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High-Magnetic-Field Studies of the Kondo Semiconductor CeNiSn*

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Synopsis

The Kondo-lattice compound CeNiSn behaves as a narrow-gap semiconductor at low temperatures below 7 K. The effect of magnetic field on the gapped state of this compound has been studied by the measurements of magnetization, magnetoresistance and specific heat on a single-crystalline sample. A weak metamagnetic transition and very large negative magnetoresiostance are observed only when a magnetic field higher than 13 T is applied along the *a* axis of the orthorhombic structure. These results show that the pseudogap is suppressed anisotropically by the magnetic field. The collapse of the gap is accompanied with the evolution of a polarized heavy-fermion state.

I. Introduction

Kondo-lattice compounds are intermetallic compounds possessing an ordered lattice of a rare earth atom with unstable 4f electrons such as Ce and Yb. The valence fluctuation and heavy-fermion behavior of these compounds have been the subject of intensive studies.1) 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 YbB_{12},2,3) CeNiSn,4-7) CeRhSb,8) and $Ce_3Bi_4Pt_3$.9) The gap energy is estimated to be 56, 6, 8 and 70 K, respectively, from the semiconducting behavior of the resistivity. The energy scale of the gap corresponds to the magnetic energy of tens of Tesla, which is now accessible with high field techniques. Thus we are able to study the magnetic response of the gapped state. In fact, the previous measurements of magnetoresistance on a polycrystalline sample of CeNiSn showed that the semiconducting gap is closed at around 20 T.10) The larger energy gap in YbB_{12} is also destructible by the application of magnetic field of 50 T.3)

Our recent investigation of single-crystalline CeNiSn has revealed that the magnetic and transport properties in zero field are strongly anisotropic.5-7) This behavior

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reflects strong hybridization of the 4f states with the 5p and 3d states of the neighboring Sn and Ni atoms in the orthorhombic structure. Therefore, we expect anisotropic effect of magnetic field on the the magnetic and transport properties. Keeping this in mind, we have performed the measurements of magnetization, magnetoresistance and specific heat on single-crystalline samples of CeNiSn. The results will be discussed in relation to the model of hybridization gap in the Kondo-lattice system.[11,12]

II. Experimental

Single crystals were grown by a floating-zone method using a single-ellipsoidal infrared mirror furnace. The crystals obtained are about 7 mm in diameter and 20 mm in length elongating along the *b* axis. The field dependence of magnetization was measured in pulsed magnetic fields up to 36 T at the High Magnetic Field Laboratory of the Research Center for Extreme Materials, Osaka University. The eddy current effect was sufficiently reduced by using a long-pulsed field of about 20 ms and samples with cross-sections less than 0.3 mm^2. The magnetoresistance was measured by four-probe dc technique in magnetic fields up to 15 T, which were produced by a water-cooled magnet at the High Field Laboratory for Superconducting Materials of Institute for Materials Research, Tohoku University. The specific-heat measurements were performed using a relaxation-type calorimeter, which was mounted into a dilution refrigerator equipped with a superconducting magnet at Technische Hochschule Darmstadt.

III. Result and Discussion

3.1 Magnetization

The field dependence of magnetization M(H) of single crystal CeNiSn at 1.3 K is represented in Fig. 1 from Ref. 13. The M(H) curve only along the *a* axis exhibits a weak metamagnetic-like transition near 13 T. It is more clearly seen in the derivative susceptibility dM/dH vs H. However, the increase in M associated with the transition is much smaller than that found in nonmagnetic heavy-fermion compounds like $CeRu_2Si_2$.[14] This fact suggests that the weak transition in CeNiSn is not due to the suppression of antiferromagnetic intersite interactions as found in $CeRu_2Si_2$ but due to the collapse of the pseudogap. Above 20 T, M(H) along the *a* axis increases linearly with increasing field and attains a value of 0.3 μ_B/Ce at 36 T. The size of the magnetization is only one fourth of that found in the isostructural, antiferromagnetic compound CePtSn, 1.2 μ_B/Ce.[15] The small and linearly increasing moment in CeNiSn can be interpreted as a result of strong Kondo-type interaction persisting even after the pseudogap has collapsed. Assuming the effective moment of 0.3 μ_B/Ce, the magnetic energy at the transition field of 13 T corresponds to the thermal energy of 2.6 K, which is comparable to the gap energy estimated from the *a*-axis resistivity.[5]

