Fermi-liquid instability of $CeRh_2Si_2$ near a pressure-induced quantum phase transition

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The electrical resistivity of single crystalline $CeRh_2Si_2$ has been measured at high pressure up to 8 GPa, and low temperature down to 2.0 K for the current along the *a* and *c* axes. Two magnetic phase transitions $T_{N1}=35$ K and $T_{N2}=24$ K at ambient pressure were observed as a function of pressure. It is found that both transitions T_{N1} and T_{N2} are suppressed by applying pressure and disappear near the critical pressure at $P_{C1}\sim 1.0$ GPa and $P_{C2}\sim 0.6$ GPa, respectively. The transition for T_{N1} is second order and the one for T_{N2} is first order. The resistivity shows T^2 dependence at low temperature in a wide pressure range from ambient pressure up to 8 GPa, indicating that a Fermi liquid state still exists near P_{C1} and P_{C2}. According to the comparison between the coefficient of T^2 term and the Sommerfeld coefficient γ, however, it is suggested that the enhancement of many-body dynamical effect occurs near the critical pressure $P_{C1}\sim 1.0$ GPa. These results are discussed on the basis of the pressure-induced quantum phase transition.

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I. INTRODUCTION

A quantum phase transition (QPT) occurs at 0 K in a highly correlated electron system due to variation of nonthermal control parameters which gives rise to fundamental change in the ground state. The typical parameters to tune QPT are the chemical composition, pressure, or magnetic field. While phase transitions in classical models are driven only by thermal fluctuations as classical systems usually freeze into a fluctuationless ground state at $T=0$, quantum systems have fluctuations driven by the Heisenberg uncertainty principle even in the ground state. The $T>0$ region in the vicinity of a quantum critical point (QCP), however, offers a fascinating interplay of effects driven by quantum and thermal fluctuations. This means that working outward from QCP is a powerful way of understanding and describing the thermodynamic and dynamic properties of numerous systems in which QPT occurs. Indeed, unusual electronic and magnetic behaviors can arise near nonzero temperature.[1,2] If the magnetic ordering temperature in a heavy fermion (HF) system is suppressed to absolute zero by tuning the control parameter, magnetic fluctuations lead to strong enhancement of the quasiparticle scattering rate and potentially to a breakdown of the Fermi liquid (FL) description.

CeT_2Si_2 (T: transition metal element) crystallizes in a $ThCr_2Si_2$-type tetragonal structure. These compounds have been well known to show many interesting electronic properties such as magnetic ordering, superconductivity, and so forth.[3] These anomalous properties are considered to be due to electronic and magnetic instability of $4f$ electron state since several kinds of interactions such as the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, the Kondo effect, and crystalline electric field are competing with each other in these compounds. Recently, a superconducting transition has been found in several compounds under hydrostatic pressure near a QCP just at the border of magnetism where $T_N\rightarrow 0$, notably in $CeCu_2Ge_2$,[4] $CePd_2Si_2$,[5] and $CeRh_2Si_2$.[6] All these systems show antiferromagnetic ordering at ambient pressure.

$CeRh_2Si_2$ is also an antiferromagnet at ambient pressure. It has been revealed by neutron diffraction that the Bragg reflection represents the magnetic modulation with the wave vector of $\mathbf{q}_1=(0.5,0.5,0)$ below $T_{N1}=36$ K, where the magnetic moments are arranged along the *c* axis. Furthermore, the second Bragg reflection due to the wave vector of $\mathbf{q}_2=(0.5,0.5,0.5)$ starts growing at $T_{N2}=24$ K.[7]

It has been reported that the resistivity and the thermal expansion show anomalies near T_{N1} and T_{N2}.[8,9] T_{N1} and T_{N2} are suppressed by applying pressure and disappear near the critical pressure at $P_{C1}\sim 1.0$ GPa and $P_{C2}\sim 0.6$ GPa, respectively. It implies that there are two phase transitions at zero temperature, that is, QPT, one of which, P_{C1}, separates magnetic ordering phase with the wave vector of $\mathbf{q}_1$ from the one with no long range order, and the other, P_{C2}, separates two magnetic phases. The superconductivity appears at 400 mK at 0.9 GPa, near the critical pressure P_{C1} required to completely suppress antiferromagnetic ordering.[6]

