Spin-density-wave state under pressure in the heavy-fermion compound $Ce(Ru_{0.95}Rh_{0.05})_2Si_2$

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We report the effects of pressure on the spin-density-wave (SDW) state in the heavy-fermion compound $Ce(Ru_{0.95}Rh_{0.05})_2Si_2$ revealed by specific heat measurements up to a pressure of 1.6 GPa. The Néel temperature (T_N) and the Sommerfeld coefficient (γ) decreased with increasing pressure. When the pressure exceeded 0.7 GPa, the SDW transition was suppressed. We give a special emphasis to the anisotropy in the $Ce(Ru_{1-x}Rh_x)_2Si_2$ system and suggest that the lattice parameter c and the distance between the Ru(Rh) and Ce atoms play an important role in the SDW transition. In terms of the decrease of the Néel temperature, the effect of chemical substitution of Rh for Ru is similar to the change observed under pressure.

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I. INTRODUCTION

In cerium and actinide compounds, the hybridization of the f electrons with electrons in the conduction band gives rise to various kinds of interesting phenomena. A magnetic or nonmagnetic ground state can be realized at low temperatures, depending on the strength of the hybridization. The heavy-fermion compound $CeRu_2Si_2$, which has the $ThCr_2Si_2$-type tetragonal structure, has been extensively studied since Haen *et al.* found that it shows a paramagnetic behavior down to 20 mK in 1987.[1] Because the substitution of a second element for each of the Ce, Ru, or Si atoms induces antiferromagnetic (AF) ordering,[2–4] this compound is supposed to be almost on the edge of the magnetic ordering phase. The effect of the substitution of La for Ce can be explained by a negative chemical pressure effect due to the large atomic volume of La compared with that of Ce. Substitution with a $4d$ transition metal such as Mo, Ru, or Pt onto the Ru site induces systematic changes in the critical field of the metamagnetic transition, H_M, and the Kondo temperature T_K. These observations have been explained by taking into consideration the alloying effects that are sensitive to the number of $4d$ conduction electrons.[2,5,6]

Two antiferromagnetic transitions exist in the $Ce(Ru_{1-x}Rh_x)_2Si_2$ system. One is in the high-Rh-concentration range with $0.6<x\leqslant 1.0$, where the ordered moment and magnetic ordering temperature (Néel temperature) T_N seem to increase with x.[7] The other is in the low-Rh-concentration range with $0.03\leqslant x\leqslant 0.3$, where the presence of inhomogeneous local fields was discovered by microscopic NMR and muon spin rotation (μSR) measurements.[8–10] The compound with $x=0.15$ in this range was studied systematically by neutron diffraction and electrical resistivity measurements.[11] It was shown that this kind of antiferromagnetic phases had an incommensurate sinusoidal spin modulation with wave vector $\tau=(0,0,0.420)$ and that there existed an anomaly in electrical resistance along the c axis around the magnetic transition temperature. These results suggest the presence of a partial gapping of the Fermi surface along the uniaxial c direction and that an itinerant spin-density-wave (SDW) transition has occurred in the heavy-electron band.[12–14]

In a heavy-fermion system, the ground state is determined by the competition between the intersite RKKY interaction and the intrasite Kondo interaction.[15] When the RKKY interaction is greater than the Kondo interaction, AF ordering will occur; on the contrary, the system will show a Fermi-liquid behavior. In this SDW transition range in the $Ce(Ru_{1-x}Rh_x)_2Si_2$ system, when the Rh substitution increases from low to high concentration, the Néel temperature T_N increases from zero to a maximum value of 5.5 K at $x=0.15$ and then decreases. This fact indicated that the substitution of Rh for Ru could adjust the balance between the RKKY and Kondo interactions. With increasing the substitution from $x=0.03$ to 0.15, the RKKY interaction will be improved, then weakened from $x=0.15$ to 0.30. The chemical substitution will bring two variables that influence the interactions: one is the change of the lattice parameters; the other is the change of the number of the d electrons. Which one is more important to the balance between the RKKY and Kondo interactions? To elucidate this question, it is effective to compare the effect of pressure on the SDW transition with the chemical substitution, because the application of pressure only induces the contraction of the lattice parameters; the effect of pressure on the SDW transition in the compound $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$ has been studied.[16] The results showed that application of pressure caused a decrease in T_N. At about 1 GPa the SDW transition was suppressed and the ground state was moved from the SDW state to that of a Fermi liquid (FL). This indicated that the contraction of the lattice parameter benefits the Kondo interaction in the compound $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$. However, it was unclear from this study if the effect of pressure is similar to increasing or decreasing the level of Rh substitution, because there is a maximum in the Néel temperature at the composition of $x=0.15$ for this system—any increase or decrease of the Rh concentration would lead to a decrease in the Néel temperature. In this paper we will report the effect of pressure on the compound whose composition is $Ce(Ru_{0.95}Rh_{0.05})_2Si_2$, which is located near the boundary between the SDW and FL, and

