it has been proposed²⁹ that such fluctuations actually are partially responsible for the creation of a large m^* at low temperatures. Thus, in $CeRu_2Si_2$ it has been proposed³⁰ that the increase in C/Tat T = 0.4 K seen²⁶ up to 7.5 T, (which mirrors our own 1.5 K results; Fig. 9) is in fact proof of an increasing m^* , due to an "increase of the ferromagnetic coupling." At higher fields above the metamagnetic transition, then this model would have m^* fall as the fluctuations are eventually suppressed with ever-increasing field. For comparison, dHvA are available³¹ for CeB₆ above 13 T, and field data for C/T of CeB₆ are available³² up to 22 T. In the region where both kinds of measurement overlap ($H \ge 13$ T), the determination of γ from the measured m^* from the dHvA data agrees well with the measured specific heat. C/T data well below 0.4 K are necessary³³ to determine γ in CeB₆, however, due to the strong influence on C/T (there is still curvature in C/T at 0.4 K in H=0) from the large anomaly in C at 2.3 K from the antiferromagnetic transition. It is not clear, a priori, to how low a temperature C/T must be measured in CeRu₂Si₂ to avoid distortion due to the magnetic fluctuation peak in C at 9 K and H = 0. The peak broadens as it is suppressed with field and is difficult to identify.

In any case, no measurable rise in C/T at low temperatures is observed at the metamagnetic transitions in $CeRu_2Si_{1.8}Ge_{0.2}$. A slight rise in C/T at 0.4 K versus H is observed¹⁷ in $Ce_{0.9}La_{0.1}Ru_2Si_2$. However, the data of Ref. 17 show very clearly that this rise is due to a peak in C/T at 2 K becoming more pronounced in height and shifting to lower temperature with increasing field. In summary, $CeRu_2Si_2$ is an interesting high- γ system which, upon doping either the Ru or the Si site, has an increasing γ and, eventually, magnetic order. Specificheat measurements in field of $Ce(Ru_{2-x}Rh_x)Si_{2-y}Ge_y$ show very strong suppression of C/T (T = 1.5 K) by field in $CeRu_2Si_{1.8}Ge_{0.2}$ (54% by 10 T) and a peak in C/Tversus H at 8 T in $CeRu_2Si_2$, where M versus H data show an anomaly. However, no such peak in γ versus H is observed in $CeRu_2Si_{1.8}Ge_{0.2}$ despite the existence

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therein of two anomalies in M versus H (so-called "metamagnetic" transitions).

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