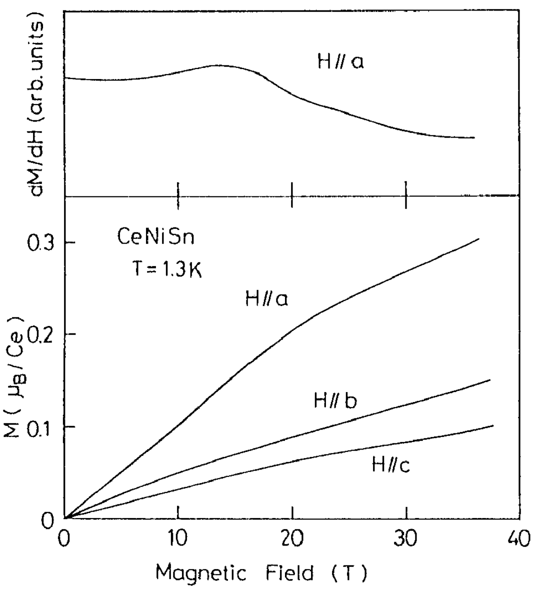

Fig. 1. Magnetization of CeNiSn along the three principal axes at 1.3 K (after ref. 13).

3.2 Magnetoresistance

The magnetoresistance of single crystalline samples show very strong anisotropy. In Fig. 2, we present the normalized magnetoresistance $\Delta\rho(H)/\rho(0)$ at 1.4 K, where $\Delta\rho(H) = \rho(H) - \rho(0)$.[13] At H = 15 T, the negative magnetoresistance for H//*a* attains -88%, which is much larger than for H//*b* and H//*c*. Thus, the energy gap is most sensitive to the magnetic field applied along the easy *a* axis. For H//*a*, the dependence of magnetoresistance on the current direction was further examined. In Fig. 2(b), the field dependence of $\Delta\rho(H)/\rho(0)$ for I//*c* is similar to that for I//*b*, whereas $\Delta\rho(H)/\rho(0)$ for I//*a* is almost constant up to 4 T and then gradually decreases with increasing field. These results suggest a strong anisotropic scattering mechanism under magnetic fields.

Shown in Fig. 3 is the temperature dependence of the resistivity along the three principal axes in fields of 0, 12 and 14 T.[16] At H = 0 T, $\rho_a(T)$ exhibits a maximum near 11.4 K, which has been ascribed to the development of antiferromagnetic correlations.[5] In a field of 14 T parallel to the *a* axis, this maximum is almost smeared out. The strong suppression of the upturn below 6 K is a result of the gap suppression by magnetic fields. A drastic effect occurs in the resistivity for the configuration I//*b* and H//*a*, which indicates metallic behavior and is in contrast to the semiconductor-like behavior for H//*c*. Furthermore, at temperatures below 4 K, it obeys a T^2 dependence with a coefficient of 1.0 $\mu\Omega cm/K^2$ and a residual resistivity of 39 $\mu\Omega cm$. The size of this coefficient is typical for moderately heavy fermion systems.

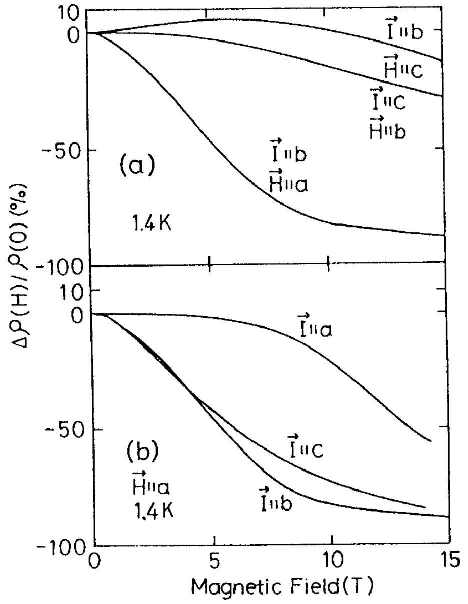

Fig. 2. Magnetoresistance of CeNiSn at 1.4 K (after ref. 13).

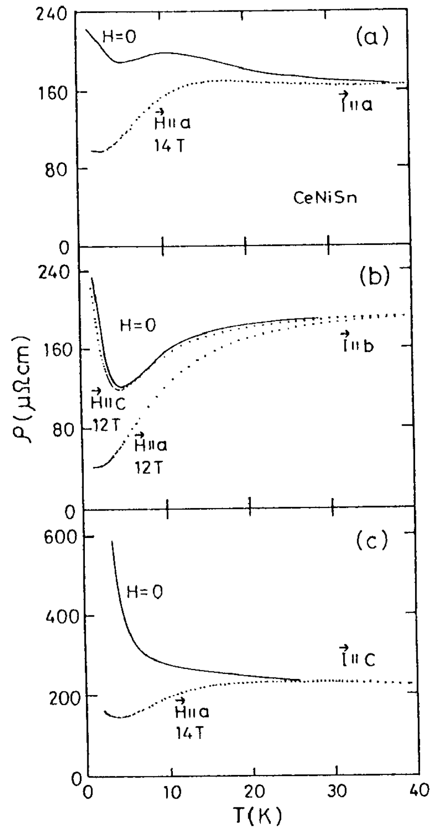

Fig. 3. Electrical resistivity vs temperature for CeNiSn in external fields 0, 12 and 14 T for electrical currents along the three principal axes (after ref. 16).