In the present work, we report the electrical resistivity and x-ray diffraction under pressure in detail and discuss the electronic properties of a single crystalline $CeRh_2Si_2$ near the pressure-induced QCP. The electrical resistivity of a FL exhibits a contribution $\rho(T)=\rho(0)+AT^2$ at low temperature, where A is usually attributed to the Umklapp process of the electron-electron collisions. However, in the vicinity of the magnetic-nonmagnetic transition, non-fermi-liquid (NFL) behaviors are often observed as a strong deviation of transport properties from FL predictions, which is indicated as a power law such as $\rho(T)=\rho(0)+AT^m$, $m<2$. Even if

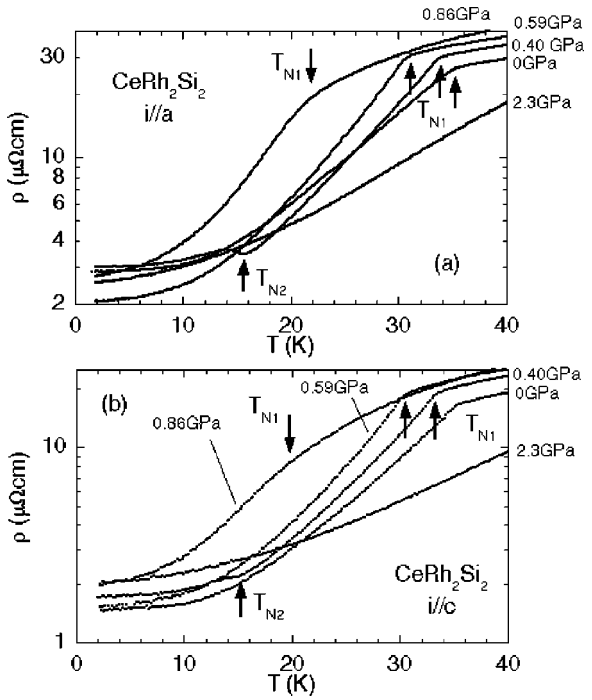

FIG. 1. Temperature dependence of the electrical resistivity of $CeRh_2Si_2$ along the (a) *a* and (b) *c* axes.

such behaviors are not observed, some crossover from NFL to FL may be found near QCP. In this paper, we discuss the electronic properties and the FL instability near QPT induced by pressure.

II. EXPERIMENTAL

Single crystal of $CeRh_2Si_2$ was grown by Czochralski pulling method in a tetra-arc furnace. The sample was annealed in a quartz tube under vacuum of 1×10^{-6} Torr at 900 °C for one week. The electrical resistance for the current along both *a* and *c* axes was measured by the usual four-probe dc method, in which four gold leads were attached to the sample by means of silver paste. The residual resistivity ratio is 32 at ambient pressure. High pressure was generated up to 2.3 GPa by using a tungsten carbide piston and a Ni-Cr-Mo-Co alloy (MP35N) cylinder.[10] The pressure was always kept constant in the temperature range between 2 and 300 K by controlling the load within $\pm1\%$. A mixture of Fluorinerts of FC70 and FC77 in ratio 1:1 was used as a pressure transmitting medium. Above 2.3 GPa, the electrical resistance was measured for the current along the *a* axis by using a cubic anvil-type high-pressure cell up to 8 GPa.[11]

The pressure dependence of lattice parameters was determined by x-ray ($MoK\alpha$) powder diffraction with a Guinier-type focusing camera and highly sensitive film. Hydrostatic pressure was generated by using tungsten-carbide Bridgman anvils and a Be sheet as a gasket. Details of the pressure apparatus have been reported previously.[12]

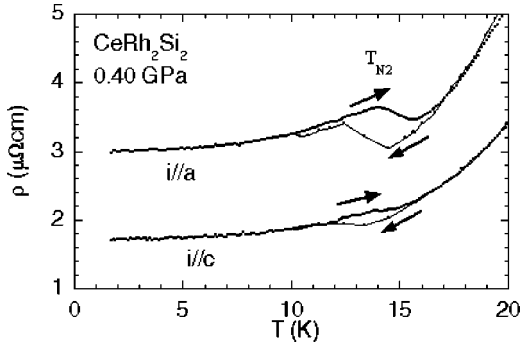

FIG. 2. The electrical resistivity at 0.4 GPa below 20 K.

