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# Multiple phase transitions in $CeRu_2(Si_{1-x}Ge_x)_2$

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**Abstract.** The magnetic-phase diagram of the system $CeRu_2(Si_{1-x}Ge_x)_2$ is studied by specific-heat, magnetic-susceptibility and electrical resistance measurements ($x = 0.1$, 0.2, 0.3, 0.6, 0.8, 0.9, 1.0). The susceptibility $\chi$ was measured by zero-field-cooling (ZFC) and field-cooling (FC) methods. If $x < 0.1$, there is no clear long-range magnetic order. When $x > 0.1$, $\chi_{FC}(T)$ starts to deviate from $\chi_{ZFC}(T)$ at $T_{sf}$. This suggests a spin-glass state below $T_{sf}$. In the range $0.9 \geqslant x \geqslant 0.2$, the specific-heat measurements of $CeRu_2(Si_{1-x}Ge_x)_2$ show multiple magnetic phase transitions. There are three phase transitions in $CeRu_2Si_{0.4}Ge_{1.6}$ and $CeRu_2Si_{0.2}Ge_{1.8}$. Except for $x = 0$, below 30 K, electrical resistivity $\rho(T)$ increases continuously up to a temperature of 10 K. The drastic drop of $\rho(T)$ below 10 K is due to magnetic ordering. The multiple phase transitions of $CeRu_2(Si_{1-x}Ge_x)_2$ are not observed in the resistance measurements. However, the slope of $\rho(T)/\rho(285\ \mathrm{K})$ of $CeRu_2Si_{0.4}Ge_{1.6}$ changes at $\sim$7 K, which further supports multiple phase transitions in $CeRu_2(Si_{1-x}Ge_x)_2$.

## 1. Introduction

Among cerium compounds that crystallize in the $ThCr_2Si_2$ structure, $CeRu_2Si_2$ and $CeRu_2Ge_2$ have attracted considerable interest. $CeRu_2Si_2$ is one of the few cases of heavy-fermion compounds in which no evidence of long-range magnetic ordering has been found down to $\sim$20 mK [1, 2]. However, $CeRu_2Ge_2$ orders ferromagnetically [3] at 7.9 K with slightly enhanced low-temperature electronic specific heat of 20 mJ $mol^{-1}$ $K^{-2}$.

Both compounds are widely studied. The difference between $CeRu_2Si_2$ and $CeRu_2Ge_2$ is caused by the different ground state. The formation of a ground state is dominated by the electronic structure of the compound and the stability of 4f electrons [4]. $CeRu_2Ge_2$ exhibits a pure magnetic ground state with well localized 4f electrons [3]. In the ground state of $CeRu_2Si_2$, the interaction between 4f electrons and conduction electrons prevents long-range magnetic ordering. Loidl *et al* [4] claimed that when the Si of $CeRu_2Si_2$ is substituted by Ge the band width of the conduction electron will increase and change the hybridization of the f band and the conduction band. Therefore, long-range magnetic ordering in $CeRu_2Si_2$ may be restored.

The susceptibility of $CeRu_2(Si_{1-x}Ge_x)_2$ ($x = 0.05$, 0.1, 0.25, 0.5, 0.75, 1.0) was first reported by Godart *et al* [5] $CeRu_2Si_{2-x}Ge_x$ exhibits a variety of magnetic behaviour. With small concentrations ($x \sim 0.05$) of Ge, the magnetic susceptibility increases without any magnetic ordering. With a further increase in $x$, the system undergoes a magnetic transition, the nature of which depends upon the value of $x$. In the report of Godart *et al*, the susceptibility of $CeRu_2SiGe$ exhibits a rather complex behaviour and shows a peak at 9 K. They mentioned that there were two kinds of order or spin-glass behaviour around 9 K. However, in $CeRu_2Si_{0.5}Ge_{1.5}$ and $CeRu_2Si_{1.5}Ge_{0.5}$, only simple magnetic

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**Table 1.** The lattice parameters of $CeRu_2(Si_{1-x}Ge_x)_2$.

| Compound | $a$ (Å) | $c$ (Å) | $c/a$ | $V$ (Å$^3$) |
|---|---|---|---|---|
| $CeRu_2Si_2$ | 4.19 | 9.80 | 2.339 | 172.05 |
| | 4.195 [5] | 9.976 [5] | 2.336 [5] | 172.17 [5] |
| $CeRu_2Si_{1.8}Ge_{0.2}$ | 4.201 | 9.81 | 2.335 | 173.13 |
| $CeRu_2Si_{1.6}Ge_{0.4}$ | 4.207 | 9.821 | 2.334 | 173.82 |
| $CeRu_2Si_{1.4}Ge_{0.6}$ | 4.214 | 9.853 | 2.338 | 174.97 |
| $CeRu_2Si_{0.8}Ge_{1.2}$ | 4.227 | 9.943 | 2.352 | 177.66 |
| $CeRu_2Si_{0.4}Ge_{1.6}$ | 4.255 | 10.037 | 2.359 | 181.720 |
| $CeRu_2Si_{0.2}Ge_{1.8}$ | 4.261 | 10.011 | 2.349 | 181.76 |
| $CeRu_2Ge_2$ | 4.273 | 10.054 | 2.352 | 183.57 |
| | 4.269 [5] | 10.035 [5] | 2.351 [5] | 182.88 |
| | 4.268 [7] | 10.07 [7] | | |

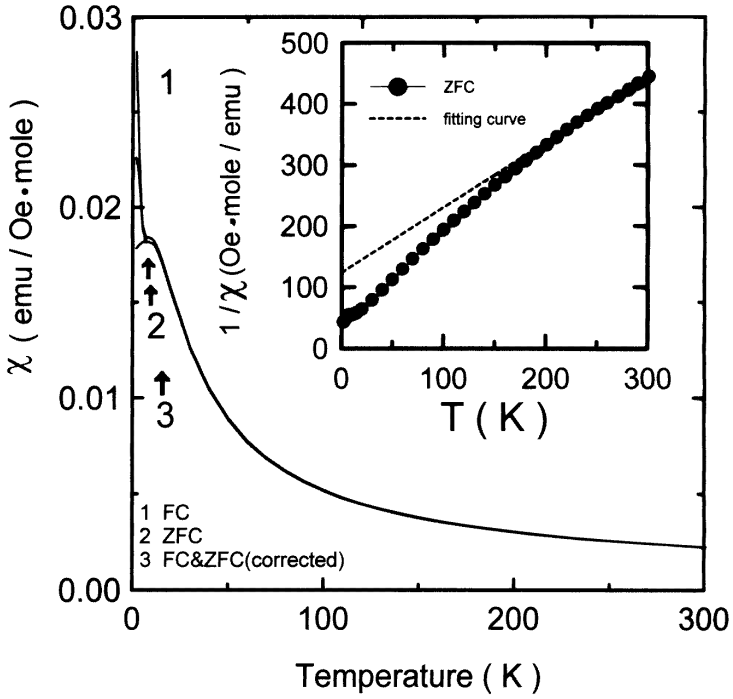


**Figure 1.** The temperature dependence of the molar susceptibility $\chi$ for $CeRu_2Si_2$ at 100 gauss. The susceptibility was measured by both field-cooling (curve 1) and zero-field-cooling (curve 2) methods. Curve 3 is the susceptibility after the correction of the impurity contribution. The inset is the temperature dependence of the susceptibility $1/\chi$ (zero-field cooling) between 2 and 300 K.

ordering is observed. $CeRu_2Si_{1.5}Ge_{0.5}$ exhibits antiferromagnetic ordering below 8 K and $CeRu_2Si_{0.5}Ge_{1.5}$ shows ferromagnetic ordering below 10 K.

