

The Kondo Insulator U_2Ru_2Sn

Silke Paschen, Michael Baenitz, Vinh-Hung Tran, Annegrit Rabis, Narayani Senthilkumaran, John A. Mydosh, André Strydom*, Paul de V. du Plessis**, Gaku Motoyama***, Noriaki K. Sato*** and Frank Steglich

Heavy-fermion semiconductors or semimetals, usually referred to as “Kondo insulators” [1], are an intriguing set of materials: The origin of the formation of a narrow gap or pseudogap at the Fermi energy is still unclear. In the last report we discussed the Kondo insulator CeNiSn [2] and our aim of finding new Kondo insulators in the class of clathrate compounds [3]. Here we report on the first tetragonal Kondo insulator, U_2Ru_2Sn . Based on the observation that the electrical resistivity increases upon cooling below 30 K, Menon et al. [4] have tentatively classified U_2Ru_2Sn (tetragonal $P4/mbm$ structure) as a Kondo insulator. Triggered by this interesting proposition we have started to investigate both poly- and single crystalline samples by various techniques. The polycrystals were prepared by melting the elements in an arc furnace [4]. The single crystals were prepared by the Czochralski technique using a 3-arc furnace. a and c axis oriented samples were prepared by polishing Laue-oriented pieces [5]. Part of the results presented below have been published [5-7].

The magnetic susceptibility $\chi(T)$ measured on a single crystalline sample both along the a and the c axis shows a distinct anisotropy (Fig. 1). It is

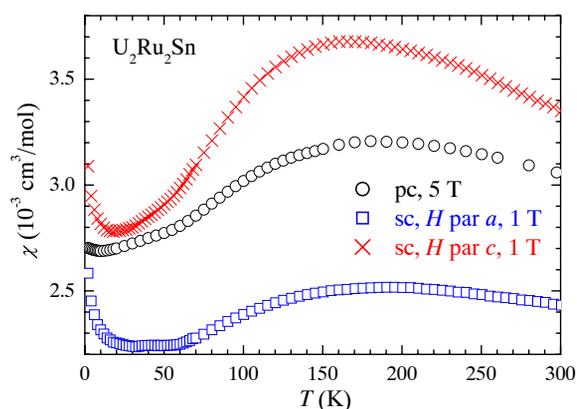


Fig. 1: Temperature dependence of the magnetic susceptibility $\chi(T)$ of a single crystal (sc) with the magnetic field H along either the a or the c axis and of a polycrystal (pc) of U_2Ru_2Sn , in fields of 1 T (sc) and 5 T (polycrystal, pc).

approximately 1.5 times larger along the c axis than along the a axis. Thus, the former may be identified as the easy magnetic axis. The data of the polycrystal fall in between the two single-crystal curves, as expected. The pronounced decrease of $\chi(T)$ below approximately 160 K is typical for Kondo insulators and reflects the opening of the energy gap.

Figure 2 shows the specific heat $C_p(T)$ of both, a single crystal and a polycrystal of U_2Ru_2Sn , as well as of a polycrystal of the non- f reference compound Th_2Ru_2Sn . Displayed on the right axis is the difference between the U_2Ru_2Sn and the Th_2Ru_2Sn data. In the temperature range above approximately 8 K, where the sample-dependent electronic contribution to $C_p(T)$ is unimportant, this difference may be regarded as the magnetic contribution. It is very similar for the single crystal and the polycrystal and may reasonably well be described with a Schottky-like anomaly, $\Delta C_p/R = (T/T_0)^2 \times \exp(T/T_0)/(1 + \exp(T/T_0))^2$ with the interlevel separation $T_0 = 80$ K. In the band picture of two sharp density-of-states peaks around the energy gap, which is frequently employed to model Kondo insulators [8], $k_B T_0$ is

