Uniaxial Fermi-surface nesting and spin-density-wave transition in the heavy-fermion compound $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$

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(Received 24 April 1997)

Resistivity parallel and perpendicular to the tetragonal c axis of the antiferromagnetic heavy-fermion compound $Ce(Ru_{1-x}Rh_x)_2Si_2$ for $x=0.15$ has been measured in various fields along the c axis of the single crystal. A sharp upturn is observed just below the Néel temperature T_N only in the parallel direction. This is interpreted in terms of partial gapping of the Fermi surface along the uniaxial c direction. The specific heat and susceptibility below T_N are, respectively, analyzed into two independent contributions in the low-temperature limit: one is an exponential contribution of the gapped part of the Fermi surface with the energy gap about 10 K, and the other is a residual heavy-fermion contribution due to the remaining ungapped part of about 65%. These facts suggest an itinerant spin-density-wave transition based on Fermi-surface nesting in a heavy-electron band. [S0163-1829(97)00241-5]

I. INTRODUCTION

The heavy-fermion compound $CeRu_2Si_2$ with a $ThCr_2Si_2$-type tetragonal structure has been extensively studied since it shows paramagnetic behaviors down to 20 mK (Ref. 1) with short-range antiferromagnetic (AF) correlations below 70 K (Ref. 2) and a metamagneticlike transition below 10 K with the critical field $H_M \sim 7.8$ T in the magnetically easy c-axis direction.[1] This transition has recently been interpreted in terms of the sharp crossover from the itinerant-electron paramagnetism to the local-moment system.[3,4] Substitution of the second element into each Ce, Ru, or Si site induces the AF ordering.[5–7] Therefore, this compound is supposed to be nearly on the edge of the magnetic ordering phase. In $(Ce,La)Ru_2Si_2$ it has been demonstrated that H_M is reduced and an AF ordering is stabilized when several percentages of the Ce atoms are replaced by La. The effect of the La substitution is mainly explained by a negative chemical pressure effect due to the larger atomic volume of La than that of Ce. Substitution of a $4d$ transition metal such as Mo, Rh, or Pd to the Ru site induces systematic changes in H_M and Kondo temperature T_K. These facts have been summarized as another interesting alloying effect sensitive to the $4d$ conduction electron number.[6,8,9] In the $Ce(Ru_{1-x}Rh_x)_2Si_2$ system with a low concentration of Rh for $0.05<x<0.3$, the AF transition has been discovered using specific heat and susceptibility measurements.[6] Microscopic NMR[10] and muon spin rotation (μSR)[11,12] measurements support the AF transition in this system with the presence of inhomogeneous local fields. A recent neutron-diffraction experiment for $x=0.15$ shows that the AF phase has an incommensurable sinusoidal spin modulation with wave vector $\tau=(0,0,0.420)$ and that the magnetic moment is polarized along the c axis with an amplitude of $0.65\mu_B/Ce$.[13] Further interest in this system arises from the question of whether the incommensurable spin structure is associated with a Fermi-surface instability and the formation of a spin-density wave (SDW) or not. Resistivity is one of the powerful measurements of probing such a Fermi surface anomaly as has been typically performed on AF Cr with a SDW transition. Recent resistivity measurements on the cubic heavy-fermion compound YbBiPt have also suggested the presence of a partial gapping of the Fermi surface and the development of an SDW in a heavy-mass conduction band.[14] Thus, we have studied the resistivity of the heavy-fermion compound $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$ parallel and perpendicular to the tetragonal c axis, respectively, in various magnetic fields. It is found that the results, combined with specific heat and magnetic susceptibility measurements, suggest the development of a partial energy-gap opening of the Fermi surface due to a uniaxial Fermi-surface nesting and SDW ordering in the present heavy-fermion Ce compound.