It is interesting to study the competition between the ferromagnetic and antiferromagnetic interaction in $CeRu_2(Si_{1-x}Ge_x)_2$ (with $0.5 < x < 1.0$). Except susceptibility measure-

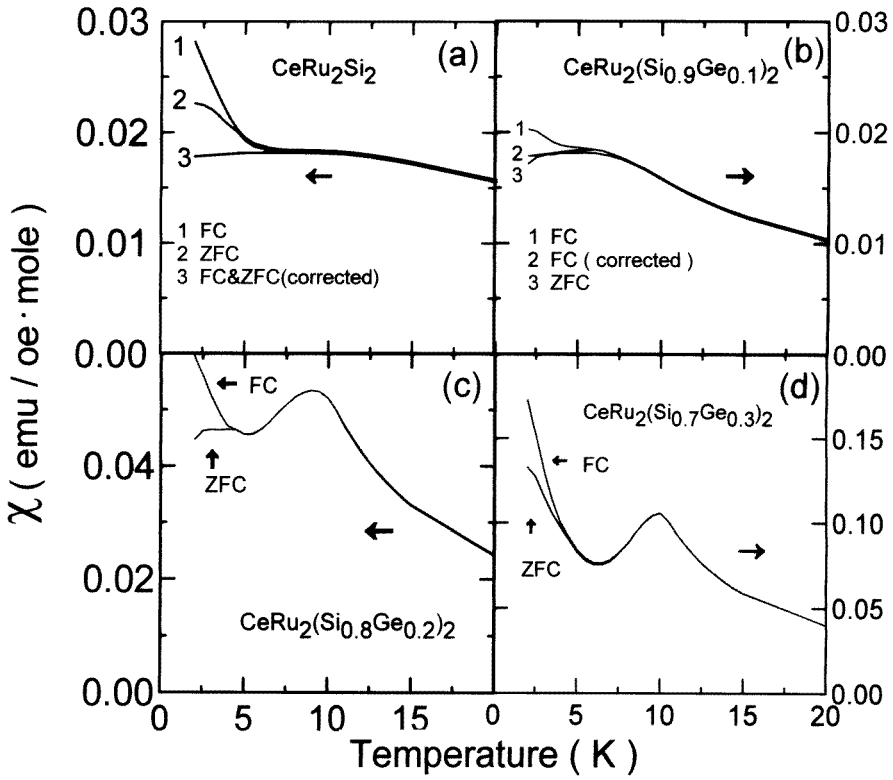


**Figure 2.** The temperature dependence of the molar susceptibility $\chi$ for $CeRu_2Si_2$, $CeRu_2Si_{1.8}Ge_{0.2}$, $CeRu_2Si_{1.6}Ge_{0.4}$ and $CeRu_2Si_{1.4}Ge_{0.6}$ at 100 gauss. The susceptibility was measured by both field-cooling (FC) and zero-field-cooling (ZFC) methods between 2 and 20 K.

ments, the $CeRu_2(Si_{1-x}Ge_x)_2$ system has not been systematically studied. The specific heat of $CeRu_2Si_{1.8}Ge_{0.2}$ was reported by Kim *et al* [6] In this paper, we report susceptibility, specific-heat and electrical resistance measurements on $CeRu_2(Si_{1-x}Ge_x)_2$ ($x = 0.1$, 0.2, 0.3, 0.6, 0.8, 0.9, 1.0). From these experiments a magnetic-phase diagram is constructed.

## 2. Experimental results

The preparation of $CeRu_2(Si_{1-x}Ge_x)_2$ was clearly described by Godart *et al* [5] Polycrystalline samples of $CeRu_2(Si_{1-x}Ge_x)_2$ were prepared by arc-melting the pure elements in their stoichiometric ratio in an atmosphere of purified argon gas. The button was flipped several times and remelted to achieve good homogeneity. The samples were annealed at 1100 K for 8 days. The overall weight loss during melting was less than 1%. X-ray measurements of the sample were carried out at room temperature and showed only a single phase. The structure is consistent with tetragonal space group $I4/mmm$. The lattice parameters $a$ and $c$ are shown in table 1. As shown in table 1, both $a$ and $c$ increase with germanium concentration.

The magnetization studies were performed in a superconducting quantum-interference device (SQUID) magnetometer. The susceptibility was measured by both the zero-field-

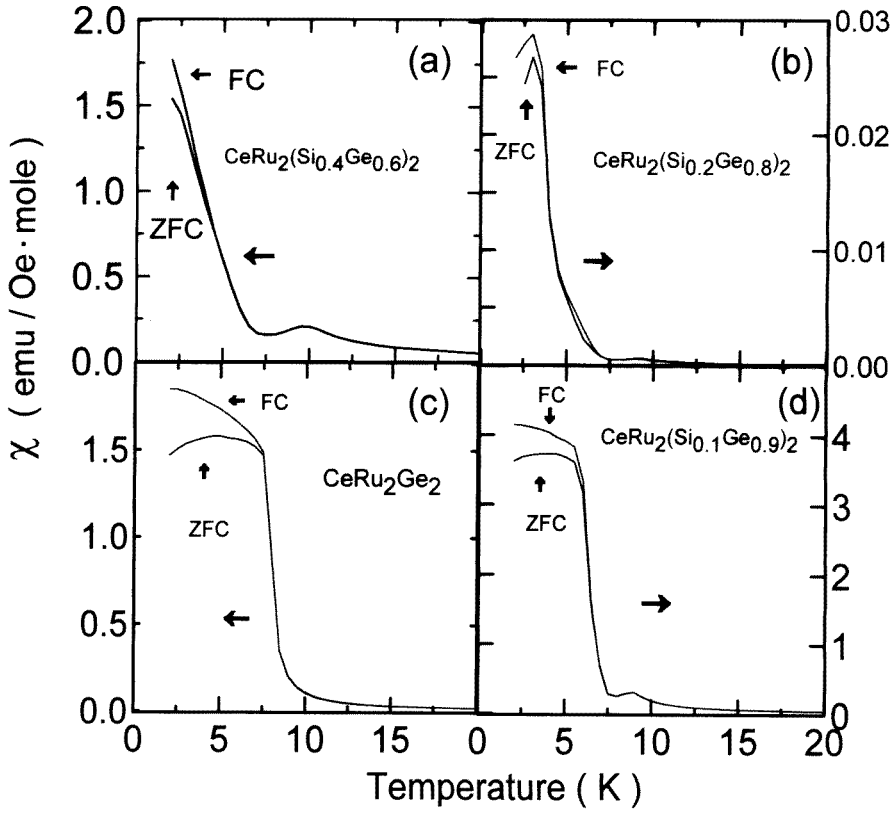


**Figure 3.** The temperature dependence of the molar susceptibility $\chi$ for $CeRu_2Si_{0.8}Ge_{1.2}$, $CeRu_2Si_{0.4}Ge_{1.6}$, $CeRu_2Ge_2$ and $CeRu_2Si_{0.2}Ge_{1.8}$ at 100 gauss. The susceptibility was measured by both field-cooling (FC) and zero-field cooling (ZFC) methods between 2 and 20 K.