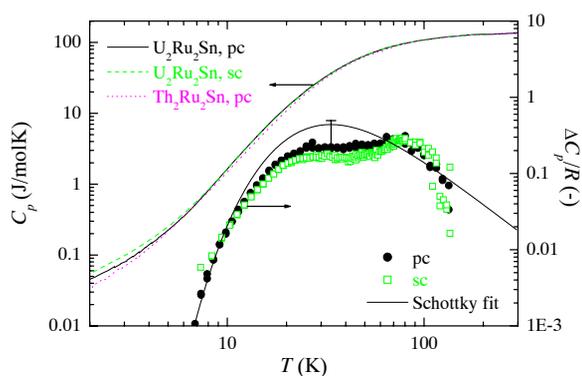


Fig. 2: Left scale: Temperature dependence of the specific heat $C_p(T)$ of a single crystal (sc) and a polycrystal (pc) of U_2Ru_2Sn , and of a polycrystal of Th_2Ru_2Sn . Right scale: Difference of the U_2Ru_2Sn and Th_2Ru_2Sn data ΔC_p , in units of the gas constant R , vs. temperature T . The large error bar for ΔC_p results from an error of 3% estimated for the C_p data. The solid line shows the best Schottky fit to both (sc and pc) data sets.

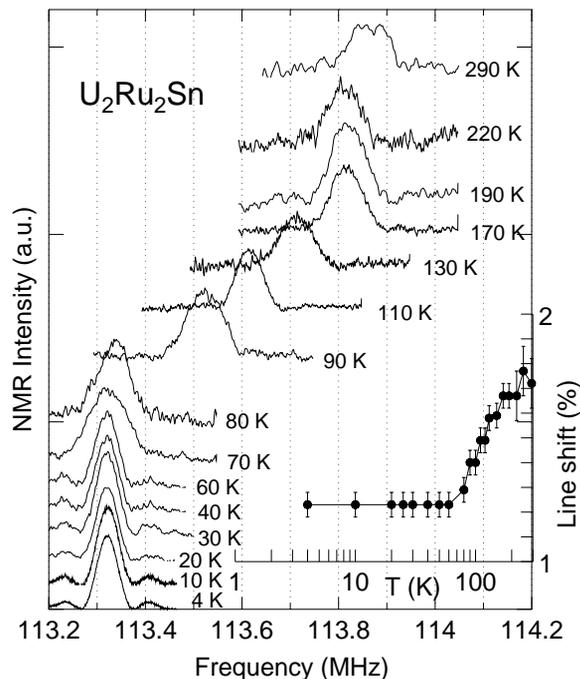


Fig. 3: ^{119}Sn NMR spectra at different temperatures. The shift is determined with the Larmor frequency of ^{119}Sn (111.91 MHz).

the activation energy. Thus, the band gap of $\text{U}_2\text{Ru}_2\text{Sn}$ is $2k_{\text{B}}T_0$ corresponding to 160 K. This is in excellent agreement with the value of 155 K extracted from ^{119}Sn NMR data, to be presented below.

Nuclear magnetic resonance (NMR), being a local probe, has proven a powerful tool to determine the energy gap also in polycrystalline samples. Therefore, we performed ^{119}Sn ($I = 1/2$) NMR measurements on polycrystalline powder samples. NMR spectra were obtained by Fourier-transforming the digitized spin echoes using a conventional pulsed NMR spectrometer (Bruker, MSL 300, $\mu_0H = 7.05$ T). Typical ^{119}Sn NMR spin echo spectra of $\text{U}_2\text{Ru}_2\text{Sn}$ are shown in Fig. 3 at various temperatures. At low temperatures the spectrum has a symmetrical Lorentzian line shape with a linewidth of approximately 122.5 kHz (FWHM, at 4 K) and a Knight shift K of 1.23 % (at 4 K). For the non-magnetic reference compound $\text{Th}_2\text{Ru}_2\text{Sn}$ we found, at 4 K, a linewidth of 41.8 kHz and $K = 0.18$ %. At low temperatures the NMR line shows practically no shift whereas, between 80 K and approximately 150 K the line is shifted strongly to higher frequencies and the linewidth is increased (see inset of Fig. 3). $K(T)$ roughly tracks $\chi(T)$ of the polycrystal

