Heavy-Fermion Behavior in a Heusler-Type Compound $YbPd_2Sb$?

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Rare-earth-based Heusler phases ***RE***Pd_2***M***, where ***M*** = In, Sn or Pb, attracted t considerable attention because of their interesting magnetic and superconducting properties [1]. Most intriguing are obviously those systems in which superconductivity coexists with long-range magnetic order, as for example $YbPd_2Sn$ [2] and $ErPd_2Sn$ [3]. In these compounds the 4***f***-electrons responsible for antiferromagnetism are fairly coupled to superconducting electrons [4], in contrast to systems like ***RE***Ni_2B_2C, ***RE***Rh_4B_4 or ***RE***Mo_6***Y***$_8$ (***Y*** = S, Se) where the two cooperative phenomena are spatially well isolated. Motivated by this finding we have recently undertaken systematic investigations of $YbPd_2$***M*** phases with ***M*** = In, Pb, Sb and Bi [5]. To our knowledge physical properties of these ternaries have not been studied before.

The compound $YbPd_2Sb$ crystallizes in a cubic unit cell of the $AlCu_2Mn$-type (space group ***Fm*3*m***), characteristic of Heusler phases (Fig. 1). The Yb atoms are located at the 4***b*** (1/2,1/2,1/2) sites, the Pd atoms occupy the 8***c*** (1/4, 1/4, 1/4) sites, while the Sb atoms are placed at the 4***a*** (0, 0, 0) sites. For comparison, in Fig. 1 there are also presented the crystal structures of YbPdSb and YbSb. The latter is of the NaCl-type (space group ***Fm*3*m***) with the very same occupation of the 4***b*** and 4***a*** sites as in $YbPd_2Sb$ but with all the 8***c*** positions being empty. In turn, in the unit cell of YbPdSb, which is of the MgAgAs-type (space group ***F*43*m***), the 4***c*** positions are occupied by the Pd atoms and the other atoms are located as in YbSb. The apparent close structural relationship between the three compounds results in similarities in their physical properties [6].

Polycrystalline samples of $YbPd_2Sb$ were obtained in a two step synthesis. First a Pd_2Sb alloy was prepared by arc melting. Small losses (< 1 wt.%) of antimony were compensated beforehand. The alloy Pd_2Sb was then placed along with Yb metal in a glassy carbon crucible, sealed into a Ta-tube, and melted in an induction furnace. Subsequently, the ingot was subjected to prolonged heat treatment at around 1173 K. All experimental steps were carried out in a inert-gas glove box system ensuring an Ar-gas atmosphere with both oxygen and water contamination at a level of less than 1 ppm. The so-prepared samples were single phase with no detectable deviation from the ideal composition, as proved by X-ray powder diffraction, metallography and EDX analysis. The refined lattice parameter ***a*** = 665.42(2) pm is larger than the value expected on the basis of the lanthanide contraction in the ***RE***Pd_2Sb series [7]. This suggests some instability of the Yb ion valence. Indeed, preliminary Yb L_{III}-edge spectroscopy studies yielded the effective valence slightly lower than +3.

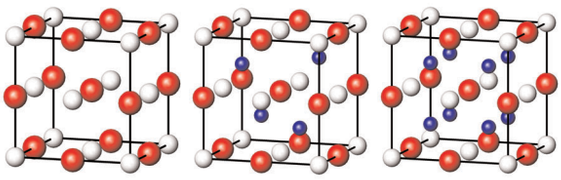

Fig.1: Crystal structure of $YbPd_2Sb$ ($MnCu_2Al$-type), compared to those of YbSb (NaCl-type) and YbPdSb (MgAgAs-type). Red balls - Yb, blue balls - Pd, gray balls - Sb.