cooling (ZFC) and the field-cooling (FC) methods. For ZFC, we cooled the sample from 300 to 2 K in the zero field and applied a field at 2 K. Then we heated the sample while measuring $\chi$ in the constant field. For FC, the sample was cooled in a magnetic field from 300 to 2 K and then heated up while measuring $\chi$. Figure 1 is the temperature dependence of the molar susceptibility $\chi$ for $CeRu_2Si_2$ at 100 gauss. For FC, below 6 K, the $\chi_{FC}$ of $CeRu_2Si_2$ exhibits a sharp rise with decreasing temperature, which is presumably due to small contribution of free $Ce^{3+}$ ions situated on grain boundaries or other defects reported by Gupta *et al* [1]. After the correction of the impurity contribution, $\chi_{FC}(T)$ has a broad peak near 8 K and saturates at low temperature. After the correction, $\chi_{FC}(T)$ is the same as $\chi_{ZFC}(T)$. Above 200 K, $\chi(T)$ of $CeRu_2Si_2$ follows the Curie–Weiss form $\chi(T) = C/(T + \Theta)$, with $\Theta = 80$ K and $\mu_{eff} = 2.54\ \mu_B$. As shown in figure 2(b), with small $x$ ($\sim$0.1), after the correction of the impurity contribution, a broad peak of $\chi_{FC}(T)$ appears at $\sim$6 K. There is no strong evidence to indicate a magnetic order in $CeRu_2Si_{1.8}Ge_{0.2}$. The corrected $\chi_{FC}(T)$ starts to deviate from $\chi_{ZFC}(T)$ at low temperature($T < 3$ K), which might suggest magnetic order at low temperature.

Although the ground state of $CeRu_2Si_2$ does not exhibit long-range order, it is a compound with antiferromagnetic instability [7]. The current idea is that $CeRu_2Si_2$ is just on

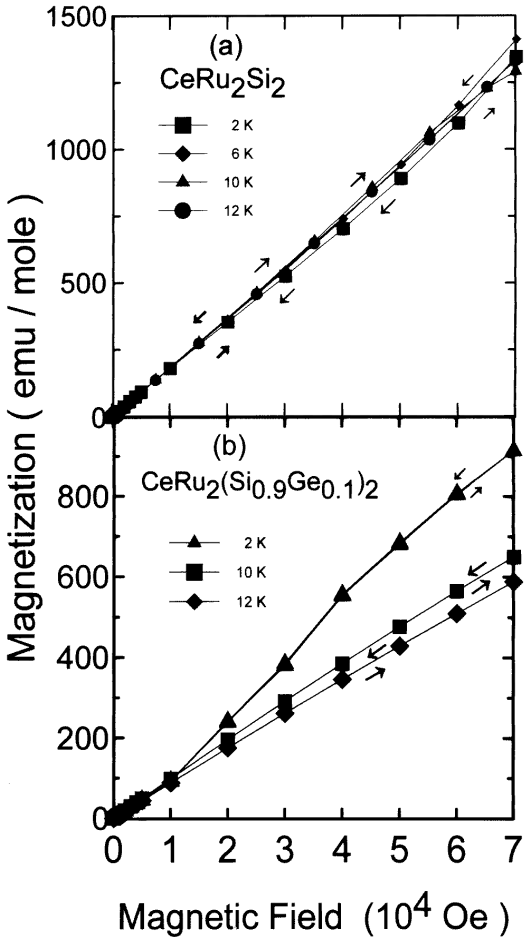


**Figure 4.** The field dependences of the magnetization $M(H)$ of $CeRu_2Si_2$ and $CeRu_2Si_{1.8}Ge_{0.2}$ at 2, 6, 10 and 12 K.

the non-magnetic to magnetic transition which can be induced by pressure [8]. Therefore, we might dope with germanium to restore a long-range magnetic ordering.

With a further increase in $x$ ($\sim$0.2), $CeRu_2Si_{1.6}Ge_{0.4}$ exhibits a rather complex behaviour (figure 2) in $\chi(T)$. After the 9 K antiferromagnetic transition, $\chi(T)$ increases again at $\sim$5.4 K. Besides, $\chi_{FC}(T)$ starts to deviate from $\chi_{ZFC}(T)$ at $\sim$5 K. This suggests a spin-glass state below 5 K. The spin-glass type order might be due to the competition between the ferromagnetic and antiferromagnetic interaction in $CeRu_2Si_{1.6}Ge_{0.4}$. A much clearer multiple transition can be found in the $\chi(T)$ of $CeRu_2Si_{1.4}Ge_{0.6}$ (figure 2). $\chi(T)$ of $CeRu_2Si_{1.4}Ge_{0.6}$ clearly indicates an antiferromagnetic transition at $T_N \sim 9$ K and a spin-glass transition at $T_{sf} \sim 4$ K. As shown in figure 3(a), with further increasing germanium concentration, the antiferromagnetic order is depressed and the ferromagnetic order is enhanced in $CeRu_2Si_{0.8}Ge_{1.2}$. If the concentration of germanium is much higher than that of silicon, for example in $CeRu_2Si_{0.4}Ge_{1.6}$, a clear peak of the $\chi(T)$ appears at $\sim$3.5 K. This suggests that there is a ferromagnetic transition at $T_C \sim 7$ K and a spin-glass transition at $T_{sf} \sim 3.5$ K (figure 3(b)). In $CeRu_2Ge_2$, the antiferromagnetic transition

