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U_2Ru_2Sn: a new Kondo insulator?

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Abstract

We present magnetic susceptibility, specific heat, electrical resistivity, magnetoresistance, Hall coefficient, thermal conductivity, thermoelectric power, and ^{119}Sn NMR data for U_2Ru_2Sn. Clear evidence is found for the formation of a gap in the electronic density of states at the Fermi level. Together with the disappearance of the local magnetic moments at low temperatures and the observation of enhanced effective masses this suggests to classify U_2Ru_2Sn as a Kondo insulator.

Keywords: Low-carrier-density system; Kondo insulator; U_2Ru_2Sn

The tetragonal compound U_2Ru_2Sn has been considered as a weakly temperature dependent paramagnet [1]. However, recently, Menon et al. [2] found that the electrical resistivity is atypical for a simple paramagnet, and is rather indicative of a semiconducting behavior. In this work, we study magnetic susceptibility, specific heat, electrical resistivity, magnetoresistance, Hall coefficient, thermoelectric power, thermal conductivity, and ^{119}Sn NMR of two polycrystalline samples of U_2Ru_2Sn. We show that the observed physical properties can be interpreted in terms of the formation of a gap at the Fermi level.

Polycrystalline samples of U_2Ru_2Sn were synthesized as described in Ref. [2]. The magnetic susceptibility $\chi(T)$ of U_2Ru_2Sn (Fig. 1(a)) behaves much as it does in known Kondo insulators, e.g., $Ce_3Bi_4Pt_3$ [3], CeNiSn [4], CeRhSb [5]. It shows Curie–Weiss behavior at high temperatures, has a broad maximum at $T_{max} = 170$ K, and decreases strongly at lower temperatures, which reflects the opening of an energy gap Δ. The relationship $\Delta/k_B \propto T_{coh} \propto 4T_{max}/(2J+1)$ was used to estimate the gap value for $Ce_3Bi_4Pt_3$ [3]. Taking $J = 8/2$ for U^{4+}, we obtain $\Delta/k_B = 76$ K for U_2Ru_2Sn. Around 25 K, a knee-like anomaly of χ at $H = 1$ kOe is found. With increasing field, this anomaly shifts down to lower temperatures and becomes completely suppressed in $H >$ 30 kOe. Therefore, this anomaly seems to be associated with antiferromagnetic correlations. We recall that a similar anomaly was observed in CeNiSn [4]. It should be noted that this anomalous low-field χ-behavior was observed only for one orientation of the sample with respect to the field. In this configuration χ for $T \geqslant 50$ K is larger by 10% than χ measured on the sample rotated by 90°, pointing to a texture in the polycrystalline sample.

The analysis of the specific-heat data using $C_p(T) = \gamma T + \beta T^3 + AT^{-2}\exp(-\Delta/k_BT)$ [7] yields Δ/k_B of approximately 71 K, in good agreement with the gap extracted from $\chi(T)$ above, $\gamma = 18 \pm 1$ mJ/K^2 mol U, and $\beta = 1 \pm 0.1$ mJ/K^4 mol U.

Similarly to $\chi(T)$, the zero-field electrical resistivity $\rho(T)$ also shows a broad maximum (Fig. 1(b)), but at a lower temperature of 125 K. Below this maximum, $\rho(T)$ strongly drops, which might be due to the onset of coherence. Below $T_{min} = 25$ K, $\rho(T)$ shows an upturn and tends to saturate with further decrease in

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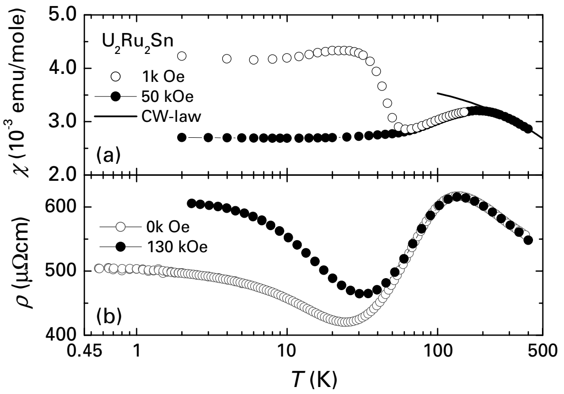

Fig. 1. (a) Magnetic susceptibility and (b) electrical resistivity of U_2Ru_2Sn as a function of temperature.