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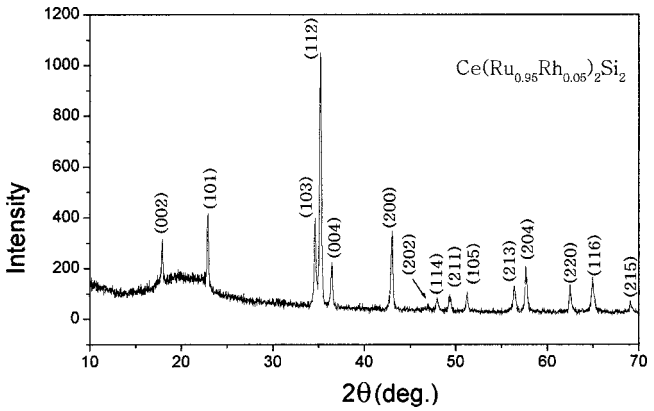


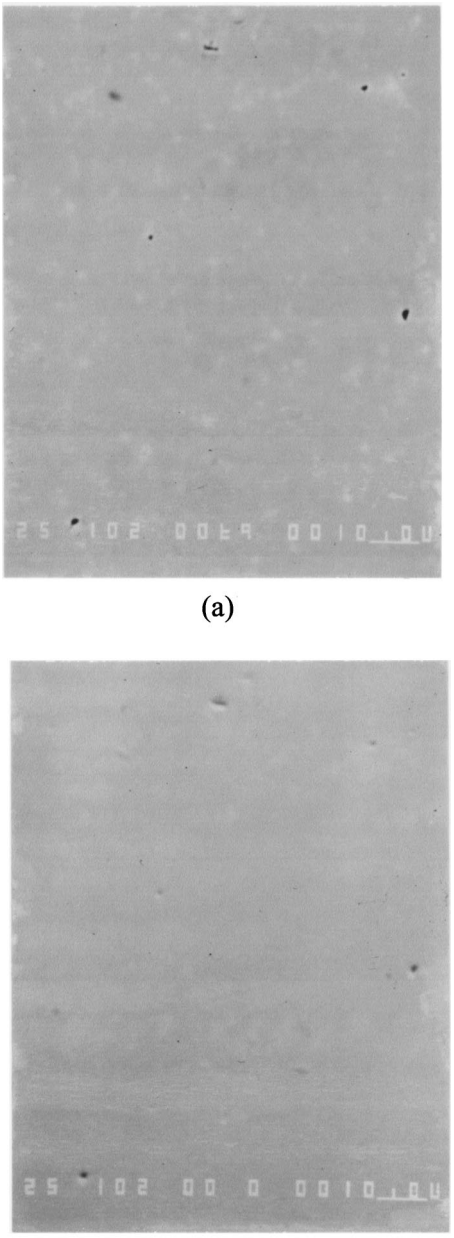
FIG. 1. θ-2θ x-ray diffraction (XRD) pattern for the powder of the $Ce(Ru_{0.95}Rh_{0.05})_2Si_2$ single crystal.

which has a SDW transition at about 2 K at ambient pressure, and compare the results with the chemical substitution.