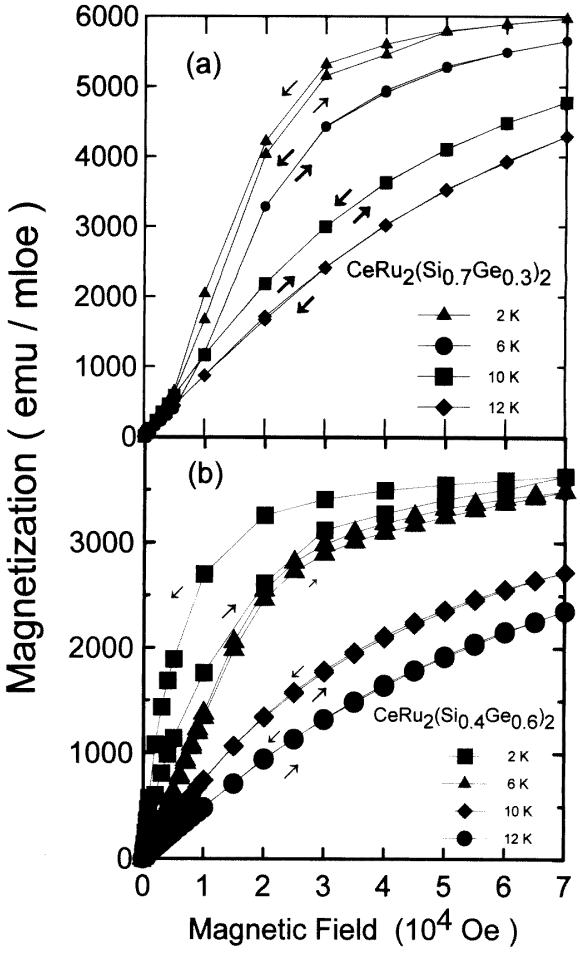


**Figure 5.** The field dependences of the magnetization $M(H)$ of $CeRu_2Si_{1.4}Ge_{0.6}$ and $CeRu_2Si_{0.8}Ge_{1.2}$ at 2, 6, 10 and 12 K.

disappears and the ferromagnetic transition appears at $T_C \sim 9$ K (figure 3(c)). However, even if 10% of the germanium of $CeRu_2Ge_2$ is substituted by silicon, this multiple magnetic phase transition still exists in $CeRu_2Si_{0.2}Ge_{1.8}$ (figure 3(d)). This seems to indicate that the ferromagnetic phase of $CeRu_2Ge_2$ is unstable. Moreover if $x$ increases, the ferromagnetic transition temperature $T_C$ increases and the antiferromagnetic transition temperature $T_N$ decreases; therefore, $T_C$ is close to $T_N$. In $CeRu_2Si_{0.2}Ge_{1.8}$, $T_C \sim 7.9$ K nears $T_N \sim 8.7$ K. Furthermore, if $0.1 < x < 0.6$, the ferromagnetic interaction is not strong enough to overwhelm the antiferromagnetic interaction; therefore, only a spin-glass state is observed. For $x \leqslant 0.1$, only a very small antiferromagnetic order is observed.

Figure 4(a) is the field dependences of the magnetization $M(H)$ of $CeRu_2Si_2$ at 2, 6, 10 and 12 K. Up to 7 T, $M(H)$ is still linear in $H$ and independent of temperature. $M(H)$ is measured as $H$ is increased from 0 to 7 T and reversed from 7 to 0 T. There is no hysteresis in $M(H)$, which further confirms that the sharp rise of $\chi_{FC}$ of $CeRu_2Si_2$ below 6 K is due to magnetic impurity instead of an intrinsic property of $CeRu_2Si_2$.

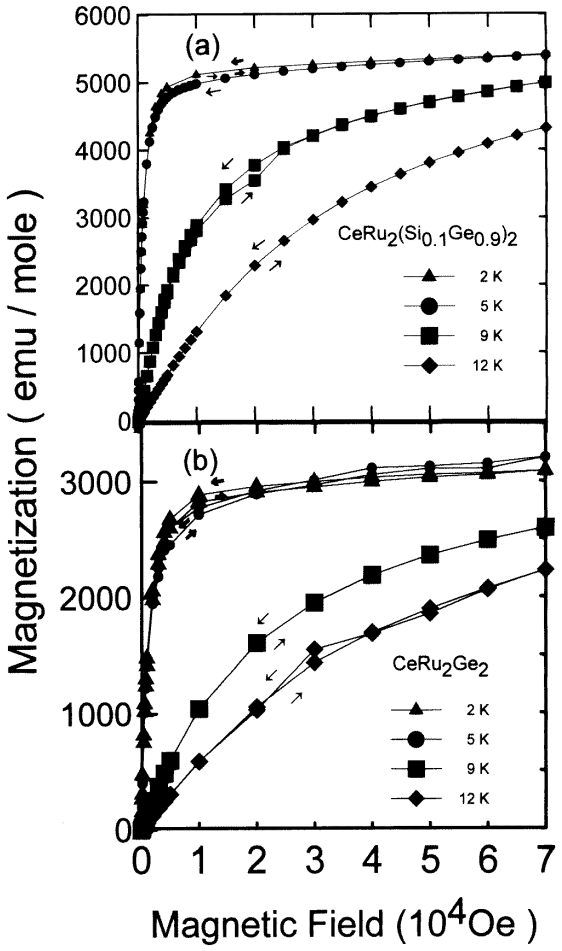


**Figure 6.** The field dependences of the magnetization $M(H)$ of $CeRu_2Si_{0.2}Ge_{1.8}$ and $CeRu_2Ge_2$ at 2, 5, 9 and 12 K.

Figure 4(b) is the field dependences of the magnetization $M(H)$ of $CeRu_2Si_{1.8}Ge_{0.2}$ at 2, 10 and 12 K. As shown in figure 4(b), there is no hysteresis. At 10 and 12 K, the magnetization $M$ is linear with the applied field $H$. In a high field ($H > 1$ T), $M(H)$ weakly deviates from a linear pattern at 2 K, which might indicate an antiferromagnetic order at low temperature. The muon experiments suggest the appearance of tiny magnetic ordered moments ($10^{-3}$ $\mu_B$) in $CeRu_2Si_2$ below $T_N \sim 1.5$ K [9]. If the germanium in $CeRu_2Si_{1.8}Ge_{0.2}$ makes the Ce moment become more stable, the non-linear behaviour of $M(H)$ will be consistent with a weak antiferromagnetic correlation below 2 K. The field dependences of the magnetization $M(H)$ of $CeRu_2Si_{1.4}Ge_{0.6}$ at 2, 6, 10 and 12 K are shown in figure 5(a). Even in a 7 T field, the magnetization $M$ is still not saturated. In high field ($H > 1$ T) below 6 K, the magnetization curves $M(H)$ start to deviate from a linear relationship. Below 2 K $CeRu_2Si_{1.4}Ge_{0.6}$ shows hysteresis. The behaviour of $M(H)$ is consistent with an antiferromagnetic order at 6 K and a spin-glass order below 2 K, which is supported by the susceptibility measurement. As shown in figure 5(b), the field dependence of magnetization of $CeRu_2Si_{0.8}Ge_{1.2}$ is similar to that of $CeRu_2Si_{1.4}Ge_{0.6}$. However, the