III. RESULTS AND DISCUSSION

A. Disappearance of antiferromagnetism at high pressure below 2 GPa

Figure 1 shows the electrical resistivity $\rho(T)$ for the *a* and *c* axes as a function of temperature below 40 K under high pressures. At ambient pressure, the resistivity shows a sudden decrease near 35 K ($=T_{N1}$) and a small anomaly near 24 K ($=T_{N2}$). The anomalies at T_{N1} and T_{N2} correspond to the magnetic phase transitions which were mentioned before. The anomaly near T_{N1} becomes less prominent with increasing pressure, corresponding to the pressure-induced decrease of the sublattice magnetization.[7] T_{N1} decreases with increasing pressure and disappears above 1.0 GPa ($\sim P_{C1}$).

T_{N2} also decreases with increasing pressure and disappears above 0.58 GPa ($\sim P_{C2}$). At 0.4 GPa, on the other hand, it is found that $\rho(T)$ shows a minimum $T_{N2}\sim14.5$ K along the *a* and *c* axes. Furthermore, as seen in Fig. 2, the hysteresis is found near T_{N2} along the *a* and *c* axes, indicating that this transition is first order. Although no hysteresis in the $\rho(T)$ curve is observed at ambient pressure, the results of strain measurements on a single crystal of $CeRh_2Si_2$ show a discontinuity in the length of the *a* and *c* axes[13] which is consistent with a first-order transition near 26 K. The discontinuous change at T_{N2} increases with increasing pressure,[14] corresponding to the enhancement of the anomaly on $\rho(T)$ curve near T_{N2} by applying pressure.

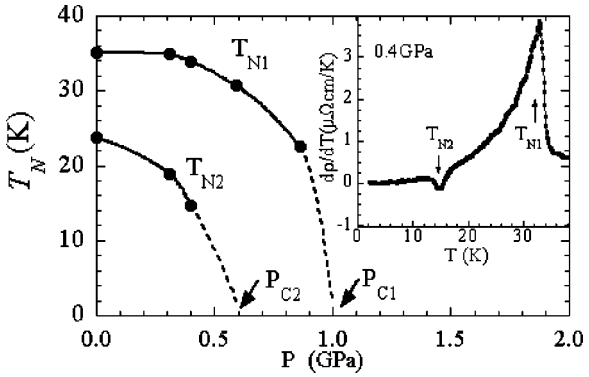

FIG. 3. Pressure dependence of the critical temperatures T_{N1} and T_{N2} of $CeRh_2Si_2$. Broken lines are extrapolations. An example of the $d\rho/dT$ at 0.4 GPa is shown in the inset.

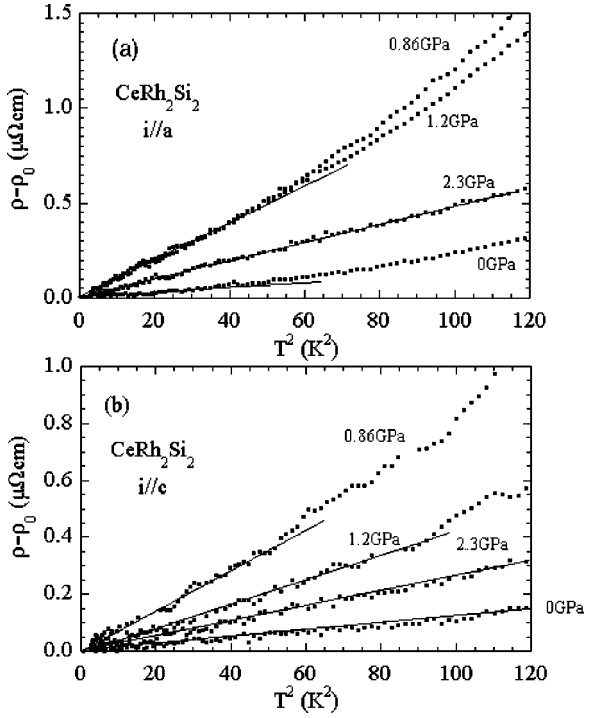

FIG. 4. T^2 dependence of $\Delta\rho=\rho-\rho_0$ of $CeRh_2Si_2$ along (a) a and (b) c axes.

