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Hall Resistivity and Magnetoresistance of a Kondo Semimetal CeNiSn in High Magnetic Fields

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The Hall resistivity and magnetoresistance of single-crystalline CeNiSn have been measured in magnetic fields up to 15 T. When the field is applied along the easy magnetization a axis, the Hall resistivity at 1.5 K changes sign from negative to positive at 5 T and a negative magnetoresistance appears for all the current directions. By contrast, a large positive magnetoresistance appears for $H /\!/ c$ in the transverse configurations at temperatures only below 5 K. These results suggest that the V-shaped density of states are overlapped by the Zeeman effect when the field $H /\!/ a$ exceeds 5 T.

KEYWORDS: Hall effect, magnetoresistance, CeNiSn, Kondo lattice

§ 1. Introduction

The Kondo-lattice compound CeNiSn has received much attention because an anisotropic energy gap of the order of 10 K opens at the Fermi level.[1] From the measurements of specific heat,[1] Hall coefficient,[2] nuclear magnetic resonance[3] and tunneling spectroscopy,[4] it has been revealed that the density of states at the Fermi level strongly decreases below 10 K. However, our recent study using high-quality single crystals has shown that the semiconducting increase in the resistivity is strongly suppressed for purer crystals.[5-7] In fact, the resistivity along the orthorhombic a axis decreases down to 25 μΩcm as the temperature is decreased to 0.4 K, like in a metallic Kondo-lattice compound. This result indicates that the ground state in CeNiSn is not insulating but semimetallic with a pseudo energy gap. A theory for the formation of a pseudogap which is closed along the a axis has been proposed by Ikeda and Miyake.[8] They have proposed that the gap closing originates in the fact that hybridization of a conduction band with the crystal-field ground state $|5/2,\pm3/2\rangle$ for the $4f$ electron vanishes at the zone boundary along the a axis.

The pseudogap in CeNiSn is unstable against high magnetic field when it is applied along the a axis, which is the easy axis of magnetization.[1] Specific-heat measurements in magnetic fields indicated that the density of states at the Fermi level recovers gradually when $H//a$ exceeds 6 T.[1] A weak metamagnetic behavior appears around 14 T in the magnetization curve at 1.3 K,[1] and the longitudinal magnetoresistance (MR) for $H//a$ reaches - 70% at 30 T.[9] These effects of magnetic field have been attributed to the Zeeman splitting of the V-shaped density of states. On the other hand, a very large positive MR appears for $H//c$ and $H//b$ in the transverse configurations.[6,7,10] This fact has led Inada *et al.* to conjecture that the ground state of CeNiSn is a compensated metal with equal carrier concentrations for electrons and holes, both of which have closed Fermi surfaces.[10]

In order to study further the field-induced collapse of the pseudogap in CeNiSn, we have measured the field dependence of Hall resistivity $\rho_{xy}(H)$ and the temperature dependence of electrical resistivity $\rho(T)$ in fields up to 15 T.

§ 2. Experimental

Single crystals of CeNiSn #8 and #9 were grown by a Czochralski method using a radio-frequency furnace. The details of crystal growth and of purification process have been described in Refs. 5 and 6. In the as-grown crystal #9, impurities of $Ce_2Ni_3Sn_2$ less than 0.2% were detected by metallographic examination and electron-probe microanalysis. The impurity is decreased below 0.1% in crystal #8 by the purification using the technique of solid-state electrotransport.

Measurements of $\rho(T)$ in magnetic fields were performed by a four-probe DC technique at the Institute for Solid State Physics, University of Tokyo. On the other hand, measurements of Hall voltage V_y were done by an AC method using lock-in amplifiers at temperatures between 1.5 and 10 K. The Hall resistivity ρ_{xy} was determined by the relation $\rho_{xy} = V_y d / J_x$ where d is the sample thickness and J_x is the electrical current. The value of V_y was calculated from two data for opposite field directions, $V_y = \{V_y(+H) - V_y(-H)\} / 2$. Magnetic fields up to 15 T were produced by a superconducting magnet at the High Field Laboratory for Superconducting Materials of Institute for Material Research, Tohoku University.

