LA-UR - 91-1536

LA-UR--91-1536

DE91 013672

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SUBMITTED TO 19th Rare Earth Conference
Lexington, KY, 14-19 JULY 91
Journal of Less-Common Metals

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PRESSURE DEPENDENCE OF THE CURIE-TEMPERATURE IN THE KONDO-LATTICE-COMPOUND YbNiSn.

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Summary

We have measured the magnetic susceptibility ($2\text{K} < T < 300\text{K}$) and the electrical resistivity under pressure ($1.2\text{K} < T < 300\text{K}$; $p < 20\text{kbar}$) of the Kondo-lattice compound YbNiSn, which may be considered as the hole analogue of the isostructural compound CeNiSn. In contrast to CeNiSn, YbNiSn does not show an energy gap at low temperatures, but instead undergoes a magnetic phase transition at $T_M = 5.5\text{K}$. The magnetic state might be either a weakly ferromagnetic or a complex antiferromagnetic state. By applying pressure the room temperature resistance (ρ_{RT}) decreases, while T_M increases. This behavior can be interpreted with respect to Doniach's Kondo-necklace model as a decrease of the local exchange coupling constant J with increasing pressure, which reduces Kondo-spin compensation and favors long-range magnetic order. These results show that the idea of YbNiSn being the hole counterpart to CeNiSn holds well for the properties such as

dT_M/dP and $d\rho_{RT}/dP$ but that it fails in more detailed aspects, like the origin of the gap formation.

1. Introduction

Recent attention has focused on the rare-earth compounds CeNiSn and YbNiSn which have the orthorhombic ϵ -TiNiSi-structure [1]. CeNiSn was found to be a Kondo-lattice compound, showing Kondo-like behavior at high temperatures ($T_K \approx 100K$) [2] and a small semiconductor-like energy gap at low temperatures ($\Delta E/k_B \approx 6K$) [3]. The physical properties of CeNiSn resemble those of SmB₆, YbB₁₂, SmS and TmSe [4-7] which are well known valence-fluctuating compounds that show semiconductor-like behavior at low temperatures. In contrast to CeNiSn and YbNiSn the latter compounds have a cubic crystal structure and exhibit an energy gap of several times 10K.

YbNiSn may be considered the hole analogue to CeNiSn, since the valence state of the magnetic configuration of Yb ($Yb^{3+}; 4f^{13}$) has one hole with respect to the fully occupied, non-magnetic 4f-shell ($Yb^{2+}; 4f^{14}$). Unlike CeNiSn, YbNiSn is metallic at ambient pressure. The absence of an energy gap in YbNiSn may be not surprising, because its formation critically depends on the detailed band structure near the Fermi energy. As shown for CeNi_{1-x}Cu_xSn, the gap formation can be depressed by substituting small amounts of Ni by Cu [8], or by applying hydrostatic pressure to CeNiSn ($p=22$ kbar) [9]. Furthermore, the

4f-valence electrons are more localized in Yb than they are in Ce.

Here we focus on changes in the relative importance of Kondo-spin fluctuations and Rudermann-Kittel-Kasuya-Yosida-interactions (RKKY) in YbNiSn as revealed from high pressure studies of the electrical resistance and the magnetic ordering temperature.

2. Experimental Procedure

The sample was prepared by melting the constituent elements in a sealed molybdenum crucible and annealing it at 750°C for five days. X-ray analysis showed the proper orthorhombic ϵ -TiNiSi-structure [1]. The electrical resistance was measured using a four-probe Lockin-technique. The high pressure experiment was performed in a BeCu piston/cylinder cell. Using a 1:1 mixture of isoamyl-alcohol and n-pentane as pressure medium provides nearly hydrostatic pressure conditions down to the lowest temperature of $T=1.2\text{K}$ reached in this experiment. The pressure was determined at $T\approx 7\text{K}$ by in. uctively monitoring the pressure-induced shift of the superconducting transition temperature of a small piece of lead mounted along with the sample [10].