The pressure dependence of T_{N1} and T_{N2} is plotted in Fig. 3. The magnetic phase transition temperatures T_{N1} and T_{N2} were determined by calculating the temperature derivatives of $\rho(T)$, $d\rho/dT$, which is shown in the inset of Fig. 3. It is in good agreement with that determined by the measurement of thermal expansion.[8] We have found that the transition at T_{N1} is second order and that at T_{N2}, first order. T_{N1} decreases gradually at low pressure and then abruptly near 1 GPa, which is the critical pressure P_{C1} where the transition disappears. Qualitatively, the phase boundary of $T_{N1}(P)$ is interpreted on the basis of Doniach's model for competing Kondo and RKKY interactions.[15]

In $CeRh_2Si_2$, the magnetic order is present because the RKKY interaction overcomes the Kondo effect at low temperature. By considering that T_{N1} decreases with pressure, $CeRh_2Si_2$ is on the right hand side of Doniach's phase diagram. The antiferromagnetic order is destroyed by increasing the coupling constant $JN(0)$, which means an increase of pressure. The ground state then becomes a FL with a rather large effective mass for the fermionic quasiparticles, indicating a large spin fluctuation near pressure-induced quantum phase transition, which will be discussed later.

Next, we discuss briefly the thermodynamic property on the slope of the phase boundary at T_{N2}. At ambient pressure, the discontinuity of the entropy ΔS is estimated to be 0.06 J/mol K.[13] ΔV is obtained from the discontinuity of the thermal expansion, $\Delta V/V=-6\times10^{-5}$ (Ref. 13) and $V=4.8\times10^{-5}$ m^{-3}/mol is taken from the lattice parameter. By using the Clausius-Clapeyron relation, $\partial T_{N2}/\partial P=\Delta V/\Delta S\sim0$. Although the discontinuity of the volume ΔV is almost independent of pressure,[8,14] the slope of $\partial T_{N2}/\partial P$ becomes steep as pressure increases. It is suggested that ΔS is suppressed by applying pressure below P_{C2}.

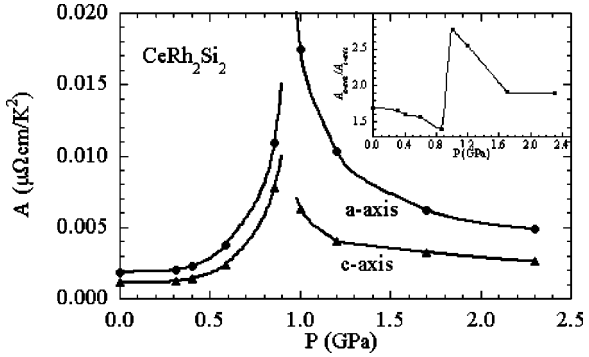

FIG. 5. Pressure dependence of the coefficient of T^2 of $CeRh_2Si_2$ along the a and c axes. The inset shows the ratio A_a/A_c as a function of pressure.

The resistivity ρ is described as $\rho=\rho_0+AT^2$ at low temperature in a wide pressure range, where ρ_0 is the residual resistivity and A the constant. Figure 4 shows the T^2 dependence below 2.3 GPa along the a and c axes. In the case of the present work, the T^2 term of ρ is attributed to the effect of spin fluctuation or the contribution from electron-magnon scattering.

The values of A for the a and c axes, A_a and A_c, are shown in Fig. 5 as a function of pressure. Broken curves are a guide to the eyes. If the T^2 term of ρ is mainly attributed to the electron-magnon scattering, the value of $A(P)$ should decrease up to $P_{C1}\sim1.0$ GPa where magnetic ordering disappears. However, it is found that both $A_a(P)$ and $A_c(P)$ show maxima near 1.0 GPa, where the effect of spin fluctuation is expected to be most significant. It means that the large enhancement in the T^2 term is not due to the electron-magnon scattering but due to the existence of large spin fluctuation around the critical pressure P_{C1}, which corresponds to the pressure-induced quantum phase transition. Qualitatively, the pressure dependence of $A(P)$ is consistent with that of Sommerfeld coefficient γ of specific heat,[16] in which $\gamma(P)$ increases from 20 mJ/mol K^2 at ambient pressure and passes through a broad maximum of 80 mJ/mol K^2 near 1.0 GPa. On the other hand, no anomaly is observed near P_{C2}. Since this transition is first order, no fluctuation may exist and therefore the coefficient A is not affected.