II. SAMPLES AND EXPERIMENTS

Single crystals were used for the specific heat measurements. They were prepared by the Czochralski method in a triarc furnace in an argon atmosphere. In the x-ray powder diffraction experiment, we did not observe any other phases except the $ThCr_2Si_2$ structure (see Fig. 1). Because in the $Ce(Ru_{1-x}Rh_x)_2Si_2$ system the compound with $x=0.05$ is very close to the quantum critical point (QCP) for suppression of antiferromagnetic ordering at the ambient pressure, the quality of the sample will have a significant influence on the experimental results. So we investigated the homogeneity and composition of the sample with an electron probe microanalyzer (EPMA) (JEOL, JCXA-733). The specimen for the EPMA experiment was prepared as follows. Small pieces of the samples were fixed by a piece of P-resin plastic into a cylindrical shape. The surface of the specimen was polished roughly by sand papers (mesh Nos. 1000, 2000, and 3000) and polishing powders (mesh No. 3000), followed by a final polishing with diamond pastes (3, 1, and 0.5 μm in diameter) on the buff. The specimen was washed supersonically, and the roughness of the surface was checked by an optical microscope after each polishing. The polished specimen was mounted into a carbon sputtering apparatus, and then the specimen was coated with a thin carbon layer (about 10 nm thickness) to avoid electric charging.

The EPMA results showed that the stoichiometry of our sample is $Ce(Ru_{0.95\pm0.01}Rh_{0.049\pm0.001})_2Si_{2.04\pm0.01}$. Figure 2 is the surface photographs for the sample. We obtained the photographs by two different techniques: (a) backscattered electron composition image (COMPO), which represents the atomic weight ratio [Fig. 2(a)], and (b) backscattered electron topological image (TOPO), which represents the roughness of the surface of the sample [Fig. 2(b)]. As shown in Fig. 2, the COMPO photograph shows a very homogeneous composition except some small dark spots. The TOPO photograph also shows the same dark spots found in the COMPO photograph. Therefore, we can attribute them to holes on the surface of the sample.



(a)

(b)

FIG. 2. EPMA photographs for the $Ce(Ru_{0.95}Rh_{0.05})_2Si_2$ compound: (a) backscattered electron composition image (COMPO) and (b) backscattered electron topological image (TOPO).

The specific heat measurements were carried out using a quasiadiabatic method at various pressures up to 1.6 GPa. The pressure cell of $\Phi 12\,\mathrm{mm}(\text{external diameter})\times 42\,\mathrm{mm}$ (length) is made of a Cu-Be alloy. A Be-Cu piston and a cylinder clamp were used to generate and retain pressure which was applied at room temperature. AgCl was used as the pressure medium.

III. RESULTS AND DISCUSSION

Figure 3 shows the temperature dependence of the specific heat of a $Ce(Ru_{0.95}Rh_{0.05})_2Si_2$ single crystal at various

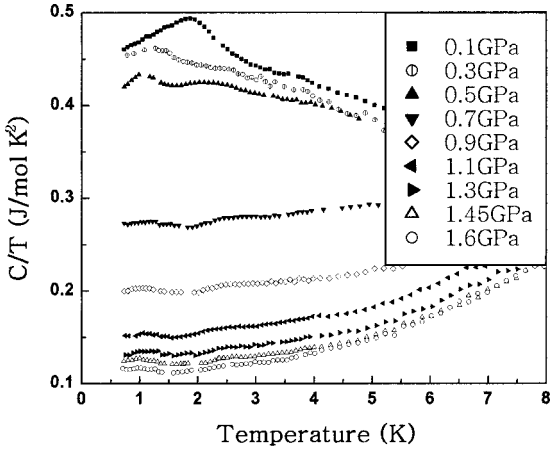


FIG. 3. Temperature dependence of the specific heat C for $Ce(Ru_{0.95}Rh_{0.05})_2Si_2$ compound at various pressures (plotted as C/T vs T).

pressures. The specific heat C, divided by the absolute temperature T, is plotted as a function of T. With application of pressure there is a decrease in the factor C/T at all temperatures. This suggests that the density of states at the Fermi surface decreases with increasing pressure. In the plot of C/T vs T at 0.1 GPa, there is a peak at about 1.8 K, which we interpreted as being a result of the partial gapping of the Fermi surface. The position of this peak corresponds to the magnetic ordering temperature,[17] below which the compound was in the SDW state. A similar anomaly at about 2 K was observed at the ambient pressure in this compound. Under increasing pressure the position of the peak moves to a lower temperature, which indicates that the SDW transition temperature is decreasing. When the pressure exceeds 0.7 GPa, the value of C/T changes very little with decreasing temperature. This fact suggests that the ground state of the $Ce(Ru_{0.95}Rh_{0.05})_2Si_2$ compound is a Fermi liquid under high pressure and that the long-range magnetic ordering is suppressed.