II. EXPERIMENT

A polycrystalline $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$ sample was prepared by arc melting a stoichiometric amount of Ce($4N$), Ru($3N$), Rh($3N$), and Si($6N$) in an argon atmosphere followed by annealing at 800 °C for one week. X-ray diffraction confirms the sample to be the body-centered-tetragonal $ThCr_2Si_2$-type structure. A single-crystal sample of $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$ was grown by the Czochralski technique in a tri-arc furnace. The resistivity measurements for the single-crystal sample were made above 0.4 K in a ^{3}He cryostat by using a standard four-probe ac technique. The specific heat was determined above 2 K on the polycrystalline sample using a semiadiabatic technique. The magnetic susceptibility was measured above 2 K on the single-crystal sample in a conventional superconducting quantum interference device magnetometer.

III. EXPERIMENTAL RESULTS

Figure 1 displays the results of the resistivity measurements for two small slices cleaved to directions parallel and

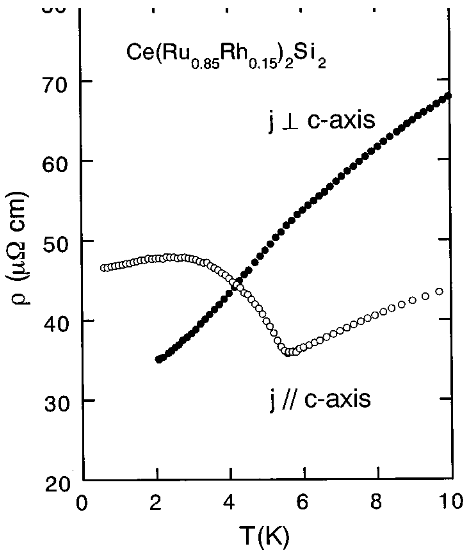

FIG. 1. Temperature dependence of the resistivity of $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$ with excitation current j applied along the c axis ($\rho_\parallel$) and in the basal plane ($\rho_\perp$).

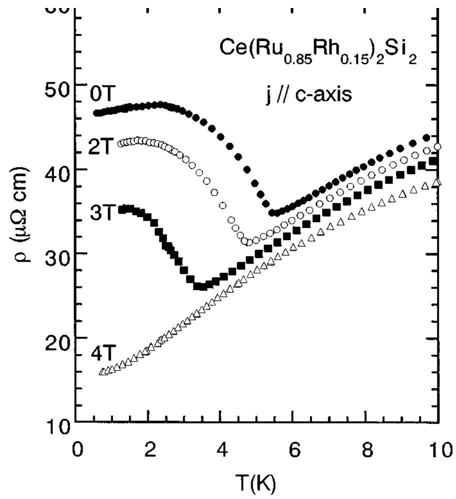

FIG. 2. Temperature dependence of the resistivity of $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$ with excitation current j applied along the c axis ($\rho_\parallel$) in various magnetic fields along the c axis.

perpendicular to the tetragonal c axis with excitation current, respectively, parallel ($\rho_\parallel$) and perpendicular ($\rho_\perp$) to the c axis. As shown in this figure, the temperature dependence of each resistivity is strongly anisotropic. With decreasing temperature from 10 K $\rho_\parallel$ decreases and shows a sharp upturn bend at around the Néel temperature $T_N = 5.5$ K. With further decreasing temperature $\rho_\parallel$ shows a broad maximum. On the other hand, $\rho_\perp$ shows rather a continuous decrease below 10 K with a subtle bend around T_N. The sharp rise in resistivity only in the parallel direction $\rho_\parallel$ suggests a Fermi surface instability and the presence of a partial gapping of the Fermi surface along the uniaxial c-axis direction. A similar resistivity anomaly around the magnetic phase transition temperature has been shown in bcc Cr metal[15] and the fcc YbBiPt heavy-fermion compound,[14] in which an itinerant-electron SDW transition is expected to occur with a partial gap in the conduction band. In these cases the resistivity anomaly can be anisotropic depending on the direction of the excitation current. However, a complete uniaxial anomaly has not been observed yet in any samples. One of the reasons should be due to a magnetic polydomain structure in the cubic crystals. An anomaly present only in the resistivity $\rho_\parallel$ would be the first clear evidence of uniaxial conduction-band gapping in metals and metallic compounds.