It is noted that the value of A shows large anisotropy between the a and c axes. The inset in Fig. 5 shows the pressure dependence of the ratio A_a/A_c. A discontinuity is observed at the critical pressure near P_{C1} and a small anomaly near P_{C2}. The resistivity is usually calculated from the displacement of the Fermi surface by an electric field. When the Fermi surface is anisotropic, the resistivity depends on the direction of the current density in the crystal. If the values of A_a and A_c come from the umklapp process of the electron-electron collisions, they may be related to the area (or cross section) of Fermi surfaces that have been sliced perpendicular to the a and c directions in k space. Because several scattering mechanisms are included in the

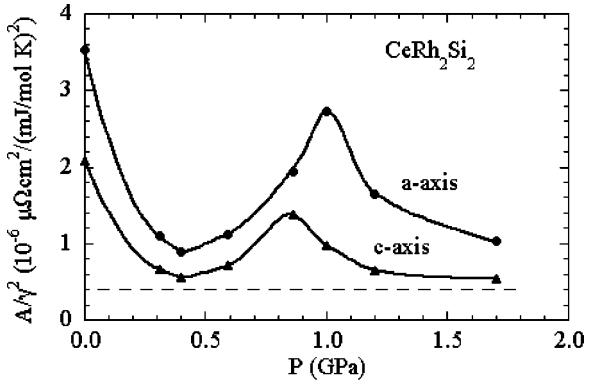

FIG. 6. Pressure dependence of the ratio of A/γ^2 of $CeRh_2Si_2$ along the *a* and *c* axes.

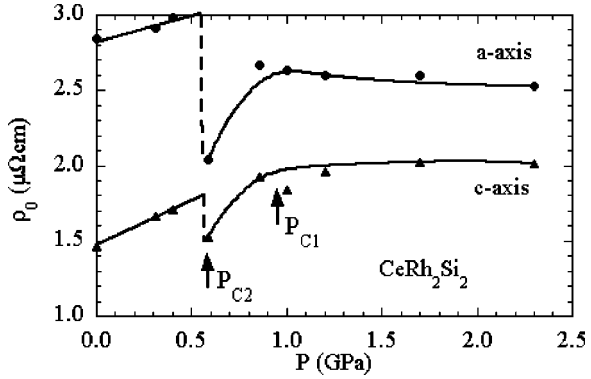

FIG. 7. Pressure dependence of the residual resistivity ρ_0 of $CeRh_2Si_2$ along the *a* and *c* axes.

electrical resistivity, it is suggested that this transition corresponds a change in the topology of the Fermi surface at high pressure.

In general, the coefficient A of T^2 term in the resistivity and the linear specific heat coefficient γ appear to have the so-called Kadowaki-Woods relation $A \propto \gamma^2$.[17,18] If the effective mass of conduction electron is essentially determined by the properties of a conventional band without a many body effect, such as in ordinary transition metals, the ratio $A/\gamma^2 \sim 0.4\times10^{-6}$ is expected. In the case for HF compounds, on the other hand, the heavy mass is essentially due to the many-body dynamical effect between the lattice of local moments and the conduction electrons. It should be noted that for the system with many-body correlations, the ratio A/γ^2 must have the value larger than that of transition metals.

In order to investigate the many-body electron correlation near QPT, the value of A/γ^2 is plotted in Fig. 6 as a function of pressure. The value of γ is taken from the result of specific heat under pressure.[16] The ratio A/γ^2 is found to depend on pressure since the behavior of $A(P)$ is different quantitatively from that of $\gamma(P)$. The dashed line means the universal value $A/\gamma^2 \sim 0.4\times10^{-6}$ which the ordinary transition metals make.[18] At ambient pressure, both A_a/γ^2 and A_c/γ^2 are much larger than those of transition metals. This indicates that the effect of magnetic fluctuation is included in both A and γ values. At low pressures below 0.4 GPa, A/γ^2 decreases rapidly, which is a result of the vanishing of magnetic order. Above 0.6 GPa, however, A/γ^2 is found to increase strongly with pressure below 0.9 GPa. Moreover, it decreases again with pressures up to 1.7 GPa, the value of which, $A_a/\gamma^2 \sim 1.0\times10^{-6}$ and $A_c/\gamma^2 \sim 0.5\times10^{-6}$ at 1.7 GPa, is almost the same as that of transition metals. This indicates that the large fluctuation is induced at the critical pressure P_{C1}, and a crossover from HF state to intermediate valence state occurs at high pressure.