3. Results and Discussion

3.a Susceptibility

Figure 1 shows the inverse magnetic susceptibility of YbNiSn as a function of temperature between T=2K and room temperature. At high temperatures the inverse susceptibility follows a Curie-Weiss law from which we deduce an effective magnetic moment $\mu_{\text{eff}}=4.32 \mu_B/\text{Yb}$, a value close to the free moment of the $J=7/2$ state of Yb^{3+} ($\mu_{\text{eff}}=4.54 \mu_B$). The paramagnetic Curie-temperature $\theta=-31\text{K}$ is in reasonable agreement with previous results ($\theta=-65\text{K}$ [11]; $\theta=-43\text{K}$ [12]). Such a variation in θ -values may arise from the presence of anisotropic magnetic interactions that might be expected from the orthorhombic crystal structure. For $T<100\text{K}$ the effective magnetic moment gradually decreases due to the onset of crystalline-electric field-splitting of the $J=7/2$ multiplet and/or the development of Kondo-interactions.

The inset in Fig.1 shows the low-temperature magnetic susceptibility in which there is evidence for a magnetic transition at $T_M=5.5\text{K}$. These data correspond well to previous observations [11-14], except that in our case the magnitude of the spontaneously ordered moment $\mu_S \approx 0.01 \mu_B$, determined by cooling the sample below T_M in an applied field of 100 Oe, is much smaller than previously reported ($\mu_S=0.4 \mu_B$) [12]. This could be due to a more complex nature of the magnetically ordered state than deduced from previous measurements. An alternative to the assumption that YbNiSn orders ferromagneti-

cally [12] is the possibility of a canted antiferromagnetic structure that would produce a small spontaneous moment, the value of which would depend on the orientation of the applied field with respect to the canted moments. This alternate possibility is consistent with the negative θ , the complete suppression of T_M by an external magnetic field of $B=6.5T$ [13] and the Kondo-lattice-like temperature dependent resistance. It also would imply that RKKY-interactions are responsible for the long range magnetic order. However we can not rule out a weak ferromagnetic-type of ordering that could arise from a net ferromagnetic exchange between Yb ions due to the spatially oscillating nature of the RKKY exchange coupling.

3.b Electrical Resistivity

The electrical resistance of YbNiSn at ambient pressure presented in Fig.2 shows the same double-peak structure reported earlier and which has been attributed to Kondo-lattice behavior [12]. With decreasing temperature, the resistance of YbNiSn shows a crystal-electric-field-derived maximum at $T_{CEF}=66K$, consistent with Mössbauer-spectroscopy data [14] and with the reduced low-temperature moment shown in Fig.1. On applying hydrostatic pressure, T_{CEF} increases slightly (see inset Fig.2), as one would expect from a simple point-charge model and as also observed in YbCuAl for pressures larger than 100 kbar [15]. At lower temperatures, the resistance develops a shoulder that we

interpret as arising from the onset of coherence in the Kondo-impurity scattering process. At still lower temperatures, the resistance drops precipitously, signaling the magnetic phase transition.

The application of pressure causes the electrical resistance at room temperature to decrease. Thinking of YbNiSn as the hole counterpart of CeNiSn, the decrease in room temperature resistance (ρ_{RT}) with increasing pressure reflects quite well its mirror properties with respect to CeNiSn, where ρ_{RT} increases with pressure [9]. This difference between Ce and Yb analogues may be understood by considering that increasing pressure moves the energy level of the hole-type excitation ($E_{f13+\delta}$) further away from the Fermi level (E_F) and thereby reduces the scattering rate; whereas, the opposite holds for Ce.

In Kondo-lattice systems exchange between the 4f- and the conduction-band electrons gives rise to a delicate competition between intra-site Kondo-interactions and inter-site RKKY-interactions, which results in a magnetic phase diagram described first by Doniach [16]. According to Doniach, a decrease in the exchange coupling constant J , i.e., a decrease of the characteristic temperature T^* , favors long range magnetic order.

