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Anisotropic suppression of the energy gap in CeNiSn by high magnetic fields

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The formation of the pseudogap in CeNiSn has been investigated by means of resistivity and specific-heat measurements in high magnetic fields. When the field is raised from 0 to 12 T along the orthorhombic a axis, the semiconductorlike behavior in the resistivity below 6 K disappears and the linear electronic term in the specific heat increases from 57 to 100 mJ/K^{2}mol. However, no significant effect was observed for fields along the b and c axes up to 15 T. These results suggest an anisotropic suppression of the pseudogap, which can be interpreted as a result of strong spin polarization of the renormalized narrow band.

Cerium-based Kondo-lattice systems exhibit unusual physical phenomena such as valence fluctuation and heavy-fermion behavior.[1] The low-temperature properties of the latter are dominated by a renormalized narrow band formed by the hybridization and a Kondo-type interaction between the $4f$-electron and the conduction-electron states. Most Kondo-lattice systems have a metallic ground state, either magnetically ordered or Pauli paramagnetic. Recently, a different type of ground state with an energy gap at the Fermi level has been found in CeNiSn (Refs. 2-4) and subsequently in $Ce_3Bi_4Pt_3$ (Ref. 5) and CeRhSb.[6] They are considered as good candidates for Kondo-lattice systems with a pronounced coherence pseudogap. The existence of a weak pseudogap was previously proposed to explain the maximum in C/T observed in the specific heat of the heavy-fermion systems $CeAl_3$ and $CeCu_2Si_2$.[7-9]

CeNiSn crystallizes in the orthorhombic ϵ-TiNiSi-type structure,[10] in which the Ce atoms form a zigzag chain along the a axis. In a previous paper,[4] Takabatake *et al.* reported transport, magnetic, and thermal properties of single-crystalline CeNiSn samples above 1.5 K. At temperatures above 50 K, the transport properties are dominated by the incoherent and strongly anisotropic Kondo scattering in the presence of a crystalline field. The temperature dependence of the magnetic susceptibility χ even for the easy a axis is weak compared to the Curie-Weiss behavior expected for trivalent Ce ions. This compound was therefore classified as a valence-fluctuating system. At 12 K, both $\chi(T)$ and resistivity $\rho(T)$ exhibit pronounced peaks only along the a axis, which is considered as an indication of the development of coherence among periodically placed Kondo impurities. The development of coherence has been confirmed by the huge positive peak near 9 K in the Hall coefficient.[11] On the other hand, $\chi(T)$ of $Ce_3Bi_4Pt_3$ and CeRhSb has a broad maximum near 100 K,[5,6] and thus these systems are thought to be closer to the valence-fluctuating regime than CeNiSn.

At temperatures below 6 K, both the resistivity and thermoelectric power of CeNiSn drastically increase like in a semiconductor. The magnetic specific heat divided by temperature C_m/T has a peak of 0.19 J/K^{2}mol near 6.7 K and then sharply diminishes on cooling. The peaking in C_m/T and $\chi(T)$ agrees with the prediction for the development of a coherence pseudogap in the Kondo lattice below a coherence temperature T_c.[9] From these results, T_c in CeNiSn is estimated to be approximately 10 K. Furthermore, the nuclear-spin-lattice relaxation rate of ^{119}Sn in this compound is proportional to T^3 between 0.4 and 1.2 K.[12] This result has led to a model that the density of quasiparticle states has a V-shaped structure. Such a ground state in CeNiSn is expected to be strongly affected by sufficiently high magnetic fields. In fact, disappearance of the semiconductorlike behavior in the resistivity was found in fields higher than 20 T for a polycrystalline sample.[13] In this paper, we present results of both the magnetoresistance and low-temperature specific heat of single-crystalline samples in high magnetic fields.