To further investigate this transition of $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$, we performed a series of resistance measurements of this compound in magnetic fields. Figure 2 displays the temperature dependence of the resistivity $\rho_\parallel$ in various fields along the c axis. With increasing field B, the sharp upturn around T_N in $\rho_\parallel$ shifts to a lower temperature with keeping the shape of the temperature dependence above T_N. In contrast to this, $\rho_\perp$ does not show any clear change by field along the c axis as shown in Fig. 3. The anomaly in $\rho_\parallel$ disappears at $B = 4$ T within the observed temperature range. These facts indicate that the gap opening of the Fermi surface goes to a lower temperature with the magnetic field along the c axis and disappears at $B = 4$ T. We have measured the resistivity $\rho_\parallel$ and $\rho_\perp$ in magnetic fields perpendicular to the c axis and found that nothing changed in the field in both resistivities. In order to investigate the field dependence of the magnetic transition, we measured the field sweep of the resistivity at various temperatures above 1.5 K. An anomalous inflection in the magnetoresistance occurs below T_N (the figure is not shown here), suggesting a collapse of the gap. The field of the inflection increases with decreasing temperature in accordance with the temperature dependence of the resistivity in field. We measured the temperature dependence of the susceptibility $\chi_\parallel$ in various fields B along the easy c axis as shown in Fig. 4. The result is basically consistent with that reported for $Ce(Ru_{1-x}Rh_x)_2Si_2$, $x = 0.1$.[6] With increasing B, T_N, defined by the temperature of a sharp inflection just below a broad maximum in $\chi_\parallel$, decreases. For $B \geqslant 3.5$ T, no anomaly can be seen in this

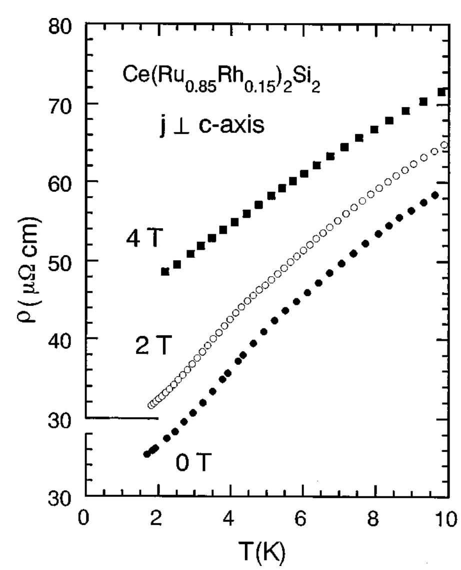

FIG. 3. Temperature dependence of the resistivity of $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$ with excitation current j applied in the basal plane ($\rho_\perp$) in various magnetic fields along the c axis.

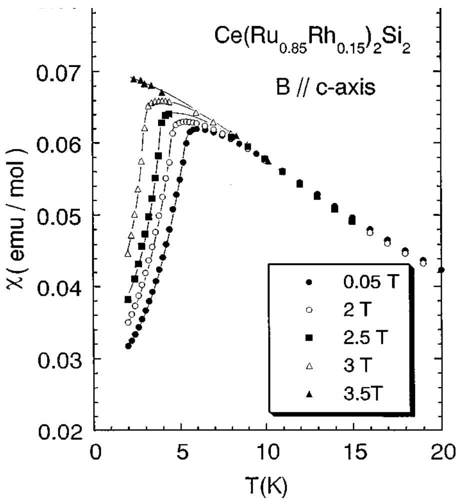

FIG. 4. Temperature dependence of the susceptibility of $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$ in various fields along the c axis.