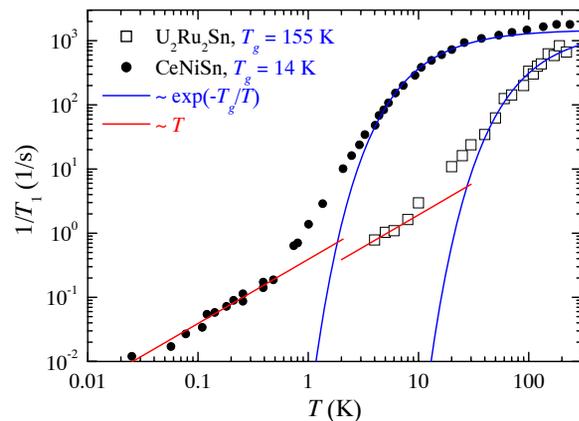


Fig. 4: T dependence of $1/T_1$ for $\text{U}_2\text{Ru}_2\text{Sn}$ and for CeNiSn [9]. The solid lines represent fits to the data, as discussed in the text.

(Fig. 1). First NMR measurements on oriented powder samples show that, in contrast to the (anisotropic) susceptibility, the effect of the magnetic anisotropy on K is negligible: $K_a - K_c \leq 0.1$ % holds over the entire temperature range. From this we conclude that the hyperfine coupling constant A_{hf} should be larger in the a than in the c direction.

The spin-lattice relaxation rate $1/T_1$ as a function of temperature obtained from the NMR signals is plotted in Fig. 4. Due to the different orientations in the polycrystalline sample, the T_1 values shown here represent an average. Investigations on single crystals are under progress. $1/T_1$ for $\text{U}_2\text{Ru}_2\text{Sn}$ is plotted together with the data for CeNiSn ($\mu_0H = 1.26$ T) from Ref. 9. Below approximately 150 K, the rate for $\text{U}_2\text{Ru}_2\text{Sn}$ decreases drastically over three orders of magnitude whereas, at very low temperatures, a linear temperature dependence is observed. As a first approach we fit the rate with an exponential curve: $1/T_1 \propto \exp(-\Delta/k_{\text{B}}T)$. This relation is well known for spin-Peierls systems or conventional superconductors. If one fits an exponential to the published data for CeNiSn [9] one obtains a gap value of $\Delta/k_{\text{B}} = 14$ K. The striking deviation from the exponential curve observed below $T \lesssim 4$ K has been ascribed to a ‘V-shaped gap’ in combination with a residual density of states within the gap [9]. For $\text{U}_2\text{Ru}_2\text{Sn}$ our results reveal a gap value of $\Delta/k_{\text{B}} = 155$ K, one order of magnitude larger than for CeNiSn .

The electrical resistivity of $\text{U}_2\text{Ru}_2\text{Sn}$ is only weakly temperature dependent but shows a distinct

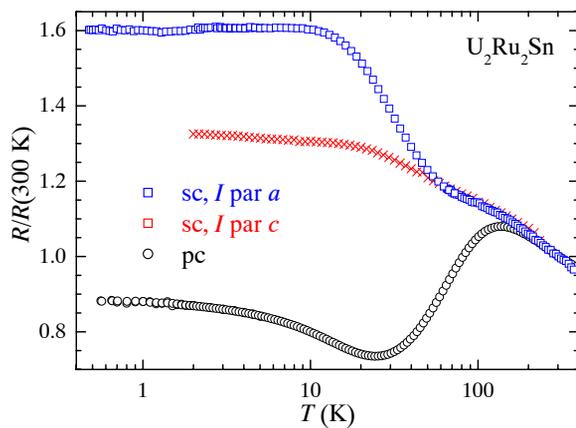


Fig. 5: Temperature dependence of the electrical resistance normalized to room temperature $R(T)/R(300\text{ K})$ of a single crystal (sc) with I along either the a or the c axis and of a polycrystal (pc) of $\text{U}_2\text{Ru}_2\text{Sn}$.