Single crystals were grown by a floating-zone method using a single-ellipsoidal infrared heating furnace (Nichiden Machinery Ltd.). We obtained crystals about 7 mm in diameter and 20 mm in length elongating along the b axis. From metallographic examination and electron-probe microanalysis, impurity phases of $CeNi_2Sn_2$ and cerium oxides were detected at the end tail and on the surface of the as-grown crystal. The central part of the crystal was shaped by means of spark erosion for resistivity and specific-heat measurements. The magnetoresistance was measured by four-probe dc techniques down to 1.4 K in magnetic fields up to 15 T, which were produced by a

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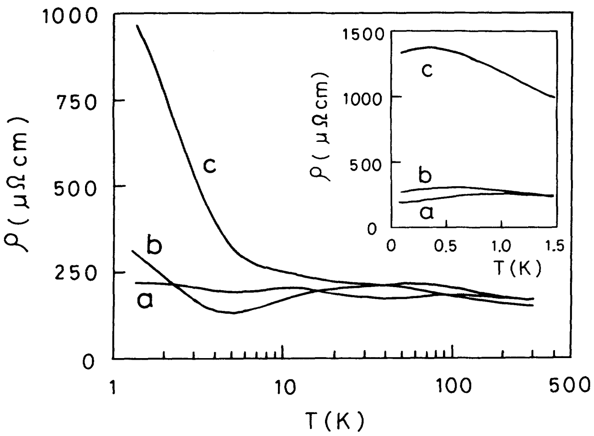

FIG. 1. Electrical resistivity vs temperature for CeNiSn along the three principal axes.

superconducting or a water-cooled magnet. The specific-heat measurements were performed between 0.06 and 0.8 K in magnetic fields up to 12 T using a relaxation method calorimetry.

Figure 1 shows the results of $\rho(T)$ along the three principal axes. Below about 6 K, $\rho_b(T)$ and $\rho_c(T)$ drastically increase due to the gap formation, whereas $\rho_a(T)$ tends to be saturated at the lowest value among the three. The strong anisotropy suggests an anisotropic gapping of the density of states on the Fermi surface. In order to check whether ρ_c and ρ_b are saturated or not, the temperature range was extended to 0.1 K using a dilution refrigerator. As shown in the inset of Fig. 1, $\rho_c(T)$ and $\rho_b(T)$ pass weak maxima at 0.4 and 0.6 K, respectively, and decrease with further decreasing temperature. The decrease in resistivity cannot be understood assuming a uniform gapping over the Fermi surface. Instead, it seems to be due to an incomplete gapping.

We measured the magnetoresistance $\Delta\rho(H) = \rho(H) - \rho(0)$ at 1.4 K and present the normalized data $\Delta\rho(H)/\rho(0)$ in Figs. 2(a) and 2(b). Comparing the two curves for **I**$\parallel b$ in Fig. 2(a), we notice that the initial slope is negative for **H**$\parallel a$ but positive for **H**$\parallel c$. At a high field of 15 T, the negative magnetoresistance for **H**$\parallel a$ attains -88%, being much larger than for **H**$\parallel b$ and **H**$\parallel c$. Thus, the energy gap is most sensitive to the application of a magnetic field along the easy a axis. Therefore, the a axis was chosen as the field direction and the dependence of magnetoresistance on the direction of the current was further examined. As can be seen in Fig. 2(b), the field dependence of $\Delta\rho(H)/\rho(0)$ for **I**$\parallel c$ is similar to that for **I**$\parallel b$, whereas $\Delta\rho(H)$ for **I**$\parallel a$ is almost constant up to 4 T and then gradually decreases with increasing field with an inflection near $H = 13$ T. These results suggest a strong anisotropic scattering mechanism under magnetic fields. It should also be noted that recent magnetization measurements have revealed a weak metamagneticlike transition near 13 T only when the field is applied along the a axis.[14] These anomalies near 13 T may reflect the disappearance of the pseudogap.

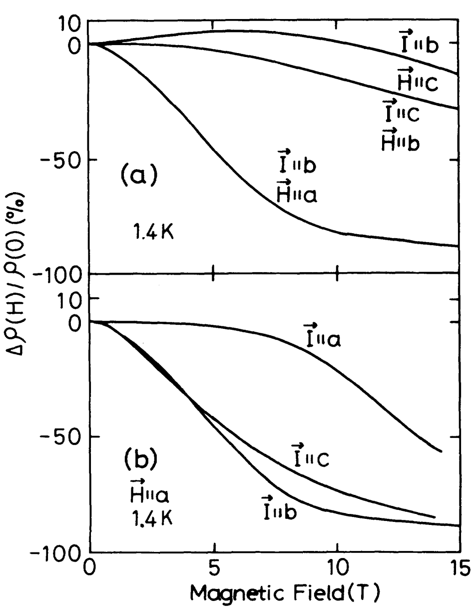

FIG. 2. Magnetoresistance of CeNiSn at 1.4 K for (a) magnetic fields along the three principal axes and (b) magnetic field along the a axis and electrical current along the three principal axes.