Such behavior has been observed in a ferromagnet UGe_2.[19,20,22] It shows ferromagnetic ordering at $T_C = 52$ K at ambient pressure. T_C and T^*, which are characteristic transition temperatures observed below T_C, decrease with increasing pressure and become zero around 1.9 and 1.2 GPa. At low pressures below 1 GPa, the value of $A/\gamma^2 \sim 1\times10^{-5}$ is near the empirical universal value for HF systems. Up to 1.1 GPa, A/γ^2 decreases to 3.3×10^{-6}, where the transition characterized by T^* disappears. Moreover it increases significantly again to 9×10^{-6} around 1.3 GPa, where the value of $A(P)$ shows a peak. This result implies that the peak in the A/γ^2 vs P curve is closely related to the occurrence of the superconductivity in these materials.

Next, the values of the residual resistivity $\rho_0(P)$ for the *a* and *c* axes are shown in Fig. 7 as a function of pressure. Solid curves are guides to the eyes. As for both the *a* and *c* axes, $\rho_0(P)$ increases slightly as pressure increases, and decreases with a minimum near 0.6 GPa$\sim P_{C2}$, where the magnetic phase transition disappears ($T_{N2}=0$). The result indicates that the critical point near P_{C2} affects the magnitude of ρ_0. The discontinuous change of ρ_0 at P_{C2} along the *a* axis is larger than that along the *c* axes.

It is difficult to show whether an anomaly on $\rho_0(P)$ exists near P_{C1}. In the case of a ferromagnetic QCP, the anomaly of ρ_0 is expected to be observed at the critical point, while the less pronounced anomalies expected in the case of AF-QCP.[21] In the case of $CeRh_2Si_2$, on the other hand, the resistivity exhibits a T^2 dependence at low temperature having the large coefficient $A(P)$ near P_{C1}. The value of A is

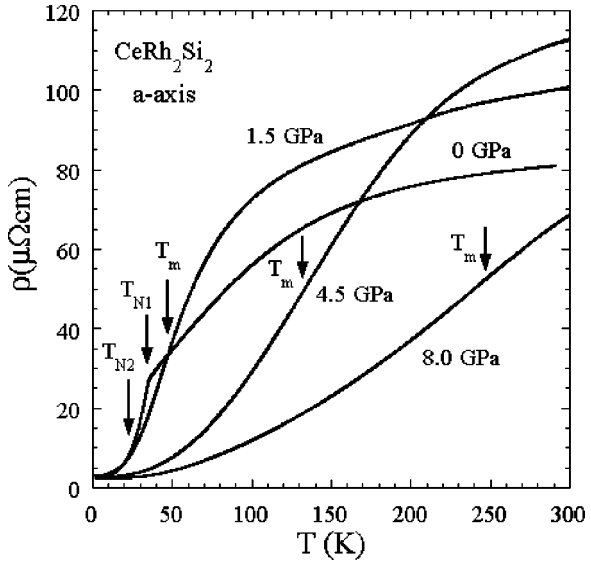

FIG. 8. Temperature dependence of the electrical resistivity of $CeRh_2Si_2$ along the *a* axis up to 8 GPa.

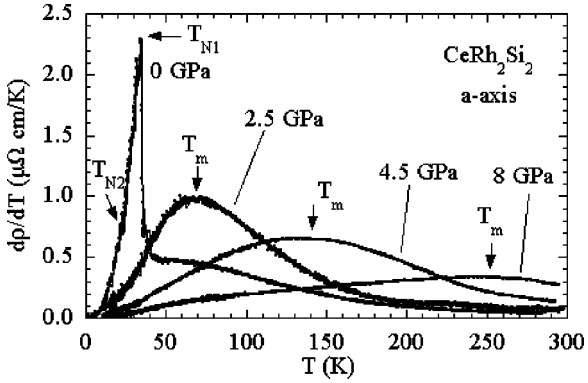

FIG. 9. Temperature derivative $d\rho/dT$ of $CeRh_2Si_2$ up to 8 GPa.

usually attributed to the Umklapp process, which is related to the magnitude of spin fluctuation in the present work. Although, it is observed that $\rho_{4.2\ \mathrm{K}}$ shows a peak at P_{C1} in the polycrystalline sample,[22] it may comes from the large value of the AT^2 term contributes to $\rho(P)$ at 4.2 K and then $\rho(P)$ may also show a maximum even when it shows no anomaly at $T \rightarrow 0$ K.