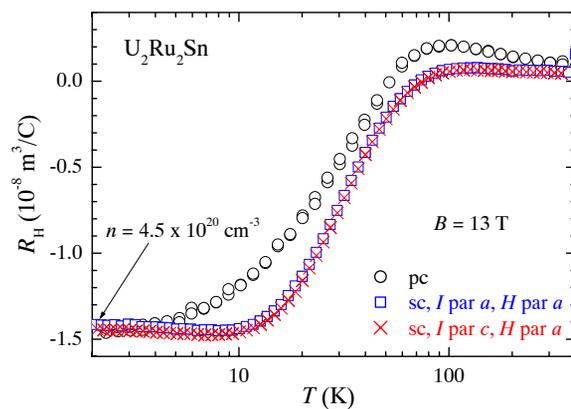


Fig. 6: Temperature dependence of the Hall coefficient $R_H(T)$ of a single crystal (sc) with I along either the a or the c axis and H along the c axis, and of a polycrystal (pc) of $\text{U}_2\text{Ru}_2\text{Sn}$.

anisotropy (Fig. 5). The absolute resistivity values are a few hundred $\mu\Omega\text{cm}$. It is not clear at present why the data of the polycrystal do not fall in between those for the two directions of the single crystal.

Figure 6 shows the temperature dependence of the Hall coefficient $R_H(T)$ of both single- and polycrystalline $\text{U}_2\text{Ru}_2\text{Sn}$. The overall behavior for both samples is similar. For the single crystal, the data with the current I along the a and the c axis virtually coincide. Thus, unlike $\rho(T)$, $R_H(T)$ is isotropic.

Therefore, the Hall mobility $\mu_H = R_H/\rho$ of $\text{U}_2\text{Ru}_2\text{Sn}$ must be anisotropic. The reason for the weak temperature dependence of the electrical resistivity in spite of the energy gap opening can now be understood as follows: As the energy gap opens and coherence sets in, the strong decrease of the charge-carrier concentration is almost exactly compensated by the strong increase of the Hall mobility. Further experiments on samples of different qualities are on the way to determine whether the low-temperature transport is intrinsic or impurity dominated.

References

- [1] G. Aeppli and Z. Fisk, *Comments Condens. Matter Phys.* **16**, 155 (1992).
- [2] S. Paschen, B. Wand, G. Sparrn, F. Steglich, Y. Echizen, and T. Takabatake, in *Development of the Institute and Scientific Report* (Max-Planck-Institut für Chemische Physik fester Stoffe, Dresden, Nov. 2000).
- [3] S. Paschen, W. Carrillo-Cabrera, M. Baenitz, V.H. Tran, A. Bentien, H. Borrmann, R. Cardoso Gil, R. Michalak, Yu. Grin, and F. Steglich, in *Development of the Institute and Scientific Report* (Max-Planck-Institut für Chemische Physik fester Stoffe, Dresden, Nov. 2000).
- [4] L. Menon, P. de V. du Plessis, and A. M. Strydom, *Solid State Commun.* **106**, 519 (1998).
- [5] S. Paschen, V. H. Tran, N. Senthilkumar, M. Baenitz, F. Steglich, A. M. Strydom, P. de V. du Plessis, G. Motoyama, and N.K. Sato, to appear in *Physica B*.
- [6] S. Paschen, M. Baenitz, V. H. Tran, A. Rabis, F. Steglich, W. Carrillo-Cabrera, Yu. Grin, A. M. Strydom, and P. de V. du Plessis, *J. Phys. Chem. Solids* **63**, 1183 (2002).
- [7] M. Baenitz, A. Rabis, S. Paschen, N. Senthilkumar, F. Steglich, V. H. Tran, P. de V. du Plessis, and A. M. Strydom, to appear in *Physica B*.
- [8] P. S. Riseborough, *Adv. Phys.* **49**, 257 (2000).
- [9] K. Nakamura, Y. Kitaoka, K. Asayama, T. Takabatake, G. Nakamoto, and H. Fujii, *Phys. Rev. B* **54**, 6062 (1996).

* Physics Department, Rand Afrikaans University, PO Box 524, Johannesburg, South Africa.

** School of Physics, University of the Witwatersrand, PO Wits 2050, Johannesburg, South Africa.

*** Department of Physics, Graduate School of Science, Nagoya University, Nagoya 464-8602, Japan.