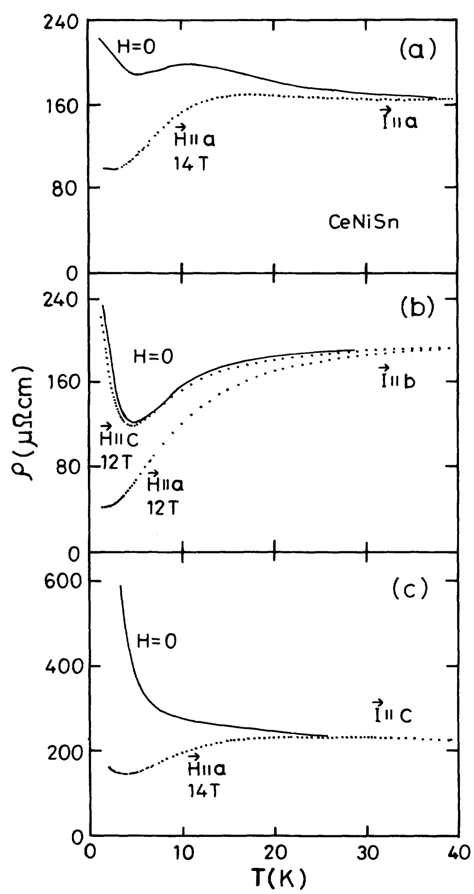

FIG. 3. Electrical resistivity vs temperature for CeNiSn at 0, 12, and 14 T for electrical currents along the three principal axes.

Figures 3(a)–3(c) show the temperature dependence of the resistivity along the three principal axes in fields of 0, 12, and 14 T parallel to the a or c axis. For **H**$\parallel a$, a negative magnetoresistance gradually increases with decreasing temperature below 30 K for the three current directions. Such a negative magnetoresistance is usually observed for Kondo-lattice systems well above the coherence temperature.[15] However, the large negative $\Delta\rho(H)$ below 6 K should be a result of the suppression of the gap by magnetic fields.

In Fig. 3(a), the a-axis resistivity at $H=0$ exhibits a maximum near 11.4 K, which has been ascribed to the development of coherence among the Kondo states at the Ce sites. In a field of 14 T parallel to the a axis, this maximum is almost smeared out and the upturn at lower temperatures is strongly suppressed. As shown in Fig. 3(b), a drastic effect occurs in the resistivity for **I**$\parallel b$ and **H**$\parallel a$, which indicates metallic behavior and is in contrast to the semiconductorlike behavior for **H**$\parallel c$. Furthermore, at temperatures below 4 K, the resistivity for **H**$\parallel a$ at 12 T obeys a T^2 dependence with a coefficient of 1.0 $\mu\Omega\,\mathrm{cm/K^2}$ and a residual resistivity of 39 $\mu\Omega\,\mathrm{cm}$. The size of this coefficient is typical for a moderately heavy-fermion system.[1] On the other hand, as shown in Fig. 3(c), the resistivity along the c axis at 14 T still exhibits an upturn below 3.6 K.

The specific heat of the single crystalline CeNiSn was measured in zero fields for $0.06 \leq T \leq 0.8$ K and $1.5 \leq T \leq 80$ K. The result in the high-temperature range was qualitatively similar to that of the polycrystalline sample[4] except that the anomaly near 2.3 K is much smaller in the single-crystalline sample. The low-temperature results at fields of 0, 4, 8, and 12 T parallel to the a and c axes are shown in Figs. 4(a) and 4(b), respectively. At $H=0$ T, C/T is almost proportional to T between 0.3 and 0.8 K and linear extrapolation yields a value of 57 $\mathrm{mJ/K^2 mol}$ as the γ value. We suppose that this size for γ is too large to be ascribed to the contribution from impurity phases if existing. Rather, it may be the contribution from the residual density of states at E_F in the pseudogap, as inferred from the saturation of resistivity below 1 K. Another interesting feature of the zero-field data in Fig. 4(b) is the upturn below 0.25 K. It should be noted that recent NMR study has revealed the appearance of hyperfine magnetic field at the ^{119}Sn nuclei in CeNiSn below 0.2 K.[16] This observation together with a sharp decrease in the NMR intensity has indicated some kind of magnetic order at around 0.2 K.[16] We found that the zero-field data of C/T can be fitted assuming the form $C/T=\gamma+aT+\delta/T^3$. Since the last term is anticipated for the nuclear contribution in a magnetically ordered state, the upturn in C/T at zero field may partly originate from the magnetic ordering. When magnetic field is raised to 12 T, the value of C/T is strongly enhanced for **H**$\parallel a$ over the measure temperature range, whereas it is almost unchanged for **H**$\parallel c$. The field dependence of C/T was further studied for **H**$\parallel a$ at 0.15, 0.42, and 0.76 K. As shown in Fig. 5, the values of C/T stay constant for **H** <4 T and then increase monotonically from about 70 to 125 $\mathrm{mJ/K^2 mol}$. This field dependence corresponds to the magnetoresistance curve for **H**$\parallel a$ and **I**$\parallel a$ in Fig. 2(b), and the largely enhanced γ value is consistent with the heavy-fermion-like behavior in $\rho_b(T)$ at $H=12$ T in Fig. 3(b). These results support the idea that the density of states in the minimum of the pseudogap is increased by application of magnetic field along the easy axis of magnetization.

