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Absence of a first-order metamagnetic transition in $CeRu_2Si_2$

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Metamagnetic transition of the heavy-electron compound $CeRu_2Si_2$ has been examined on high-purity single crystals by means of a static magnetization measurement at very low temperatures down to 90 mK. No hysteresis is observed in the magnetization process. The peak value of the differential susceptibility at the metamagnetic field $B_M \sim 7.7$ T follows a simple T^2 law below $T \sim 0.5$ K, and saturates to a finite value $1.88 \mu_B$/T Ce as $T \rightarrow 0$. From these results for two single crystals with slightly different quality, we have confirmed the nonexistence of a first-order phase transition at B_M in the clean limit of this system. The itinerant nature of the $4f$ electrons is most likely preserved in the magnetized state at $B \geqslant B_M$.

The tetragonal $ThCr_2Si_2$-type compound $CeRu_2Si_2$ is known to be an nonmagnetic heavy-electron system having an electronic specific-heat coefficient $\gamma \sim 350$ mJ/mol K^2.[1] One of the unique properties of this compound is that its magnetization exhibits an abrupt nonlinear increase at the field of $B_M \sim 7.7$ T applied along the c axis, or the [001] direction.[2–6] Well above this field, the Ce ions acquire a relatively large magnetic moment of $\sim 1.5 \mu_B$/Ce. This phenomenon is often referred to as a "metamagnetic transition," though no sign of a real phase transition is observed so far.

The metamagnetic behavior of $CeRu_2Si_2$ is known to be strongly temperature dependent and becomes discernible only below ~ 15 K. It is also very sensitive to a sample quality. Whether or not the metamagnetic behavior evolves to a real phase transition as $T \rightarrow 0$ in a clean limit is of current interest,[4–7] not only in elucidating a possible mechanism of the phenomenon, but also in interpreting the recent de Haas–van Alphen (dHvA) experiments;[8–10] a rather sudden change of the extremal areas of the Fermi surface is indicated at B_M, as if the nature of the $4f$ electrons is changed from itinerant to localized above this field.[9] According to the neutron-diffraction experiments,[11] short-range antiferromagnetic correlations existing at low fields disappear at B_M and the system remains in the paramagnetic state at higher fields. There is no change in the magnetic symmetry in fields. The possible phase transition that would be compatible with the dHvA analysis, if any, should then be of *first* order, analogous to a liquid-gas transition. In such a case the phase transition would be observed below a critical temperature T_{cr}. In order to explore this point, we have performed the *static* magnetization measurements on high-quality single crystals of $CeRu_2Si_2$ at very low temperatures.

We prepared two single crystals No. 1 and No. 2 by the Czochralski pulling method, starting from Ce (99.99%), Si (>99.999%) and 99.9% (No. 1) or 99.99% (No. 2) pure Ru. The single-crystal rod No. 2 was purified using a solid-state electro-transport method. The rod was heated up by a dc current flow with the density of 1300 A/cm^2 in average through the rod, and kept at a temperature from 1200 °C to 1300 °C for 150 h in high vacuum below 2×10^{-9} Torr. The obtained crystals had the resistivity ratio $\rho(300\ K)/\rho(T \rightarrow 0)$ of ~ 100 (No. 1) and ~ 400 (No. 2). Sample No. 2 showed even better quality than those used in the dHvA experiments.[8,9] These crystals were cut into the parallelepiped of $2 \times 2 \times 3$ mm^3, with the longest axis oriented along the c axis. High-resolution magnetization measurements down to 90 mK were performed by a Faraday force method.[7,12] Most measurements were done with an optimum field gradient value of $dB/dz = 3$ T/m.

In Fig. 1 we show the magnetization curves for sample No. 2 obtained by field scans at various temperatures below 4.2 K. At each temperature the magnetization data near the

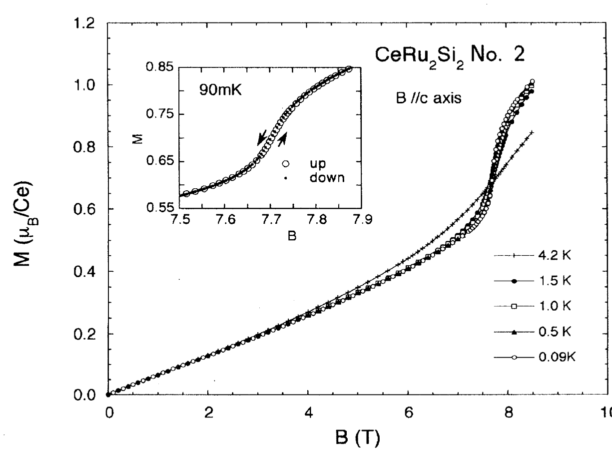

FIG. 1. Magnetization of $CeRu_2Si_2$ (No. 2) in fields $B \| c$ axis measured at various temperatures. The inset is the results for the field-increasing and -decreasing sweeps near the metamagnetic transition at 90 mK, showing no hysteresis in the magnetization process.