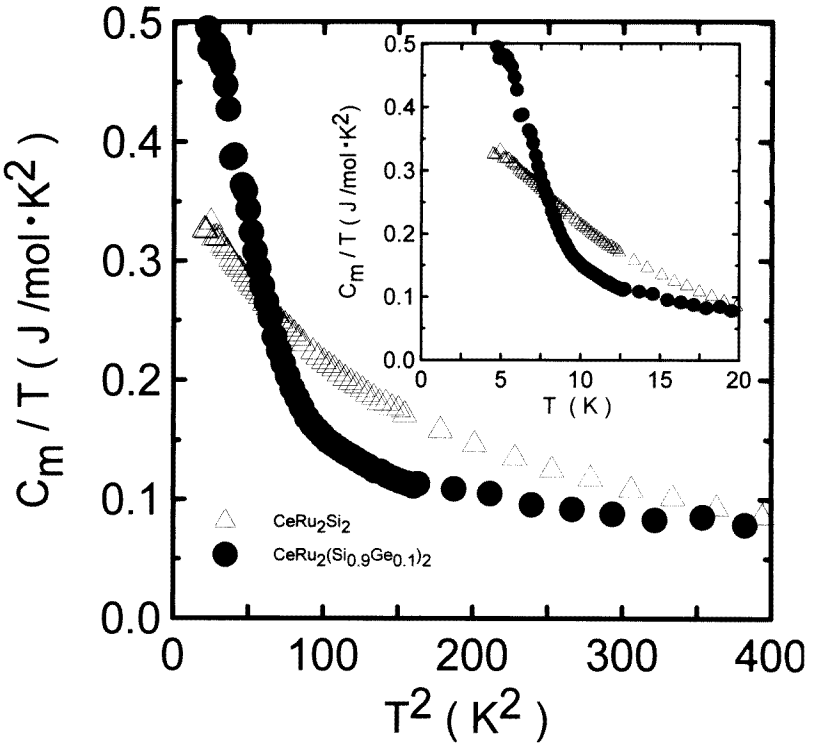


**Figure 7.** $C_m/T$ versus $T^2$ of $CeRu_2Si_2$ and $CeRu_2Si_{1.8}Ge_{0.2}$. The inset is $C_m/T$ versus $T$.

hysteresis of $CeRu_2Si_{0.8}Ge_{1.2}$ is much more pronounced than that of $CeRu_2Si_{1.4}Ge_{0.6}$ at 2 K. The hysteresis is significantly reduced in $CeRu_2Si_{0.2}Ge_{1.8}$ and $CeRu_2Ge_2$ (figure 6). This seems to suggest that the hysteresis is mainly due to the spin-glass state instead of ferromagnetic order.

The specific-heat measurements were performed in an adiabatic calorimeter by the heat-pulse method. Figure 7 shows the $C_m(T)/T$ versus $T^2$ of $CeRu_2Si_2$ and $CeRu_2Si_{1.8}Ge_{0.2}$, where the magnetic specific heat is defined as

$$C_m(T) = C(CeRu_2(Si_{1-x}Ge_x)_2) - C(LaRu_2Si_2).$$

$C_m/T$ of $CeRu_2Si_2$ at 5 K is $\sim$350 mJ mol$^{-1}$ K$^{-2}$ which agrees very well with that found in the report of van der Meulen *et al* [7]. $C_m/T$ of $CeRu_2Si_{1.8}Ge_{0.2}$ at 5 K is $\sim$500 mJ mol$^{-1}$ K$^{-2}$, which is in agreement with the report of Kim *et al* [6]. The large $C_m/T$ of $CeRu_2Si_{1.8}Ge_{0.2}$ at 5 K is due to the magnetic contribution of an antiferromagnetic order. The susceptibility measurement on $CeRu_2Si_{1.8}Ge_{0.2}$ failed to detect any occurrence of antiferromagnetic order. By neutron-scattering experiments, Mignot *et al* [10] reported that below 6 K $CeRu_2Si_{1.8}Ge_{0.2}$ was antiferromagnetic. Therefore, the specific-heat measurement further confirms the existence of antiferromagnetic order in $CeRu_2Si_{1.8}Ge_{0.2}$ below 5 K.

The temperature dependences of the specific heat $C(T)$ of $CeRu_2Si_{1.6}Ge_{0.4}$ and $CeRu_2Si_{1.4}Ge_{0.6}$ are shown in figure 8. As the temperature decreases, $C(T)$ starts to increase around 10 K with a broad maximum at $\sim$7 K and $\sim$8.5 K in $CeRu_2Si_{1.6}Ge_{0.4}$ and $CeRu_2Si_{1.4}Ge_{0.6}$ respectively, which might be corresponding to the antiferromagnetic transitions.

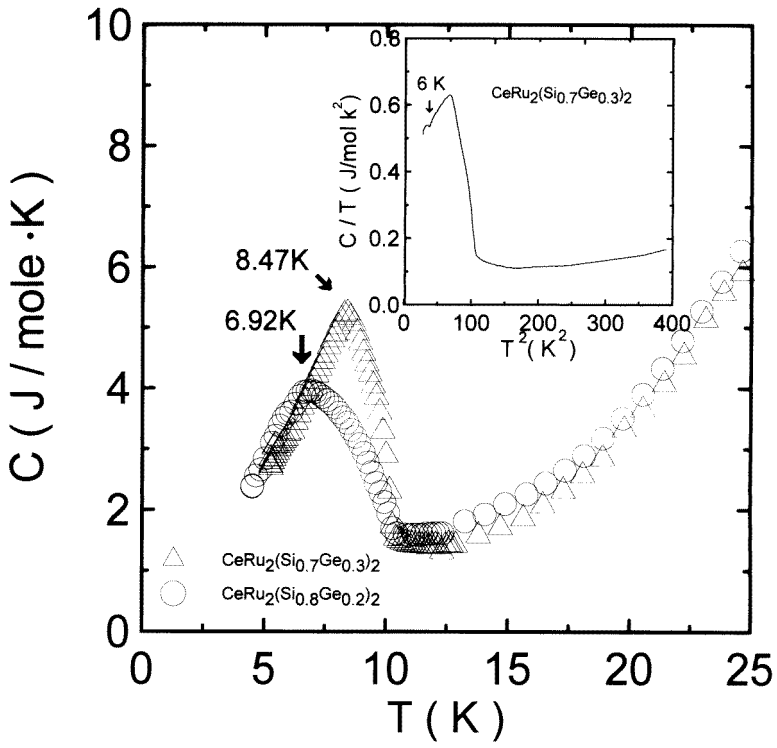


**Figure 8.** $C$ versus $T$ of $CeRu_2Si_{1.6}Ge_{0.4}$ and $CeRu_2Si_{1.4}Ge_{0.6}$. The inset is $C/T$ versus $T^2$.

As shown in the inset of figure 8, $C/T$ of $CeRu_2Si_{1.4}Ge_{0.6}$ exhibits a small hump at $\sim$6 K. To compare $\chi(T)$ with $M(H)$ of $CeRu_2Si_{1.4}Ge_{0.6}$, $\chi(T)$ begins to increase below 6 K, and $M(H)$ has a non-linear dependence below 6 K. The anomalous hump $\sim$6 K might further support a spin-glass order below 6 K.