In the present study, we have performed measurements of magnetoresistance and specific heat of single crystalline CeNiSn in high magnetic fields. The magnetoresistance is highly anisotropic with respect to the directions of both the magnetic field and the electrical current. We found that the semiconductorlike behavior in the resistivity disappears and the γ value doubles when the field parallel to the a axis is raised to 12 T. For the field direction parallel to the b or c axis, no drastic effect was observed. Considering the fact that a weak metamagneticlike transition was observed at 13 T for **H**$\parallel a$,[14] we infer the disap-

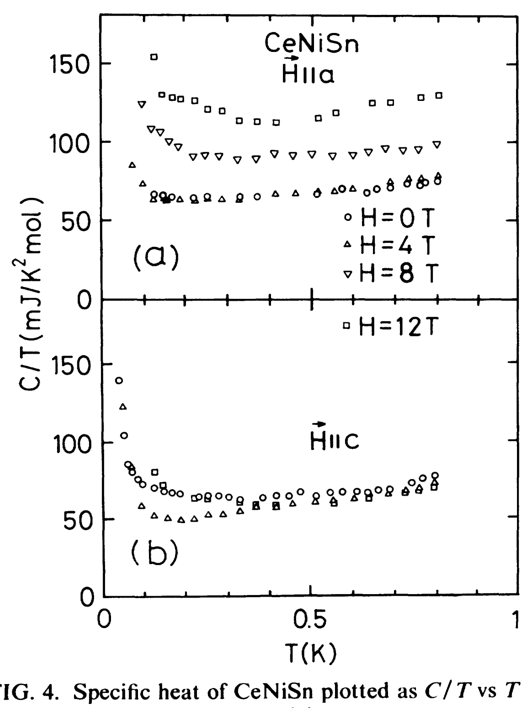

FIG. 4. Specific heat of CeNiSn plotted as C/T vs T in magnetic fields for (a) **H**$\parallel a$ axis and (b) **H**$\parallel c$ axis.

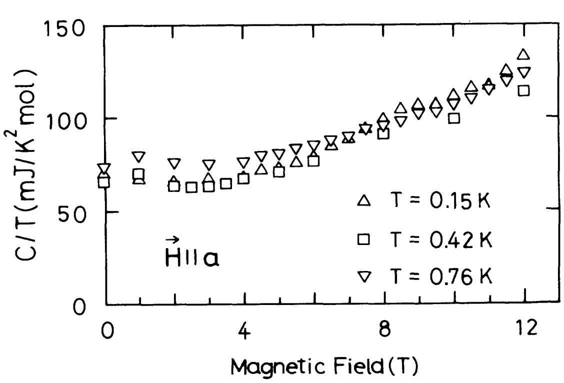

FIG. 5. Field variation of C/T of CeNiSn for **H**$\parallel a$ axis.

pearance of the pseudogap near 13 T only when the field is applied along the easy a axis. In this anisotropic suppression of the pseudogap, a strong spin polarization of the renormalized band should play an important role. It is therefore likely that anisotropic effects arising from magnetic correlations and from the hybridization of $4f$ states with conduction electron states play important roles in the formation of the energy gap in CeNiSn. Theoretical studies taking into account such anisotropic effects are highly anticipated for better understanding of the mechanism of the gap formation. Further experimental work is also needed to clarify the origin of the upturn in C/T and the decrease in the resistivity below 0.5 K.

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