In Ce-compounds it uniformly appears that $dT^*/dP > 0$, favoring a non-magnetic state with increasing pressure; whereas, in Yb-compounds $dT^*/dP < 0$ favoring the magnetically ordered state [17]. Equivalently, applying pressure favors a higher valence state,

which is non-magnetic for Ce but magnetic for Yb. This general statement is also supported by results of Mignot and Wittig on YbCuAl [15]. At ambient pressure the temperature dependent resistance of YbCuAl is typical of Kondo-lattice systems having a relatively large T^* . On applying hydrostatic pressure to YbCuAl the shape of the resistance curve changes towards that observed in YbNiSn, forming a double-peak structure with the peaks located at $T \approx 50\text{K}$ and $T \approx 10\text{K}$ respectively. In addition a sharp decrease in resistivity occurs at $T = 5\text{K}$ which is ascribed to the onset of long-range magnetic order. Thus, at high pressure YbCuAl appears to have a much reduced T^* , like YbNiSn. This behavior can be described consistently by the arguments given above for YbNiSn.

Because T^* or equivalently J decreases when pressure is applied to Yb-compounds, we would expect T_M of YbNiSn to increase with increasing pressure. This is indeed observed as shown in Fig.3, which gives an expanded view of our resistivity data near the magnetic phase transition. At ambient pressure the transition temperature $T_M = 5.5\text{K}$ (defined by the maximum in dp/dT) is the same as reported by Kasaya et al. and D'Onofrio et al. [12,13] and as found in our susceptibility measurements. At 16.7 kbar T_M reaches 7.6K. As shown in the inset of Fig.3 the rate dT_M/dP is initially 0.3 K/kbar but dT_M/dP decreases to ≈ 0.03 K/kbar at the highest pressures. This pressure dependence of T_M is consistent with Doniach's phase diagram, assuming that YbNiSn at ambient pressure is in the regime of large J -values where

Kondo-spin-fluctuations start to dominate RKKY-interactions. With increasing pressure, i.e., decreasing J , dT_M/dP should go to zero as T_M approaches its maximum. The data in the inset of Fig.3 suggest that YbNiSn at 16.7 kbar is close to having its maximum T_M . To support this suggestion, high pressure experiments in a Bridgeman-cell are under way to follow T_M to still higher pressures. If our interpretation is correct, T_M should reach a maximum and then decrease at the highest pressures at which point J is sufficiently small that RKKY-interactions completely dominate Kondo-spin-fluctuation effects.

In conclusion, we have shown that in many respects YbNiSn acts as the hole analogue of CeNiSn. The most notable discrepancy in this analogy, however, is the absence of a gap in YbNiSn. We believe this difference may be due to a detailed difference in electronic structure. Finally, we have shown that qualitatively the pressure dependence of T_M in YbNiSn follows the behavior expected from the competition between RKKY- and Kondo-interactions. To our knowledge, this is the first clear example of this behavior in a magnetically ordered Yb-compound.

Acknowledgement

Work at Los Alamos was performed under the auspices of the U.S. Department of Energy.

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Figure Captions

- Fig.1 Inverse magnetic susceptibility vs. temperature of YbNiSn. Straight line represents Curie-Weiss fit with coefficients $\theta = -31\text{K}$, $\mu_{\text{eff}} = 4.32 \mu_{\text{B}}/\text{Yb}$. Inset shows a magnification of the data in the vicinity of the magnetic phase transition ($T_{\text{M}} = 5.5\text{K}$).
- Fig.2 Electrical resistance vs. temperature ($1.2\text{K} < T < 300\text{K}$) of YbNiSn at various pressures. Inset shows the pressure dependence of the crystal-electric-field-derived resistance maximum (T_{CEF}).
- Fig.3 Electrical resistance vs. temperature ($1.2\text{K} < T < 20\text{K}$) of YbNiSn at various pressures. Inset shows the pressure dependence of the magnetic ordering temperature (T_{M}).

Fig. 1