§ 3. Results and Discussion

Figure 1 shows the temperature dependence of electrical resistivity $\rho(T)$ for the purified crystal CeNiSn #8 along the three principal axes. The data down to 4 K are in good agreement with that for crystal #5 reported in Ref. 6. Whereas $\rho_c(T)$ of #5 increased below 2 K, both $\rho_a(T)$ and $\rho_c(T)$ for #8 continue to decrease down to 0.4 K, suggesting better quality for crystal #8. Both $\rho_a(T)$ and $\rho_c(T)$ pass through a maximum at around 14 K. Below this temperature, development of antiferromagnetic spin correlations has been found from neutron scattering experiments.[10,11]

As shown in Fig. 2, the maximum is smeared out when a field of 14.5 T is applied along the *a* axis, leading to a large negative MR for all the current directions. The negative MR may originate in two contributions; suppression of antiferromagnetic spin fluctuations and suppression of the pseudogap by the magnetic field. Above 5 K, suppression of antiferromagnetic spin fluctuations may be dominant. At low temperatures, however, the density of states at the Fermi level recovers by the Zeeman splitting of the quasiparticle band, as suggested by specific-heat and NMR measurements.[1,12] Then, the carrier concentration is also increased to lead to the negative MR.

For the field parallel to the *c* axis, a positive MR appears below 10 K for *I*//*a* and below 5 K for *I*//*b* and *I*//*c*. The positive MR is much larger for the transverse configurations (*I*//*a* and *I*//*b*) than that for longitudinal configuration (*I*//*c*). With increasing field up to 15 T at 0.45 K, the resistivity for *I*//*a* reaches a large value of 770 μΩcm without showing a saturated behavior.[10] These facts indicate that the positive MR is a result of cyclotron motion of carriers. The strong decrease of carrier concentrations due to the gap formation may increase the relaxation time of carriers so that the cyclotron motion becomes possible.[7]

The above results of MR have suggested that the density of states at the Fermi level is reconstructed when high field is

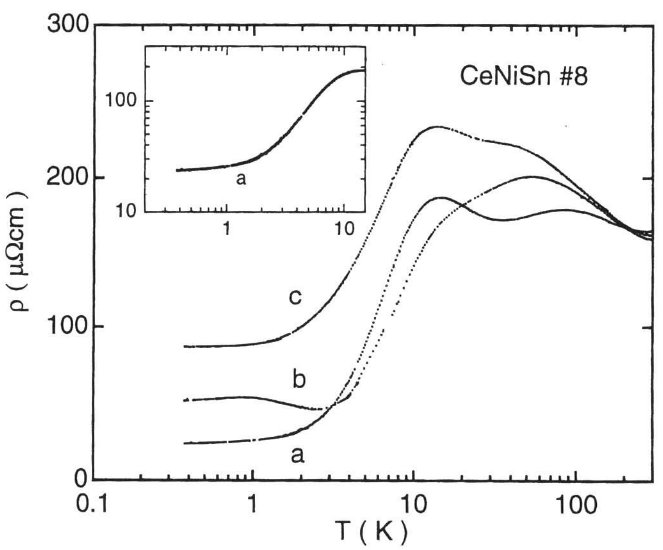

Fig. 1. Temperature dependence of electrical resistivity for CeNiSn crystal #8.