B. Kondo scattering above 2 GPa

Figure 8 shows the temperature dependence of the electrical resistivity in a wide temperature range under high pressures up to 8 GPa. No anomaly was detected in the $\rho(T)$ above 1.5 GPa since magnetic orderings were suppressed completely. Instead, an inflection point at T_m is observed in the $\rho(T)$ curve above 1.5 GPa. T_m is found to shift rapidly from 44 K at 1.5 GPa to higher temperature with increasing pressure. To show the results more clearly, the temperature derivative of $\rho(T)$ curve was calculated. The values of $d\rho/dT$ are shown in Fig. 9 as a function of T. A peak is clearly seen above P_{C1}. T_m, which is defined as the temperature showing the peak, increases with increasing pressure.

T^2 dependence was observed at low temperature below T_m. The coefficients A_a decrease significantly with increasing pressure: the value of A_a at 8 GPa is smaller that at 1.5 GPa by two orders of magnitude. Figure 10 shows the pressure dependence of the values of A_a and T_m^{-2} up to 8 GPa. It

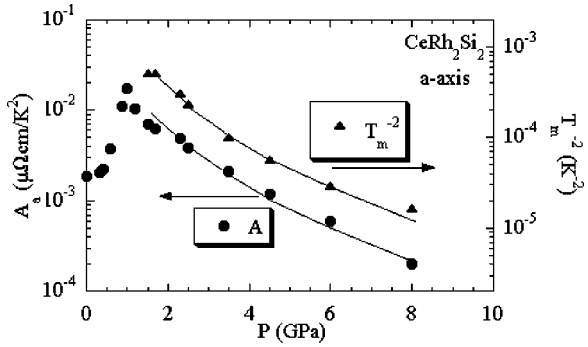

FIG. 10. A_a and T_m^{-2} as a function of pressure. Two solid lines are guides to eye to show that the curve of A vs P is the same as that of T_m^{-2} vs P.

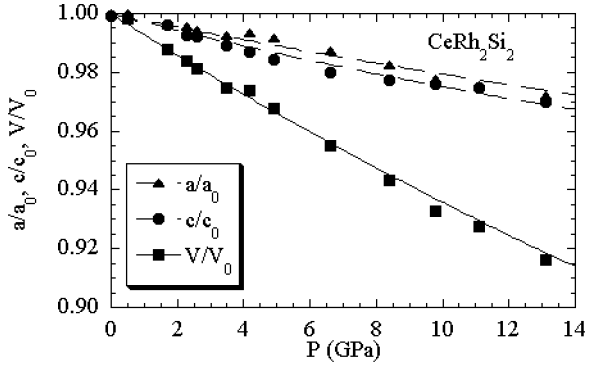

FIG. 11. Pressure dependence of a/a_0, c/c_0, and V/V_0 of $CeRh_2Si_2$. The solid line for the pressure dependence of V/V_0 shows the result of the least square fitting of Murnaghan's equation. The dashed lines for a/a_0 and c/c_0 are guides to the eye.

is found that the pressure vs A curve is proportional to that for T_m^{-2}, i.e., A is approximately proportional to T_m^{-2} above P_{C1}. According to the theory of Yoshimori and Kasai,[23] the value of A is proportional to T_K^{-2}. It has been confirmed that the relation $A \propto T_m^{-2}$ is valid in the heavy fermion materials.[24] These facts indicate that T_m is proportional to a characteristic temperature or Kondo temperature T_K, $T_m \propto T_K$.

T_K is also estimated from the temperature where magnetic susceptibility shows the peak. At ambient pressure, T_K is obtained to be $\sim$35 K and increases with pressure.[25] The slope of dT_K/dP is calculated to be 0.046 K/GPa from the result up to 1 GPa, while the large value $dT_m/dP > 15$ K/GPa above 1.5 GPa is obtained in the present work. It is considered that below P_{C1}, two kinds of interactions, the RKKY interaction and the Kondo effect, are competing with each other and T_K is related not only to the Kondo effect but also to the RKKY interaction. Above P_{C1}, where the antiferromagnetic interaction disappears, T_K is expected to become sensitive with pressure as typical heavy fermion compounds.

C. X-ray diffraction at high pressure

In order to investigate pressure dependence of lattice parameters, x-ray diffraction measurements were carried out.