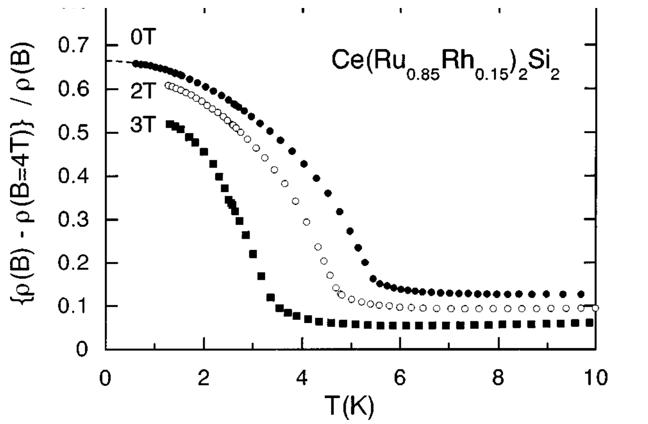

FIG. 6. Relative resistivity ratio of $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$ with excitation current j applied along the c axis ($\rho_{\parallel}$) in various fields along the c axis.

figure. Figure 5 shows the B-T magnetic phase diagram of $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$, where the temperature of the upturn in $\rho_{\parallel}$, the inflection in the magnetoresistance, and $\chi_{\parallel}$ are plotted. These temperatures follow a single curve well. Therefore, it is suggested that a gapping of the Fermi surface parallel to c axis is well linked with the antiferromagnetic SDW transition in the field along the c axis. The solid curve in this figure is a BCS energy gap scaled to pass through the points ($T_N = 5.4$ K, $B = 0$) on the x axis and ($T = 0$, $B_c = 3.3$ T) on the y axis. The curve fits the data very well, indicating that the transition can be described in the framework of weak-coupling BCS theory or mean-field theory of itinerant antiferromagnetism (SDW) based on the two-band mechanism.[16] Note that the value of the critical field B_c is almost the same as T_N in energy scale.

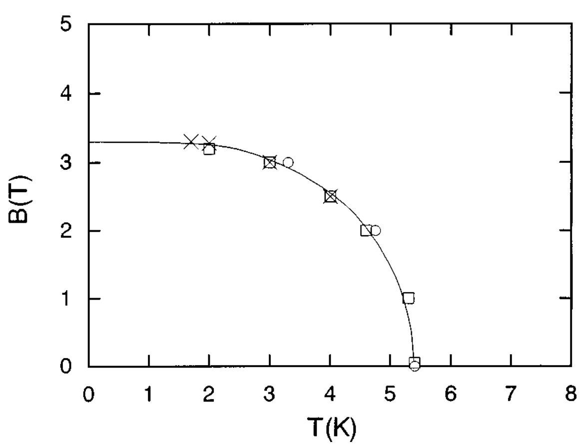

FIG. 5. B-T phase diagram of $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$ in the field parallel to the c axis. The open circle and cross are results of the temperature sweeps of resistivity and magnetization, respectively, in various fields. The open square is the result of the field sweep of resistivity in various temperatures. The solid line is a BCS energy gap scaled fixed by the points ($T_N = 5.4$ K, $B = 0$) and ($T = 0$, $B_c = 3.3$ T).

IV. DISCUSSION

Here, we discuss the heavy-fermion contribution to the resistivity $\rho_{\parallel}$ along the c axis of $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$ within the SDW picture. Following a former precise analysis of the conductivity of Cr by McWhan and Rice[17] and the recent analysis of YbBiPt by Movshovich *et al.*,[14] the total conductivity is written as $\sigma = \sigma_1 + \sigma_2$ if one assumes that the Fermi surface is divided into two independent parts, where indices 1 and 2 refer to, respectively, the ungapped and gapped magnetic regions of the Fermi surface at zero magnetic field. Then, the relative change in conductivity produced by the SDW is given by

$$\frac{\sigma_p - \sigma_g}{\sigma_p} = \frac{\rho_g - \rho_p}{\rho_g} = \frac{\sigma_{2p}}{\sigma_p}\left(1 - \frac{\sigma_{2g}}{\sigma_{2p}}\right). \quad (1)$$