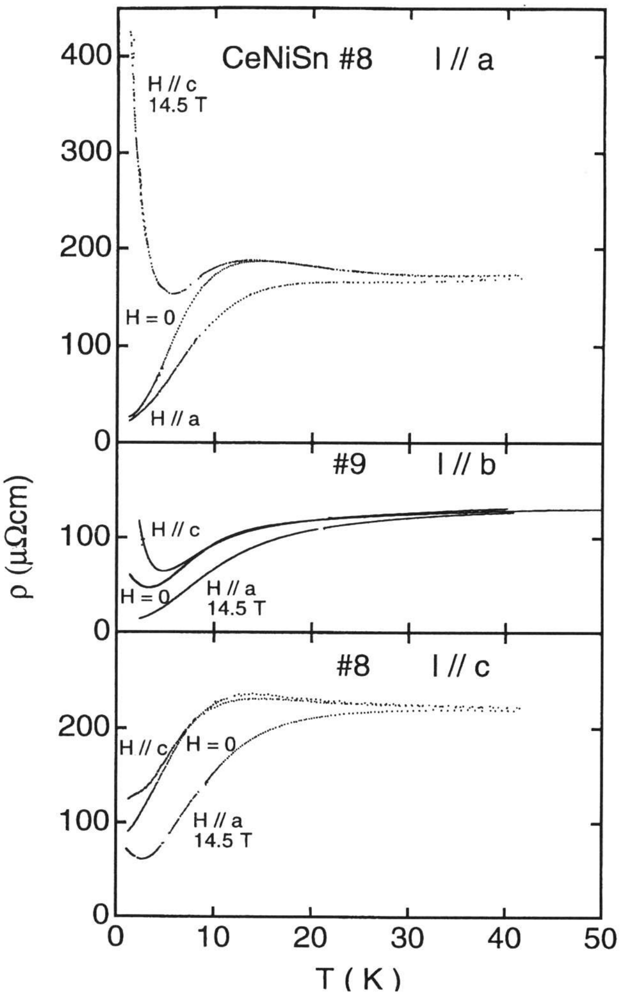

Fig. 2. Electrical resistivity vs temperature for CeNiSn in fields 0 and 14.5 T applied along the *a* and *c* axes for electrical currents along the three principal axes.

applied along the *a* axis. Thereby, the nature of the carriers would be largely altered. In order to study this effect, we have measured the field dependence of Hall resistivity for *H*//*a* and *H*//*c*. The Hall resistivity in usual systems depends on the direction of the field but does not depend on the direction of current as a consequence of the Onsager symmetry principle of the kinetic coefficients. We have examined whether or not this principle is applicable to the present system with strongly anisotropic properties. As shown in Figs. 3(a) and 3(b), the data of $\rho_{xy}(H)$ for *H*//*a* and *J*//*b* at 1.5 and 4.2 K are almost identical to those for *H*//*a* and *J*//*c*, respectively, proving the symmetry principal to hold in the present system. In Figs. 3(c) and 3(d), two sets of data of $\rho_{xy}(H//c)$ for *J*//*a* and *J*//*b* are again similar each other.

The initial slope of the $\rho_{xy}(H)$ cueve yields the Hall coefficient R_H. The values for *H*//*a* and *H*//*c* at 1.5 K are - 6.9 × 10^{-2} and -11.2 × 10^{-2} cm^3/C, respectively, which agree with those reported for crystal #4 in Ref. 7. At 1.5 K, $\rho_{xy}(H)$ for *H*//*a* initially decreases and passes through a minimum at 2.5 T. After changing sign from negative to positive at 5 T, $\rho_{xy}(H)$ exhibits a maximum at 11 T. There is no anomaly around 14 T, where a metamagnetic behavior was found in the magnetization curve.[1] The values of the fields at the