The pressure dependence of the Néel temperature T_N is shown in Fig. 4. The value of T_N at ambient pressure was

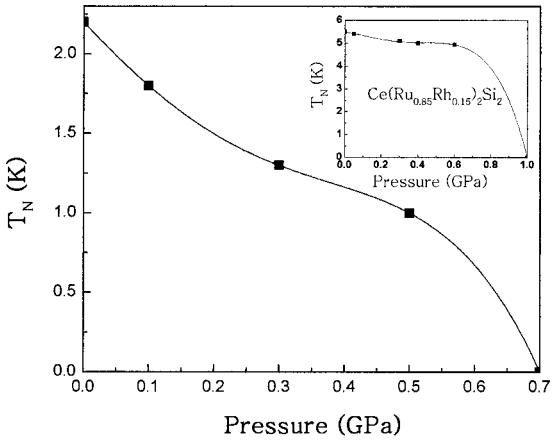


FIG. 4. Pressure dependence of the Néel temperature T_N in $Ce(Ru_{0.95}Rh_{0.05})_2Si_2$. The inset shows the pressure dependence of T_N in $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$.

obtained by measuring the susceptibility and specific heat of the material. When the applied pressure is lower than 0.5 GPa, T_N shifts slightly to a lower temperature. When the pressure is greater than 0.5 GPa, T_N decreases quite drastically to zero. It suggests that a critical pressure exists at which the magnetic ordering begins to collapse. The inset in Fig. 4 shows the pressure dependence of T_N in the compound $Ce(Ru_{0.85}Rh_{0.15})_2Si_2$, and it shows a similar behavior. From these two observations it is suggested that the Néel temperature (T_N) decrease with application of increasing pressure in the SDW phase region when the degree of Rh substitution is lower than 15%. In this region T_N also decreases with decreasing Rh concentration. Therefore, it is proposed that the effect of pressure is similar to decreasing the degree of substitution of Rh for Ru in the SDW state.

The antiferromagnetic correlation caused by the substitution of La for Ce in the compound $CeRu_2Si_2$ can be explained by a simple volume effect, because La has a larger atomic size than Ce.[2] The substitution of Rh for Ru decreases the unit-cell volume only slightly in this compound, so the correlation that we observed in the $Ce(Ru_{1-x}Rh_x)_2Si_2$ system could not be attributed to the volume effect, and it was explained as being due to the alloying effects. However, we must emphasize that the AF ordering in this system is anisotropic. Susceptibility and electrical resistance measurements indicated that the antiferromagnetic correlation only occurred along the C axis and that the energy gap partially opened at the Fermi surface perpendicular to the C^* axis.[11] Therefore, it is more important to examine the contribution of the change in the lattice parameter along the c axis and the distance between Ce and Ru/Rh atoms (d) than the volume change. Figure 5 shows the measured and calculated values of a (lattice parameter of a axis), c (lattice parameter of the c axis), d, and v (volume of the unit cell) for various values of the Rh concentration in the $Ce(Ru_{1-x}Rh_x)_2Si_2$ system. It is clear from these results that, although the volume of the unit cell and the lattice parameter a decrease when we increase the level of the substitution of Rh for Ru, the lattice parameter c and the value of d are increasing. Therefore, the application of pressure and a decrease in the concentration of Rh can induce the same effect, which is a reduction in both the c parameter and the distance between the Ru(Rh) and Ce. This should play an important role in the SDW transition.

Figure 6 shows the pressure dependence of the Sommerfeld coefficient γ in the compound $Ce(Ru_{0.95}Rh_{0.05})_2Si_2$. The value of γ decreases with increasing pressure. But the rate of decrease is strongly pressure dependent. At high pressure the compound is in the FL state and the value of γ decreases slowly. At low pressure the compound is in the SDW state and the value of γ decreases slowly. But at the critical pressure at which the SDW transition is suppressed, the value of γ decreases drastically. In the SDW state, the Sommerfeld coefficient γ consists of two parts: γ_1 and γ_2,[11] where γ_1 originates from the remaining ungapped part of the Fermi surface and it is proportional to the reverse of the Kondo temperature T_K and decreases with increasing pressure; on the other hand, γ_2 comes from the gapped magnetic region and it increases with increasing pressure.[17] The compensation for the decrease in γ_1 and the increase in γ_2 with in-