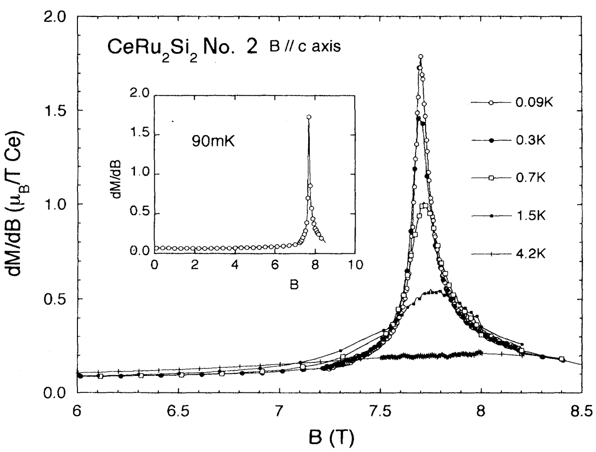

FIG. 2. Differential susceptibility of $CeRu_2Si_2$ (No. 2) near the metamagnetic transition in fields $B\|c$ axis. The inset shows the differential susceptibility at 90 mK in the full range of field.

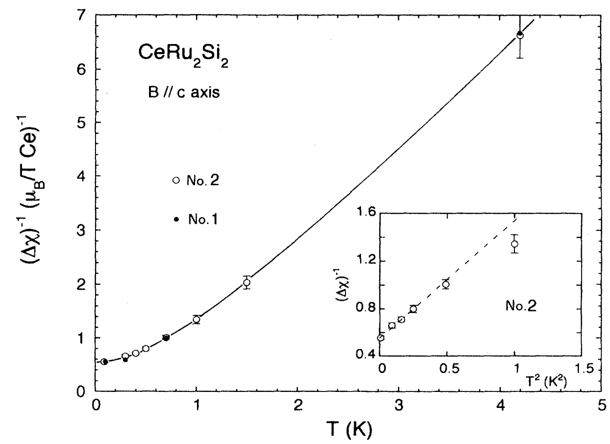

FIG. 3. Reciprocal of the peak height of the differential susceptibility of $CeRu_2Si_2$ as a function of temperature. Open circles are the data points for sample No. 2 while dots are those for No. 1. The solid line is a guide for the eyes. The inset shows the data points for No. 2 as a function of T^2.

metamagnetic region were collected every 5 mT, not all of which are shown in the figure. A strong sharpening of the metamagnetic behavior near $B\sim8$ T is evident at low temperatures. The inset of Fig. 1 shows the magnetization process around the metamagnetic field at $T=90$ mK; the data points for both field-increasing and -decreasing scans are shown. Neither discontinuity nor any appreciable hysteresis can be seen in the data, excluding a first-order transition for $T\geqslant90$ mK.

In order to judge where T_{cr} exists at still lower temperature or not, we examine the temperature variation of the metamagnetic behavior in more detail. Figure 2 shows the differential susceptibility $\chi_B\equiv dM/dB$ for sample No. 2 at various temperatures, obtained from the slopes of two successive data points on the magnetization curves. It is natural to define the transition field B_M by the peak position which corresponds to an inflection point of the magnetization curve. On cooling below 4.2 K, there is a small shift of B_M to the lower-field side, in agreement with the previous magnetization measurement above 1.3 K.[2] The $T=0$ extrapolated value $B_M(0)$ in the present measurement is 7.70 T, in good agreement with previously reported values.[3] The width ΔB of the transition estimated at 75% of the peak value χ_{B_M} is strongly temperature dependent and reduces to $\Delta B\sim40$ mT at 90 mK. It should be noticed that ΔB is still much larger than the field inhomogeneity of $\sim$9 mT within the sample due to the field gradient (3 T/m) of the Faraday method. In fact, we confirmed that further reducing dB/dz to 1.5 T/m does not cause any appreciable change in ΔB at 90 mK. Moreover, we observed the same magnitude of ΔB of sample No. 1 as well. From these, we may conclude that ΔB observed is an intrinsic width of the metamagnetic behavior of this compound.