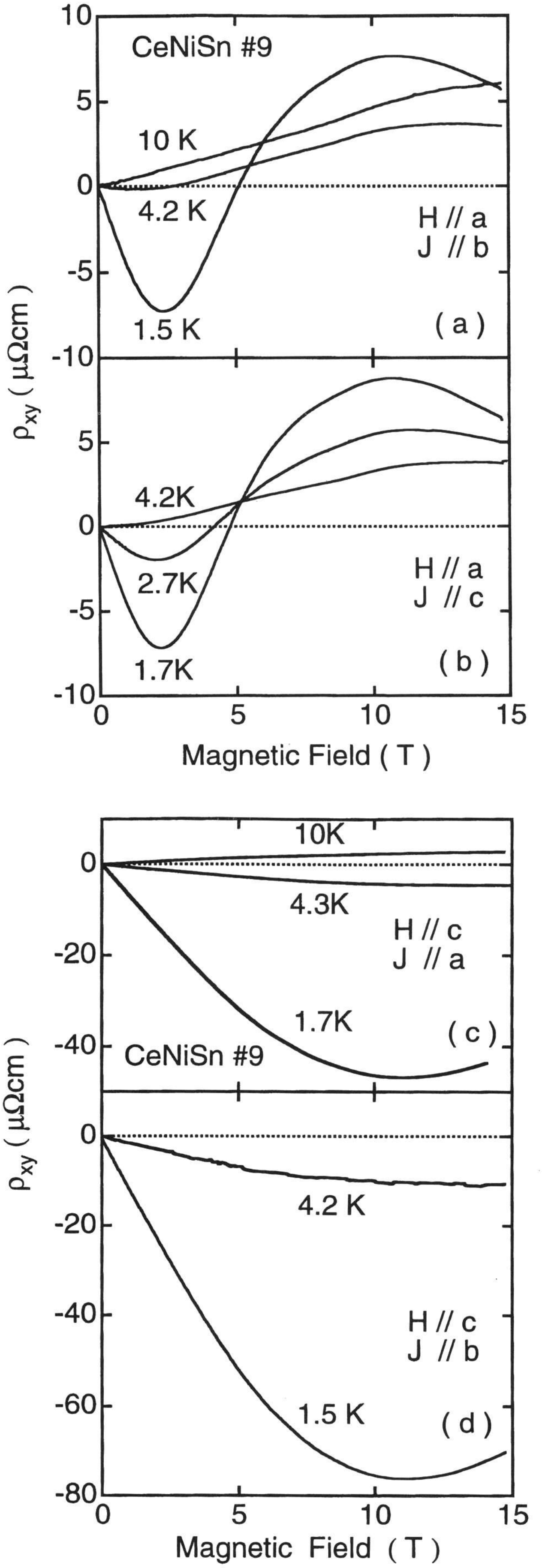

Fig. 3. Hall resistivity of CeNiSn as a function of magnetic field applied along the *a* and *c* axes at various temperatures.

minimum and maximum agree respectively with those of the two maxima in the curve of MR for *H//a* and *J//c* at 0.45 K.[10] The double-extreme structure in $\rho_{xy}(H)$ becomes weak with increasing temperature, and $\rho_{xy}(H)$ at 10 K increases linearly with *H* up to 15 T. Recalling that the V-shape gap develops only below 10 K, the double-extreme structure must be related to the field suppression of the V-shape gap. When the field is applied along the *c* axis, the V-shaped gap is hardly affected because the magnetic susceptibility for *H//c* is much smaller than that for *H//a*. [1] Nevertheless, a nonlinear behavior in $\rho_{xy}(H)$ becomes evident above 7 T and the minimum at 11 T mimics that observed at 2.5 T for *H//a*. It is therefore interesting to test whether $\rho_{xy}(H//c)$ becomes positive in still higher fields.

It is generally difficult to separate the observed Hall coefficient R_H for Ce-based compounds into the normal part and the anomalous part originating from the skew-scattering due to orbital momentum of 4*f* electrons. In the case of CeNiSn, the temperature dependence of resistivity becomes weak below 2 K, as shown in Fig.1, but R_H changes as $\ln T$ (see Fig. 6 in Ref. 7). Since the anomalous part in the coherent temperature region is known to be proportional to the square of resistivity,[14] its contribution in CeNiSn would be almost independent of temperature below 2 K. Therefore, we may conclude that the normal part dominates in the $\ln T$ dependence of R_H in zero field. Accordingly, the unusual field dependence of $\rho_{xy}(H//a)$ is assumed to reflect the field dependence of the normal part.

The strongly nonlinear behavior in $\rho_{xy}(H//a)$ distinguishes the ground state of CeNiSn from a compensated metallic state, for which a field independent Hall coefficient is expected from the usual two-band model. In fact, for a compensated metal $CeSn_3$, ρ_{xy} is a quasilinear function of *H*, and R_H depends only weakly on the temperature.[15] The inversion of sigh of $\rho_{xy}(H//a)$ for CeNiSn suggests that the ratio of concentrations for electrons and holes changes significantly with increasing field. Such a change is expected to occur when the V-shaped density of states with a small flat region at the Fermi level are overlapped by the Zeeman splitting. This model is consistent with recent NMR measurements in high magnetic fields, which have suggested that the overlapping takes place above 4 T.[12]

§ 4. Summary

We measured the Hall resistivity and electrical resistivity of high-quality single crystals of CeNiSn in magnetic fields up to 15 T. For *H//a*, a large negative MR appears below 40 K. This negative MR can be attributed to two effects of magnetic field; one is the suppression of antiferromagnetic spin fluctuations which is dominant between 40 and 5 K and the other is the suppression of the pseudogap which becomes dominant below 5 K. However, for *H//c*, a positive MR was observed below 5 K for the transverse configurations. This originates in cyclotron motion of carriers remaining at the Fermi level. On the other hand, the field dependence of the Hall resistivity for *H//a* is strongly non-linear at temperatures below 5 K. The inversion of the sign of Hall resistivity around 5 T suggests that the V-shaped density of states overlaps by the Zeeman effect.

Acknowledgments

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