3.3 Specific Heat in Magnetic Fields

The suppression of the energy gap by magnetic field was further studied by specific-heat measurements.6) Temperature variations of the specific heat of CeNiSn in magnetic fields 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 the linear extrapolation to T= 0 K yields a γ value of 57 mJ/K^{2}mol. This size of γ value seems to be too large to be ascribed to the contribution from impurity phases.Rather, it may be the contribution from the residual density of states at E_F in the pseudogap, as inferred from the saturation of the resistivity below 1 K.6) The origin of the upturn in C/T below 0.2 K is not clear yet.

When magnetic field is raised to 12 T, the value of C/T is strongly enhanced for H//*a*, whereas it is almost unchanged for H//*c*. For H//*a*, the field dependence of C/T was measured at 0.15, 0.42 and 0.76 K. As shown in Fig. 5, the values of C/T at these

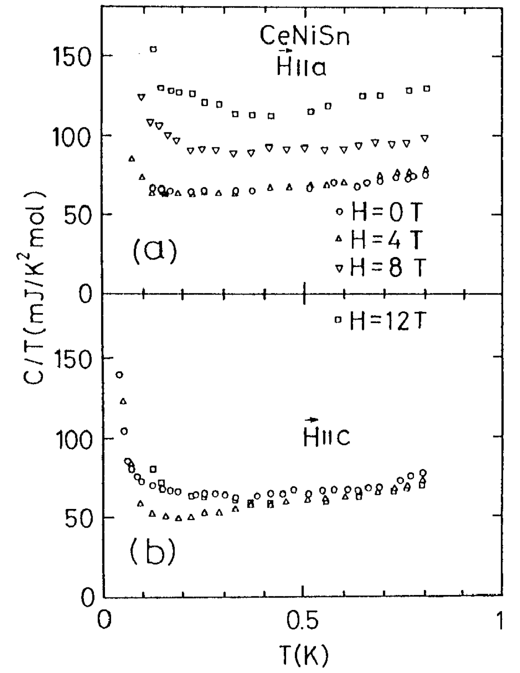

Fig. 4. Specific heat of CeNiSn plotted as C/T vs T in magnetic fields for (a) H//*a* and (b) H//*c* (after ref. 6).

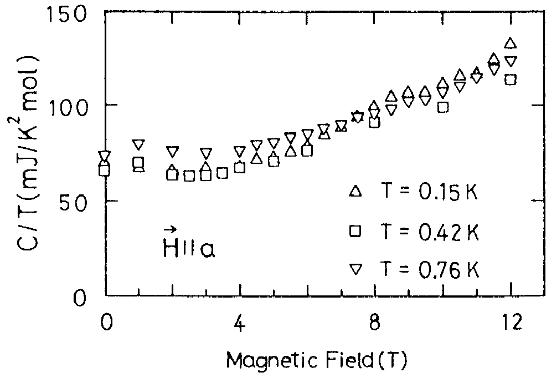

Fig. 5. Field variation of C/T of CeNiSn for H//*a* axis at low temperatures (after ref. 6).

temperatures stay constant for H < 4 T and then increase monotonically from about 70 to 125 mJ/K^{2}mol. This large enhancement is consistent with the heavy-fermion behavior in $\rho_b(T)$ at 12 T in Fig. 3(b). These results support the idea that the density of states in the minimum of the V-shaped pseudogap is increased by application of magnetic field along the easy axis of magnetization. In this anisotropic suppression of the pseudogap, a strong spin polarization of the renormalized band should play an important role. According to the model of the hybridization gap,[11,12)] the collapse of the pseudogap is attributed to the magnetic excitation of an electron from a state in the lower hybridized band to a state in the upper hybridized band with spin flipping.

IV. Summary

The effect of magnetic field on the gapped state of CeNiSn has been studied by the measurements of magnetization, magnetoresistance and specific heat on a single-crystalline sample. A weak metamagnetic transition and very large negative magnetoresiostance are observed only when a magnetic field higher than 13 T is applied along the *a* axis of the orthorhombic structure. These results indicate an anisotropic suppression of the pseudogap, which is interpreted as a result of strong spin polarization of the renormalized narrow band. The collapse of the gap is accompanied with the evolution of a polarized heavy-fermion state.

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