Here, the subscripts g and p refer to the case when the magnetic region 2 is, respectively, gapped or made paramagnetic by application of field $B = 4$ T. Then, we estimate the ratio (1) experimentally for various fields as shown in Fig. 6. As is seen in Figs. 2 and 6 we observe a small negative temperature-independent resistivity shift by field in the whole temperature range. Correcting this contribution as a trivial paramagnetic background effect, we can roughly estimate zero-temperature value of the ratio (1) in zero field as ~ 0.5 taking the extrapolation of $T \rightarrow 0$ K in this figure. The ratio σ_{2g}/σ_{2p} can be calculated by using the formula given by a SDW gap Δ and T_N (see Refs. 10 and 13). For $T \rightarrow 0$ K the contribution of the gapped magnetic part σ_{2g}/σ_{2p} is zero in any case. Therefore, we obtain $(\sigma_p - \sigma_g)/\sigma_p = \sigma_{2p}/\sigma_p = 0.5$ for zero field. The ratio σ_{2p}/σ_p is related to the percentage of the Fermi surface that is gapped along the c axis. For comparison, in Cr and YbBiPt it was found that the σ_{2p}/σ_p value is 0.3 and 0.33, respectively.

For the next step, previous results of specific heat measurements on the $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$ polycrystalline sample[6] have been analyzed by using the SDW picture. The magnetic part of the specific heat C_m in the temperature range from 2 to 10 K is obtained from the substraction of the reference material $LaRu_2Si_2$ data as the lattice and the non-$4f$ electronic contributions. The result is given in Fig. 7. As

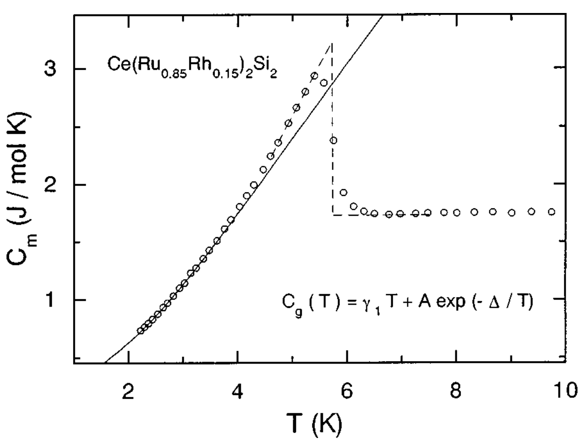

FIG. 7. Specific heat of the $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$ polycrystal as a function of temperature. The solid line is the best fit curve of Eq. (2) in the text. The dashed line is an ideal step determined from an equal area construction.

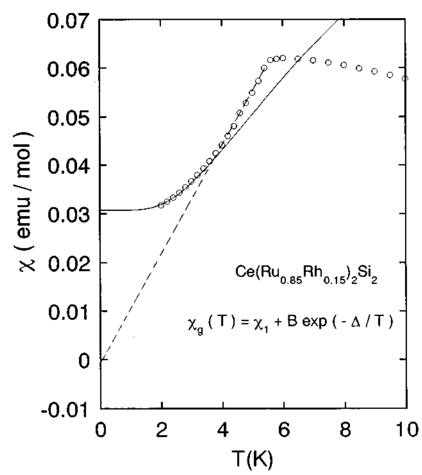

FIG. 8. Temperature dependence of the susceptibility of $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$ in field $B=0.05$ T along the c axis. The solid line is the best fit curve of Eq. (3) in the text. The dashed line is a theoretical asymptotic line described in the text.