To show how the sharpness of the transition evolves on cooling, it is convenient to define a peak height of the differential susceptibility: $\Delta\chi=\chi_{B_M}-\chi_0$, where the initial susceptibility χ_0 has an almost temperature-independent value of $0.06\mu_B$/T Ce below 4.2 K. Figure 3 summarizes the temperature variation of the reciprocal of the peak height $(\Delta\chi)^{-1}$ for the two samples No. 1 (dots) and No. 2 (open circles). In these plots we have done a demagnetization field correction, which was at most less than 10% of the $\Delta\chi$ value even for the sharpest transition at 90 mK. Above 1 K, $(\Delta\chi)^{-1}$ is strongly temperature dependent and varies almost proportional to T, in agreement with the previous results.[2] Below 1 K, however, $(\Delta\chi)^{-1}$ levels off and approaches a finite value. Very recently, a similar saturating tendency of $\Delta\chi$ is also observed by Holtmeier *et al.* independently.[6] It is very important to point out that we obtained the same results for two samples (Nos. 1 and 2) of slightly different quality. This fact implies that our samples are already in the clean limit and the observed saturation of $\Delta\chi$ is intrinsic to the system. The inset of Fig. 3 shows a T^2 plot of $(\Delta\chi)^{-1}$ for sample No. 2. Below $T^*\sim0.5$ K, $(\Delta\chi)^{-1}$ seems to follow a T^2 law, from which we obtain the $T=0$ extrapolated value of $\Delta\chi(T)$ as $1.82\mu_B$/T Ce, or equivalently $\chi_{B_M}=1.88\mu_B$/T Ce. These results strongly exclude the possibility of $T_{cr}\geqslant0$, since $\Delta\chi$ should diverge at T_{cr}. The T^2 variation of $(\Delta\chi)^{-1}$ below T^* rather suggests that the system remains in a Fermi-liquid state even at $B\sim B_M$. A similar crossover to the Fermi-liquid state is also suggested in the recent thermal expansion experiments.[5] From the results in Figs. 1–3, we may conclude that the magnetization process of $CeRu_2Si_2$ is a continuous function of field as $T\to0$ and *no* first-order metamagnetic transition is expected.

The electronic specific heat C/T of $CeRu_2Si_2$ is known to grow in field $B\|c$ and takes a pronounced maximum near B_M, followed by a gradual decrease at higher fields.[3,13,14] Whether C/T is singular at B_M or not is of importance. We have reexamined this point by a thermodynamical analysis of our magnetization results. A similar analysis was given by Paulsen *et al.*,[3] who measured $M(T)$ of $CeRu_2Si_2$ down to $\sim$200 mK at fixed fields using a superconducting quantum interference device magnetometer. Their results indicate a

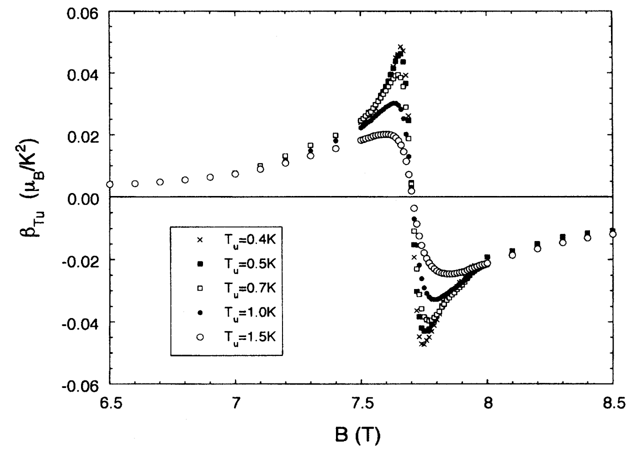

FIG. 4. Approximated coefficient of the T^2 term of the magnetization $\beta_{T_u}(B)$ as a function of field. Here $\beta_{T_u}(B)$ is evaluated in the temperature interval between 0.09 K and T_u, T_u=0.4–1.5 K.

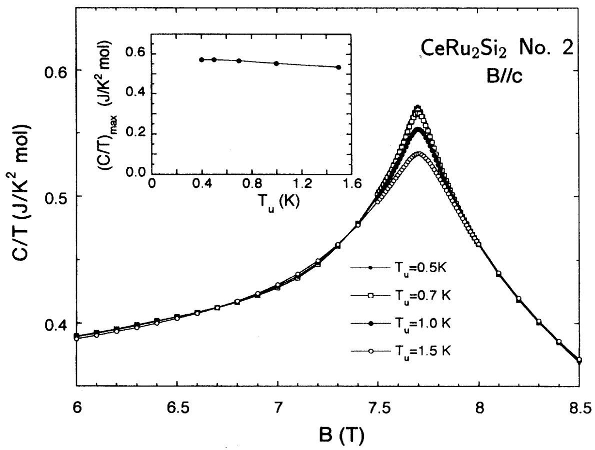

FIG. 5. Field variation of the C/T value of $CeRu_2Si_2$ (No. 2), obtained by integrating the curves in Fig. 4. The inset shows the peak value of C/T at B=7.7 T as a function of T_u for 0.4 K $\leqslant T_u \leqslant$1.5 K. Solid lines are guides for the eyes.