Figure 9 is the temperature dependence of the specific heat of $CeRu_2Si_{0.4}Ge_{1.6}$ and $CeRu_2Si_{0.2}Ge_{1.8}$. As shown in figure 9, $C(T)$ of $CeRu_2Si_{0.4}Ge_{1.6}$ and $CeRu_2Si_{0.2}Ge_{1.8}$ exhibit rather complex behaviours. $C(T)$ of $CeRu_2Si_{0.4}Ge_{1.6}$ and $CeRu_2Si_{0.2}Ge_{1.8}$ begins to increase at 10 K and reaches two peaks at $\sim$8.4 and $\sim$6.5 K. Furthermore, there is a shoulder $\sim$5 K in $CeRu_2Si_{0.4}Ge_{1.6}$ and $\sim$6.2 K in $CeRu_2Si_{0.2}Ge_{1.8}$.

A double-peak behaviour of $C(T)$ was observed in $CeRu_2Ge_2$ by Besnus *et al* [3]. In the polycrystal $CeRu_2Ge_2$, they observed two peaks at 7.51 and 7.90 K and a further rate change at 8.0 K. However, for a single crystal of $CeRu_2Ge_2$, Besnus *et al* observed only one peak at 8.0 K. Therefore, the anomalous double-peak behaviour in $C(T)$ of $CeRu_2Si_{0.2}Ge_{1.8}$ might be related to two peaks of polycrystal $CeRu_2Ge_2$, which was observed in $CeRu_2Ge_2$ by Besnus *et al*. Our specific-heat measurements of polycrystal $CeRu_2Ge_2$ are shown in figure 10. As indicated in figure 10, $C(T)$ variation draws a typical second-order transition at 8 K, which is similar to the single-crystal results of Besnus *et al* [3]. Lack of double-peak behaviour in our specific-heat measurements further confirms the high quality of our samples.

Electrical resistance was measured by a four-probe method. Figure 11 gives the temperature dependences of relative resistivity $\rho(T)/\rho(285\ \mathrm{K})$ of $CeRu_2(Si_{1-x}Ge_x)_2$. The results for $CeRu_2Si_2$ and $CeRu_2Ge_2$ are consistent with previous reports [1, 3]. Except

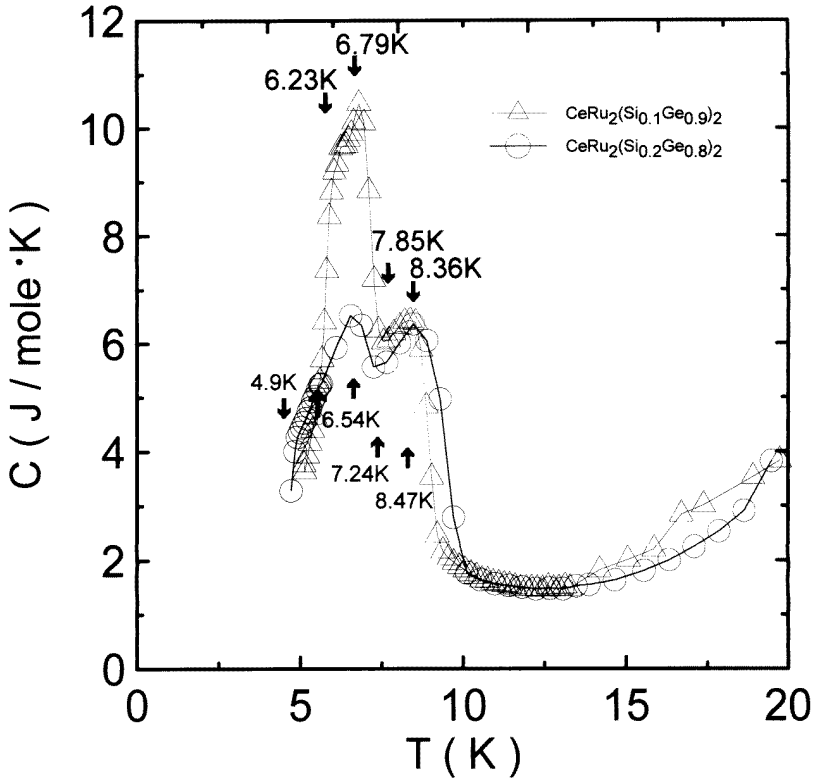


**Figure 9.** $C$ versus $T$ of $CeRu_2Si_{0.4}Ge_{1.6}$ and $CeRu_2Si_{0.2}Ge_{1.8}$.

$x = 0$, below 30 K, $\rho(T)$ increases continuously up to a temperature of 10 K. Between 30 and 10 K, the increase of $\rho(T)$ with decreasing temperature is due to a continuously increasing spin-disorder contribution of the magnetic component [3]. The drastic drop of $\rho(T)$ below 10 K is caused by magnetic ordering. The multiple phase transitions of $CeRu_2(Si_{1-x}Ge_x)_2$ are not observed in the resistance measurements. However, as shown in figure 12, the slope of $\rho(T)/\rho(285\ \mathrm{K})$ of $CeRu_2Si_{0.4}Ge_{1.6}$ changes at $\sim$7 K, which further supports multiple phase transitions in $CeRu_2(Si_{1-x}Ge_x)_2$.

## 3. Discussion

If $x \geqslant 0.2$, before the magnetic transition, the specific heat of $CeRu_2(Si_{1-x}Ge_x)_2$ is the same as that of $CeRu_2Ge_2$ and much smaller than that of $CeRu_2Si_2$. Therefore, if $x \geqslant 0.2$, the effective electron mass of $CeRu_2(Si_{1-x}Ge_x)_2$ is almost the same. Moreover if $x \geqslant 0.2$, the temperature dependence of the resistivity $\rho(T)$ of $CeRu_2(Si_{1-x}Ge_x)_2$ is all the same and obviously different from that of $CeRu_2Si_2$. The compound $CeRu_2Si_2$ is considered to be an exemplary system of the competition between the Kondo effect and the RKKY interaction. Therefore, if $x \geqslant 0.2$, the lack of Kondo effect indicates the properties of $CeRu_2(Si_{1-x}Ge_x)_2$ depend entirely on the RKKY interaction.