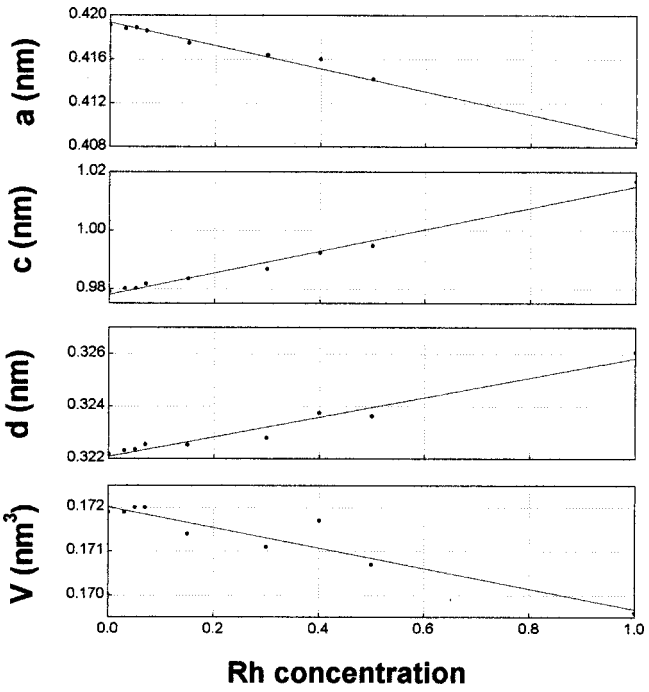


FIG. 5. Dependence of the lattice parameters of the *a* axis, *c* axis, *d* (the distance between the Ce atom and Ru or Rh atom), and *v*, the volume of the unit cell of the $Ce(Ru_{1-x}Rh_x)_2Si_2$ system on the Rh concentration.

creasing pressure leads to the phenomenon that the Sommerfeld coefficient decreases slowly. When the SDW state collapses at high pressure, the contribution of the gapped magnetic part disappears and the Sommerfeld coefficient decreases drastically.

In the $Ce(Ru_{1-x}Rh_x)_2Si_2$ system, when it is near the critical boundary that separates the magnetic and nonmagnetic phases in the chemical substitution diagram (concentration versus temperature), the value of γ is about 480 mJ/mol K^2 (for $x=0.03$ at ambient pressure).[18] At the same critical boundary in the pressure-temperature diagram, γ is just about 270 mJ/mol K^2 (when $x=0.05$ and $P=0.7$ GPa). We noticed that there was a large difference. This indicated that, at the same SDW-FL boundary, the densities of states at the Fermi surface under these two conditions are different. The application of pressure just brings about a change in the distance between the atoms, and the substitution of Rh for Ru not only causes a change in the distance, but also a change in the number of *d* electrons.

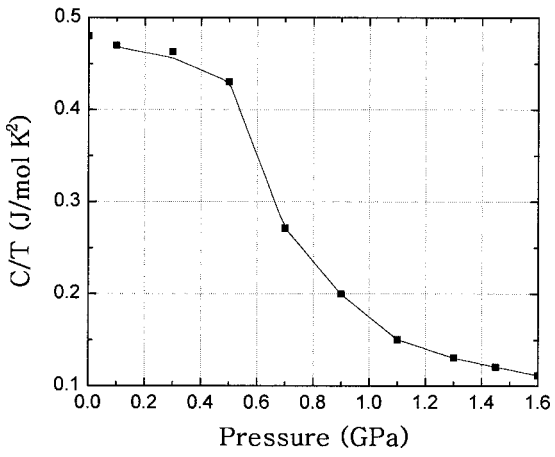


FIG. 6. Pressure dependence of the Sommerfeld coefficient γ for $Ce(Ru_{0.95}Rh_{0.05})_2Si_2$ compound.

IV. SUMMARY

In this paper, we have measured the specific heat of a single crystal of the heavy-fermion compound $Ce(Ru_{0.95}Rh_{0.05})_2Si_2$ between 0.7 and 8 K at pressures 0.1–1.6 GPa. The application of pressure decreased the SDW transition temperature first and finally suppressed the long-range magnetic ordering, which suggests that the Kondo compensation of the local moments should dominate the RKKY interactions and suppress the development of a long-range magnetic ordering at 0.7 GPa. The application of pressure and a decrease in the chemical substitution of Rh for Ru all lead to a decrease of the *c* parameter and the distance between the Ru(Rh) and Ce, which plays an important role in the SDW transition. It was also proposed that the effect of pressure is similar to that of decreasing the degree of the substitution of Rh for Ru in the SDW state.

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