T^2 variation of the magnetic moment at low temperatures below ~0.5 K. From the thermodynamic Maxwell relation, the field variation of C/T can then be calculated as $\Delta(C/T) = 2\int\beta(B)dB$,[3] where $\beta(B)$ is a coefficient of the T^2 term of the magnetization. It is shown that $\beta(B)$ is positive (negative) for $B<B_M$ ($B>B_M$), changing its sign at $B\sim B_M$.[3] Of interest is its strong field variation near B_M. The amplitude of $|\beta(B)|$ becomes maximum at fields very close to B_M, resulting in a sharp peak in the C/T value at $B\sim B_M$.[3] In the following, we examine the behavior of $\beta(B)$ near B_M from our field-scan measurements with improved field resolution.

Assuming the T^2 law, the low-temperature magnetization can be expressed as $M(B,T)=M(B,0)+\beta(B)T^2$, where $M(B,0)$ is shown in the present experiment to be a *continuous* function of field. Defining the magnetization difference $\Delta M(B,T)=M(B,T)-M(B,T=0.09$ K), we may approximate the coefficient $\beta(B)$ in the temperature interval between 0.09 K and T_u as $\beta_{T_u}(B)=\Delta M(B,T_u)/[T_u^2-(0.09)^2]$. Some of the results are plotted in Fig. 4 as a function of B for T_u=0.4–1.5 K. When the field is not very close to B_M ($B\leqslant7$ T and $B\geqslant8$ T), the value of $\beta_{T_u}(B)$ is nearly independent of T_u as is expected. Near B_M, however, the amplitude of $|\beta_{T_u}(B)|$ begins to increase for lower T_u. This implies that the T^2 law is valid only at very low temperatures in this field region.[3] The curve for T_u=0.5 K in Fig. 4 seems to show a sharper structure than the corresponding plot in Ref. 3. Nevertheless, we consider that there is no singularity in $\beta(B)$ near B_M. It should be noticed that each curve in Fig. 4 sharply changes its sign at a field B^*=7.703 T, which field coincides with $B_M(0)$. Surprisingly enough, the value of B^* does not show any appreciable change for $0.5\leqslant T\leqslant1.5$ K. This means that all the magnetization curves for $T\leqslant1.5$ K intersect each other at the same field $B=B^*$, although B_M increases by ~0.1 T on warming up to 1.5 K. From the T independence of B^* and the T^2 saturation of $\Delta\chi$, we can show $M(B,T)-M(B,0)\propto\Delta\chi(0)T^2(B^*-B)$ near B^* below ~0.5 K, which assures a nonsingular variation of $\beta(B)$ around B_M.

Figure 5 shows the C/T vs B plot, obtained by integrating the curves of Fig. 4 with an initial value of C/T=350 mJ/mol K^2 at $B=0$. A pronounced peak of C/T occurs just at $B=B^*$ $[=B_M(0)]$. The inset of Fig. 5 shows $(C/T)_{\max}$, the peak value of C/T, as a function of T_u. The saturation tendency of $(C/T)_{\max}$ for $T_u<0.7$ K is closely related to the existence of a crossover temperature $T^*\sim0.5$ K of the differential susceptibility at $B=B_M$, leading to a finite electronic specific coefficient at this field. This result strongly suggests that the system has a Fermi-liquid ground state even just at $B=B_M$. Noting that the Kondo temperature T_K of $CeRu_2Si_2$ is considered to be of the order of ~20 K,[1,2] the existence of the very low temperature scale T^* is surprising. It should be compared with $T^*\sim10$ K at $B=0$.[5,6] Some kinds of intersite fluctuations might grow as $B\rightarrow B_M$, which should also be responsible for the strong mass enhancement at this field.

Our low-temperature magnetization measurements on high-quality single crystals of $CeRu_2Si_2$ have confirmed that there is no first-order phase transition at B_M. Instead, the system seems to possess a *finite* crossover temperature $T^*\sim0.5$ K to the Fermi-liquid ground state at $B\sim B_M$. As the consequence, the low-field $4f$-itinerant state is continuously connected to the high-field magnetized one. We may then expect that the $4f$ electrons are still contributing to the Fermi surface volume at $B\geqslant B_M$.[15] These results are in contrast with those of the dHvA experiments[9] which indicate that the metamagnetic transition is a change of the f-electron nature from itinerant to localized.

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