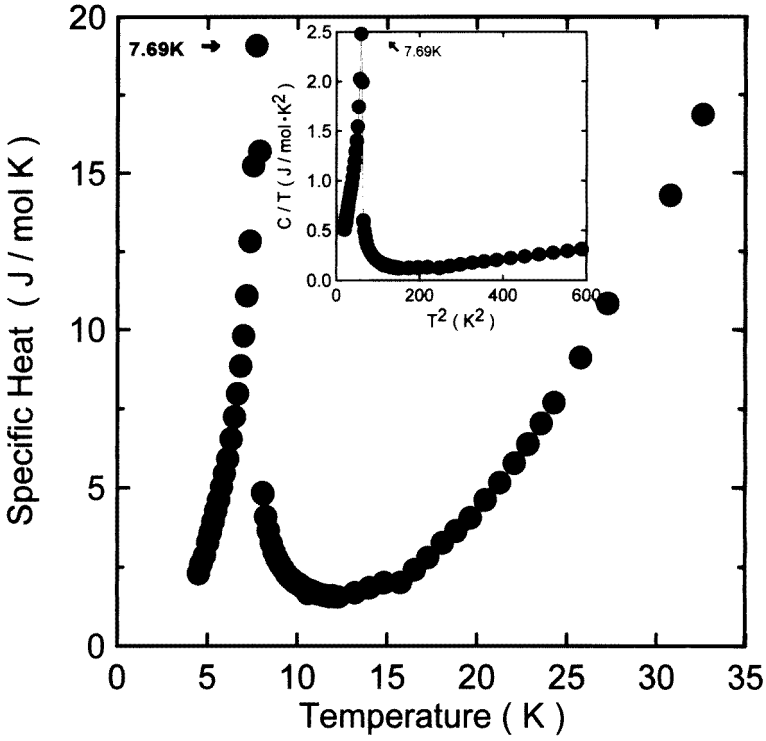


**Figure 10.** $C$ versus $T$ of $CeRu_2Ge_2$. The inset is $C/T$ versus $T^2$.

The lack of Kondo effect could be explained by an argument of Loidl *et al* [4]. By inelastic neutron scattering, Loidel *et al* claimed that for ternary compounds, such as $CeRu_2Si_2$ and $CeRu_2Ge_2$ with large volumes, the RKKY interaction between well localized f electrons dominates. In those compounds ordinary magnets are found, whereas the Kondo effect plays a minor role. In their report, only in a narrow range of unit-cell volumes ($165\ \text{Å}^3 < V < 185\ \text{Å}^3$) heavy-fermion systems can be found characterized by a delicate balance between the binding energy of the RKKY antiferromagnetic state and the Kondo singlet.

The unit-cell volume of $CeRu_2Si_{1.6}Ge_{0.4}$ ($x = 0.2$) is 174 $\text{Å}^3$ and that of $CeRu_2Si_2$ is 172 $\text{Å}^3$. It seems that the small changes of the volume greatly modify the hybridization between f electrons and conduction electrons. By band-structure calculation [11], the electronic structure of $CeRu_2Si_2$ is mainly determined by Ru instead of Si. However, the distance between Ce and Ru is $[(0.5a)^2 + (0.25c)^2]^{1/2}$; between Ce and Si, $[(0.5a)^2 + (0.18c)^2]^{1/2}$ [5]. Therefore, replacing Si with Ge will increase the distance between Ce and Ru even more than that between Ce and Si. It might also reduce the hybridization between the Ru 4d band and the Ce 4f band, such that Kondo interaction vanishes.

According to specific-heat measurements, $CeRu_2Si_{1.6}Ge_{0.4}$ is antiferromagnetic and $CeRu_2Ge_2$ is ferromagnetic at low temperature. By pressure dependence of magnetic coupling, Thompson *et al* [12] claimed that the magnetism in $CeRu_2Ge_2$ strongly depends on the Ce–Ce separation. For small separation the coupling is antiferromagnetic, and for large Ce–Ce separation the coupling is ferromagnetic. Substitutions on the Ge site by smaller Si correspond to a pressure effect. Therefore, the evolution of $CeRu_2(Si_{1-x}Ge_x)_2$

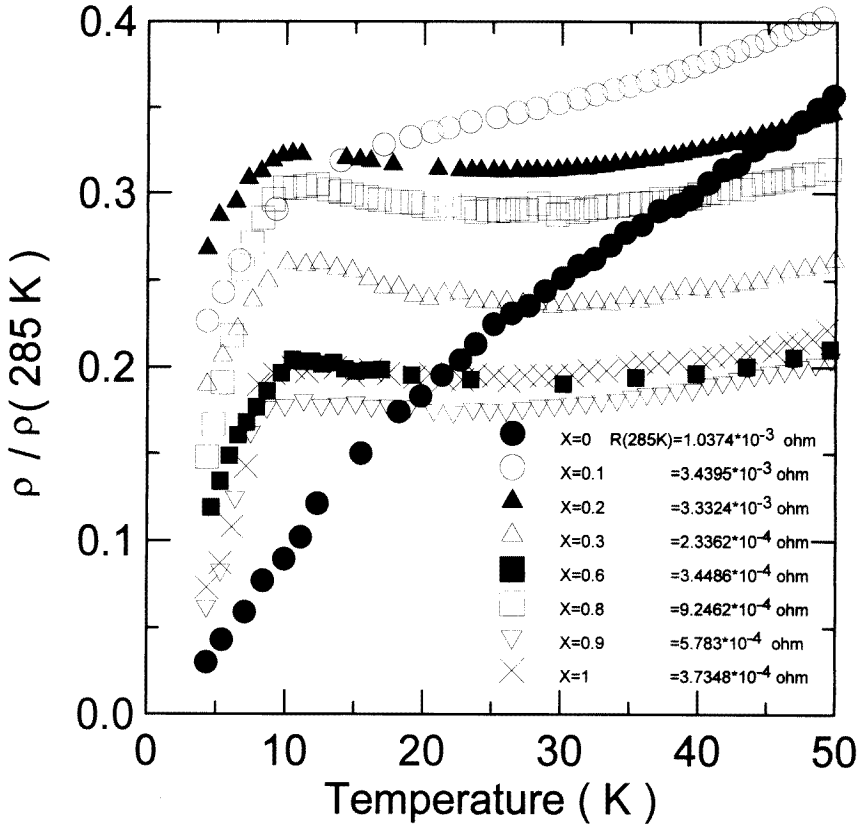


**Figure 11.** The temperature dependence of the relative resistivities of $CeRu_2(Si_{1-x}Ge_x)_2$.

from antiferromagnetism to ferromagnetism can be explained by the spatially oscillatory nature of the RKKY interaction which depends on the Ce–Ce separation. However, this simple pressure effect can not explain the multiple phase transition [11].

Two (or three) transitions of polycrystal $CeRu_2Ge_2$ reported by Besnus *et al* [3] were carefully examined by many other research groups. In an annealed polycrystal $CeRu_2Ge_2$, Thompson *et al* [12] also observed a sharp peak at 7.5 K with a weak anomaly near 8.3 K. If $T_1$ is the temperature of the high-temperature peak and $T_2$ is the temperature of the low-temperature peak, Thompson *et al* measured the pressure dependences of $T_1$ and $T_2$. They reported that $T_1$ increases and $T_2$ decreases with pressure $P$, $\partial T_1/\partial P = 0.13$ K kbar$^{-1}$ and $\partial T_2/\partial P = -0.23$ K kbar$^{-1}$. They associated $T_1$ with antiferromagnetism and $T_2$ with ferromagnetism.