a decrease of temperature the heat capacity $C_m(T)$ shows a step at T_N which implies a mean-field-like phase transition as a BCS superconductor. This step is evalued as $\Delta C=1.5$ J/mol K from a dashed curve in Fig. 7 determined from an equal area construction. According to weak-coupling BCS theory, $\Delta C/\gamma T_c$ should be equal to 1.43 with full gap formation on the Fermi surface (here, T_c is the superconducting temperature). Therefore, if we know the electronic specific heat coefficient γ_p in the paramagnetic region, $\Delta C/\gamma_p T_N$ would be one of the measures for the Fermi surface with a heavy mass that is partly gapped in the present system. It is rather difficult to determine the coefficient γ_p in the paramagnetic state because it is still temperature dependent. Here, we try to estimate γ_p from the Kondo temperature T_K as $\gamma_p \propto 1/T_K$. T_K was determined from the temperature of a broad maximum in the specific heat as 24 K for $x=0$ and 20 K for $x=0.15$.[6] Thus, from the experimental value of $\gamma=380$ mJ/mol K^2 for $x=0$,[6] γ_p for $x=0.15$ is evaluated as 460 mJ/mol K^2. The ratio $\Delta C/\gamma_p T_N$ is found to be 0.60. This value indicates that ~40% of the Fermi surface with heavy mass is gapped for $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$. The remaining 60% of the Fermi surface with heavy mass is expected to be still in the ungapped Fermi-liquid state below T_N. The data in the low-temperature region below $T_N/2$ fits the expression

$$C_g(T)=\gamma_1 T+A\ \exp(-\Delta/T) \tag{2}$$

well, as shown by a solid line in Fig. 7. The first term indicates the contribution of the conduction electron in the remaining ungapped part of the Fermi surface. The second term suggests the electronic contribution in the gapped magnetic part described within BCS theory and the SDW hypothesis. From best fits to the data below $T_N/2$ we estimate the energy gap in the low-temperature limit as $\Delta=10$ K, the residual γ value in the AF temperature range as $\gamma_1=300$ mJ/mol K^2, and the coefficient A as $A=7$ J/mol K. The ratio Δ/T_N is calculated as 1.8, which is comparable to 2.3 in Cr (Ref. 15) and consistent with the minimum value of 1.764 for the two-band model of itinerant antiferromagnetism.[16] There are so far, two heavy-fermion systems, URu_2Si_2 (Ref. 18) and YbBiPt (Ref. 14), analyzed within the SDW hypothesis. In these cases ratios of $\Delta/T_N=7.2$ and 1.65 has been reported, respectively. The obtained residual γ_1 value for $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$ is about 65% of the γ_p estimated in the paramagnetic temperature range. The ratio γ_1/γ_p can be one of the best measures for the remaining ungapped part of the Fermi surface at 0 K and is in agreement with the percentage extracted from the ratio of $\Delta C/\gamma_p T_N$. It is necessary to confirm the residual γ_1 value from the low-temperature specific heat measurements below 1 K. Above $T_N/2$ the fitted curve does not follow the data. In such a high-temperature region it is necessary to consider the temperature dependence of $\Delta(T)$. Furthermore, expression (2) would not be valid any more.

Within an SDW picture of the 5.5 K phase transition with the gap temperature $\Delta=10$ K determined from the specific heat in $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$, we try to fit the susceptibility data below $T_N/2$ for $B=0.05$ T using the expression