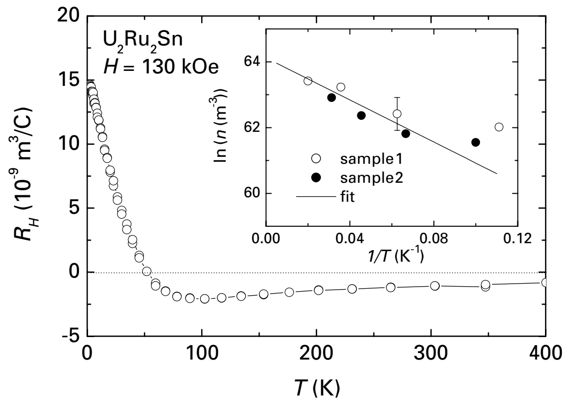

Fig. 2. Temperature dependence of the Hall coefficient of U_2Ru_2Sn. Inset: Semi-logarithmic plot of the carrier concentrations of the heavy holes of two different samples versus $1/T$.

temperature below 1 K. In magnetic fields, ρ is slightly reduced between 100 and 400 K. A big difference between $\rho(T, H = 0)$ and $\rho(T, H = 130\ \text{kOe})$ appears only below 25 K. The field shifts T_{min} towards higher temperature and enhances ρ, resulting in a positive magnetoresistance of about 22% at 2 K and 130 kOe.

In Fig. 2 we show the temperature dependence of the Hall coefficient R_H. The room temperature Hall resistivity $\rho_H = R_H B$ varies linearly with the magnetic field, with the slope $-1.08 \times 10^{-9}\ m^3/C$, indicating n-type conduction. In a one-band model, this corresponds to a carrier concentration $n_e = 5.70 \times 10^{27}\ m^{-3}$ (= 0.57 electrons/f.u.) and a Hall mobility of $1.92 \times 10^{-4}\ m^2/V\ s$. With decreasing temperature, the Hall coefficient changes sign from negative to positive near 50 K. Therefore, there are at least two different kinds of carriers involved in the electrical transport. Below 30 K, ρ_H is no longer linear in field. Using a two-band analysis [8], we have estimated the carrier concentrations and the mobilities of two sets of carriers: high- and low-mobility holes, which we refer to as ''light'' and ''heavy'' holes. At 2 K, the carrier concentration of the light holes is, with $4.44 \times 10^{23}\ m^{-3}$, much smaller than that of the heavy ones, which is $5.59 \times 10^{26}\ m^{-3}$ (= 0.06 /f.u.). Combining this latter value with the γ-value from above in a free electron expression, we obtain an effective mass of approximately $48m_0$ for the heavy holes. In the inset of Fig. 2 we show the temperature dependence of the carrier concentration n of the dominating (heavy) carriers. Between 16 and 50 K, n may be described by $n \propto \exp(-\Delta/2k_B T)$ with $\Delta/k_B \approx 60(10)$ K. The Hall mobilities increase with decreasing temperature. The mobility of the light holes at 2 K is $0.176\ m^2/V\ s$, which is about two orders of magnitude higher than the mobility of the heavy holes.

The thermoelectric power S of U_2Ru_2Sn is positive over the investigated temperature range. S has a hump around 120 K with a relatively large value of 30 μV/K and a second smaller hump with a maximum at approximately 20 K. The gap temperature estimated from $\chi(T)$ and $C_p(T)$ corresponds to the crossover between the two humps.

The thermal conductivity κ of U_2Ru_2Sn decreases smoothly between 200 and 50 K, where it suddenly increases and then passes over a maximum at approximately 30 K. The reduced Lorenz number L/L_0, where $L = \rho\kappa/T$ and $L_0 = 2.45 \times 10^{-8}\ (V/K)^2$, increases with decreasing temperature in the high temperature range, assuming a value of about 15 at 50 K. Below this temperature, a stronger increase of L/L_0 sets in. This increase may be related to the gap opening. The large L/L_0 values in the entire temperature range indicate that κ is dominated by phonons. The phonon contribution to the thermal conductivity is related to the lattice heat capacity C_{ph}, the sound velocity v, and the relaxation time τ_{ph} via the relation $\kappa_{ph} = C_{ph} v^2 \tau_{ph}/3$. Since both C_{ph} and v are expected to vary smoothly with temperature, the observed sudden enhancement of κ_{ph} is most probably associated with an enhancement of τ_{ph}. The increase of κ below 50 K could be due to a reduced scattering rate of the phonons from the freezing-out charge carriers, a mechanism which was also used to explain a similar anomaly in CeNiSn [9].

A strong support for the formation of a spin gap is given by the temperature dependence of the spin lattice relaxation rate $1/T_1$ of ^{119}Sn NMR measurements at 70 kOe. Below 150 K, this quantity decreases markedly. Between 120 and 20 K, the data are well described by the law $1/T_1 T \propto \exp(-\Delta/k_B T)$, with $\Delta/k_B = 75(5)$ K. At lower temperatures the decay of $1/T_1$ is much weaker, which is very similar to the findings for CeNiSn [10].

In summary, we have reported bulk and microscopic measurements for U_2Ru_2Sn. We have found clear evidence for the opening of a narrow gap of approximately 70 K (6 meV) in U_2Ru_2Sn. Many similarities to the behavior of CeNiSn suggest that the formation of the gap in both compound may have the same origin.

Uncited reference

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