Double phase transitions are also observed in $CeRh_2Ge_2$. From results of specific heat, one appears at $T_1 = 14$ K and another at $T_2 = 8.3$ K. Since neutron diffraction [13] indicates incommensurate antiferromagnetic order below 15 K, the transition at $T_1$ is suggested to be incommensurate antiferromagnetism; at $T_2$, an incommensurate to a commensurate transition. Both $T_1$ and $T_2$ increase with pressure $P$, $\partial T_1/\partial P = 0.16$ K kbar$^{-1}$ and $\partial T_2/\partial P = 0.23$ K kbar$^{-1}$.

The most dramatic behaviour of $CeRu_2(Si_{1-x}Ge_x)_2$ is that the antiferromagnetic transition temperatures $T_1$ are almost independent of $x$. In our specific-heat measurements

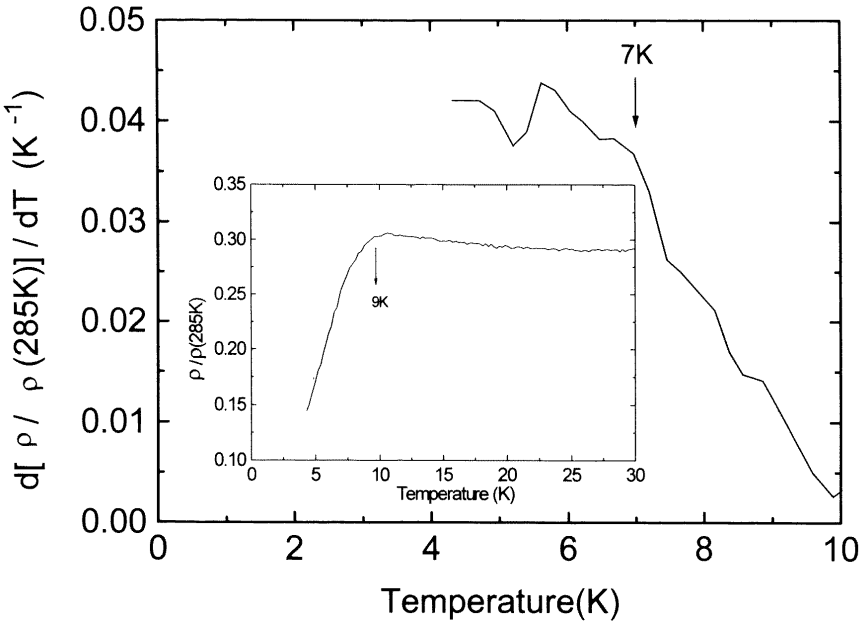


**Figure 12.** The temperature dependence of $\mathrm{d}(\rho(T)/\rho(285\ \mathrm{K}))/\mathrm{d}T$ of $CeRu_2Si_{0.4}Ge_{1.6}$. The inset is $\rho(T)/\rho(285\ \mathrm{K})$ between 4.2 and 30 K.

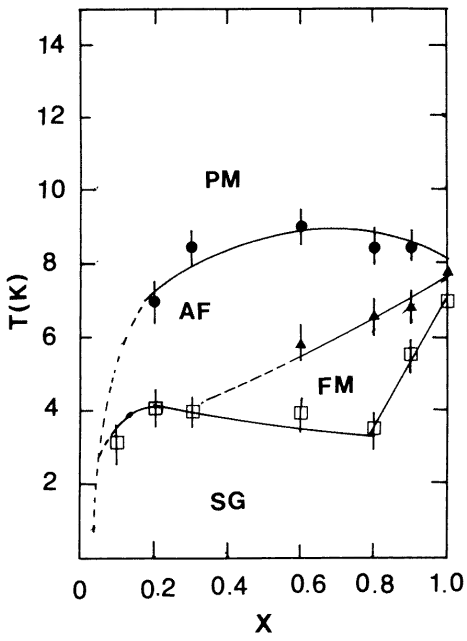


**Figure 13.** Phase diagram of $CeRu_2(Si_{1-x}Ge_x)_2$. The solid circles indicate the $T_N$ which are observed by specific-heat measurements. The triangles represent ferromagnetic transitions. The open squares denote the $T_{sf}$ which are observed by susceptibility measurements. (PM: paramagnetism, AF: antiferromagnetism, FM: ferromagnetism, SG: spin glass.)

of $CeRu_2(Si_{1-x}Ge_x)_2$, if $0.3 \leqslant x \leqslant 0.9$, there is always a peak at 8.5 K. Therefore, either $T_1$ is independent of pressure or all the chemical pressure in $CeRu_2(Si_{1-x}Ge_x)_2$ is too low to show a significant pressure dependence.

Apart from peaks at $T_1 = 8.5$ K and $T_2 = 6.5$ K, $C(T)$ of $CeRu_2Si_{0.4}Ge_{1.6}$ exhibits a shoulder around $T_3 = 5$ K, and $C(T)$ of $CeRu_2Si_{0.2}Ge_{1.8}$ has a shoulder around $T_3 = 6.2$ K. By the competition between the ferromagnetic and antiferromagnetic interaction, $CeRu_2(Si_{1-x}Ge_x)_2$ might form a spin-glass state below $T_3$. Based on the following arguments: (1) neutron diffraction [6] indicates incommensurate modulations of Ce in $CeRu_2Si_{1.8}Ge_{0.2}$ below 7 K, (2) below 2 K $CeRu_2Si_{1.4}Ge_{0.6}$ shows hysteresis, (3) in $CeRu_2Si_{0.4}Ge_{1.6}$ and $CeRu_2Si_{0.2}Ge_{1.8}$, $\chi_{FC}$ deviates from $\chi_{ZFC}$ at $\sim$5 K and 6 K respectively; we might conclude that the 8.5 K transition is an antiferromagnetic transition, the 6.5 K transition an antiferromagnetic–ferromagnetic transition and at the shoulder (around 5 K in $CeRu_2Si_{0.4}Ge_{1.6}$ and 6.2 K in $CeRu_2Si_{0.2}Ge_{1.8}$) there is a ferromagnetic to spin-glass transition. Although there are three phase transitions in $C(T)$ of $CeRu_2Si_{0.4}Ge_{1.6}$ and $CeRu_2Si_{0.2}Ge_{1.8}$, $\chi(T)$ indicates two transitions only. One of the three transitions might be insensitive to susceptibility measurements. (One should also notice that the transition temperature measured by magnetic measurements or by thermal measurements might not be exactly the same.)

The best identification of an antiferromagnetic transition temperature $T_N$ is determined by the maximum in $\mathrm{d}(\chi T)/\mathrm{d}T$ [14, 15]. Besides, magnetic transition temperatures can be also determined by the peaks in $C(T)$. However, in our measurements, $T_N$ decided either by $\mathrm{d}(\chi T)/\mathrm{d}T$ or by $C(T)$ is all the same. For $x < 0.1$, there is no clear long-range magnetic order above 2 K, and $\chi_{FC}(T)$ starts to deviate from $\chi_{ZFC}(T) < 3$ K. If the spin-glass transition temperatures $T_{sf}$ are determined when $\chi_{FC}(T)$ starts to deviate from $\chi_{ZFC}(T)$, the phase diagram is as shown in figure 13.

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