$$\chi_g(T)=\chi_1+B\ \exp(-\Delta/T). \tag{3}$$

Here, χ_1 is interpreted as the residual Pauli paramagnetic contribution coming from the ungapped Fermi surface at 0 K. The second term suggests the longitudinal susceptibility arising from the gapped part of the Fermi surface described by mean-field SDW theory.[16] A good fit has been done for $\chi_1=0.031$ emu/mol and $B=0.13$ emu/mol as shown as a solid line in Fig. 8. Above $T_N/2$ the fitted curve does not follow the susceptibility data. This may be due to a similar reason described before for the specific heat data above $T_N/2$. The initial decrease just below T_N follows an asymptotic proportional line from $\chi_p=0.062$ emu/mol at T_N to almost zero at 0 K well, as shown as a dashed line in Fig. 8, written as $\chi(T)=\chi_1+(\chi_p-\chi_1)(2T/T_N-1)$. This form is consistent with SDW theory with a correction of the residual χ_1 term. Neglecting the weak temperature dependence of the susceptibility in the paramagnetic state, χ_p can be regarded as the Pauli paramagnetic susceptibility just before the gap opening. Then, the ratio $\chi_1/\chi_p \sim 0.5$ may also represent an approximate percentage of the remaining ungapped part of the Fermi surface with a heavy mass at 0 K. This value is a little smaller than $\gamma_1/\gamma_p=0.65$ estimated from the specific heat analysis, but is rather comparable with the residual resistivity ratio given as $\sigma_1/\sigma_p=1-\sigma_{2p}/\sigma_p=0.5$. For the moment, we do not consider the anisotropy, namely, the susceptibility $\chi_\perp$ along the magnetic hard plane. In order to explain the specific heat, susceptibility, and resistivity at the same time, we need a more detailed discussion after taking account of the strong anisotropy and the shape of the Fermi surface. However, the present resistivity, specific heat, and susceptibility of the heavy-fermion $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$ are, in total, described within an itinerant SDW picture based on the Fermi-surface nesting along the c axis in a heavy-fermion conduction band.

We discuss the uniaxial energy-gap opening parallel to the c axis partially on the Fermi surface from the resistivity anomaly only for the c axis and the exponential temperature dependence of the specific heat and susceptibility below $T_N/2$. In order to proceed with our discussion, we may consider the contribution of continuous gapless states on the Fermi surface as has been discussed in anisotropic superconductivity.[19,20] The existence of gapless states would give rise to a power-law temperature-dependent term in thermal properties. This effect should be prominent in the low-temperature limit. At present we are not ready to discuss such an intrinsic low-temperature excitation because of the lack of our data below 1 K.

V. CONCLUSION

In summary, just below $T_N=5.5$ K a sharp rise in the resistivity of $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$ was observed only in the tetragonal c-axis direction. Its temperature decreases with increasing field along the c axis in agreement with a theoretical curve of the weak-coupling BCS energy gap $\Delta(T)$. This suggests the development of a partial gap opening of the Fermi surface along the c axis following the uniaxial Fermi-surface nesting and a SDW transition. The residual conductivity along the c axis after the gap opening below T_N has been estimated as $\sigma_1/\sigma_p=0.5$ for $T\rightarrow 0$ K. The percentage of the ungapped part for the heavy-fermion conduction band below T_N has been determined from the ratio of the electronic specific coefficient as $\gamma_1/\gamma_p=0.65$. The value $1-\gamma_1/\gamma_p=0.35$ is consistent with another experimental measure $\Delta C/\gamma_p T_N$ for the gapped part on the assumption of the weak-coupling BCS theory. The residual Pauli paramagnetic contribution along the c axis due to the ungapped part is also estimated as $\chi_1/\chi_p=0.5$. Thus, we conclude that the gap is formed mainly in the $4f$ conduction band with the heavy mass due to the SDW transition. From the exponential temperature dependence of the specific heat and susceptibility in the low-temperature limit, the energy gap has been obtained as $\Delta=10$ K at 0 K and therefore, $\Delta/T_N=1.8$. This value is consistent with a gap parameter determined from the SDW model based on the BCS theory.

ACKNOWLEDGMENTS

The authors would like to express their sincere thanks to Professor F. J. Ohkawa, Hokkaido University, for valuable discussion. This work was supported by a Grant-in-Aid for Scientific Research on Priority Areas from the Ministry of Education, Science, Sports and Culture, Japan.

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