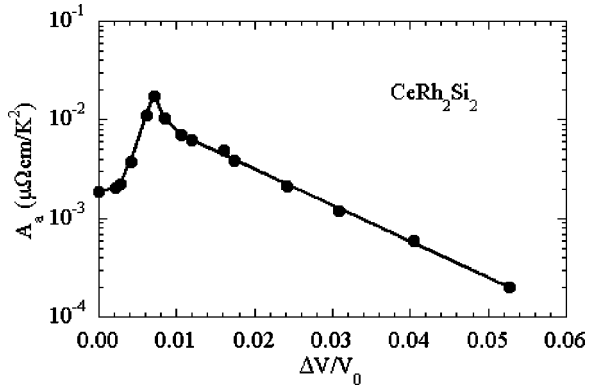

FIG. 12. A_a as a function of fractional change in volume $\Delta V/V_0$.

At ambient pressure, the crystal structure is tetragonal having the lattice parameters of $a=4.070$ Å and $c=10.156$ Å, which are consistent with previous experiments.[13] The lattice constants are determined mainly from the reflections (101), (103), and (112). Figure 11 shows the pressure dependences of the relative lattice parameters $a/a_0, c/c_0$ and the relative volume V/V_0 as a function of pressure at room temperature, where a_0, c_0, and V_0 are the values at ambient pressure. It was revealed that the tetragonal structure is stable up to 13 GPa at room temperature. Both a and c decrease with increasing pressure, and no discontinuous change is observed within the experimental errors. The c axis is more compressible than the a axis, and linear compressibilities $\kappa_i=-i^{-1}\partial i/\partial P$, $i=a$ or c, are estimated to be 2.2×10^{-3} and 3.0×10^{-3} GPa^{-1}.

We attempted a least-squares fit of the data of V/V_0 to the first-order Murnaghan's equation of state $P=(B_0/B_0')\times[(V_0/V)^{B_0'}-1]$, where B_0 denotes the bulk modulus at ambient pressure and B_0' is its pressure derivative. The results are shown in Fig. 11 as a solid curve for V/V_0. The agreement between the observed points and the calculated ones is satisfactory. B_0 and B are estimated to be 139 and 2.2 GPa, respectively. The value of B_0 of $CeRh_2Si_2$ is comparable with those of typical heavy Fermion compounds such as $CeCu_6$ and $CeInCu_2$.[26]

Here we estimate the Grüneisen parameter to evaluate the stability of electronic state above 2 GPa. The Grüneisen parameter Γ of T_K is written[26]

$$\Gamma=-\left.\frac{\partial \ln T_K}{\partial \ln V}\right|_{V=V_0}=\frac{1}{2}\left.\frac{\partial \ln A}{\partial \ln V}\right|_{V=V_0}=\frac{1}{2}\frac{\Delta(\ln A)}{\frac{\Delta V}{V_0}},$$

where A is the coefficient of T^2 term. In Fig. 12, the values of A are plotted in logarithmic scale as a function of $\Delta V/V_0$. The linear relationship is found in the plot as is shown by solid lines above 1.5 GPa ($\Delta V/V_0>0.011$). From the result, Γ is estimated to be 42 for $CeRh_2Si_2$, which is extremely large and comparable with those of heavy fermion compounds 59 and 65 for $CeInCu_2$ and $CeCu_6$, respectively.[26] Below 1.5 GPa, on the other hand, the observed values deviate significantly from the linear relation, suggesting a large enhancement of the Grüneisen parameter Γ near P_{C1}. This indicates that the electronic state is very unstable near P_{C1}.

IV. CONCLUSION

We have observed the electrical resistivity along the a and c axes of single crystalline $CeRh_2Si_2$ under high pressure. As for the two magnetic-phase transitions, it has been found that the transition at T_{N1} is second order and at T_{N2}, first order. Temperature dependence of the resistivity shows T^2 dependence at low temperature in a wide pressure range from ambient pressure up to 8 GPa, implying classical Fermi liquid behavior. The pressure dependence of the value of A/γ^2, however, suggests that the many-body dynamical effect is enhanced near the critical pressure P_{C1}. On the other hand, a discontinuous change near P_{C2} has been observed in the pressure dependence of the residual resistivity $\rho_0(P)$.

From the measurements of the electrical resistivity and lattice parameters of $CeRh_2Si_2$ at high pressure, the volume dependence of Kondo temperature T_K was discussed. The Grüneisen parameter of T_K is estimated to be 42 above 1.5 GPa. The magnitude is comparable with those of heavy fermion compounds.

ACKNOWLEDGMENTS

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