Compatible with the above finding is the reduced value of the effective magnetic moment μ_{eff} = 4.02 μ_B, obtained from a Curie-Weiss description of the magnetic susceptibility of $YbPd_2Sb$ above 80 K. The calculated paramagnetic Curie temperature θ_{CW} = −18 K may result from antiferromagnetic exchange interactions. However, its large magnitude in conjunction with the lack of long range magnetic order at least down to 40 mK (see below) hints at Kondo interactions contributing to θ_{CW}. Below 80 K the susceptibility deviates from the Curie-Weiss law, presumably mainly due to a crystal field (CF) effect. The magnetization σ(***B***) measured at ***T*** = 0.1 K exhibits a curvature characteristic of paramagnetic saturation within the CF Kramers ground state. The extrapolation of these data to ***B*** = 0 yields a value corresponding to the magnetic moment of 1.23 μ_B. It is worthwhile noting that this estimate is considerably smaller than μ = 1.71 μ_B of a Γ_7 doublet expected to be a CF ground state in $YbPd_2Sb$ [6].

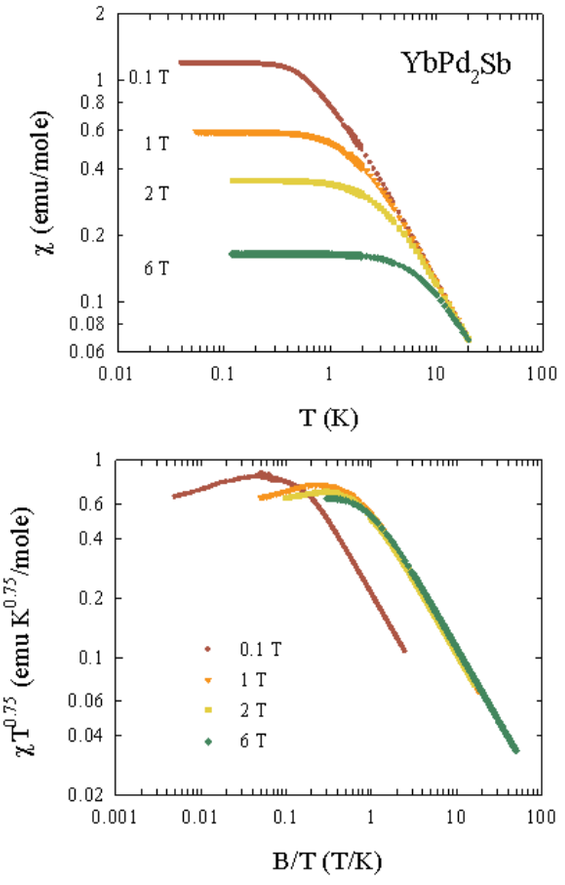

Fig.2: a) Temperature dependence of the low-temperature dc magnetic susceptibility of $YbPd_2Sb$ measured in several magnetic fields. b) The data from panel (a) plotted as $\chi T^{0.75}$ versus B/T .

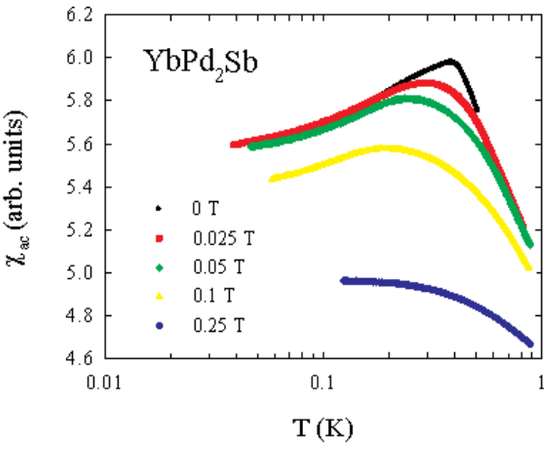

Fig.3: Temperature variation of the ac magnetic susceptibility of $YbPd_2Sb$ measured in B_{ac} = 10 Gs with f = 16.67 Hz in several steady magnetic fields.

At low temperatures the susceptibility is featureless down to a temperature T^* below which $\chi(T)$ saturates at quite high values in a manner characteristic for Kondo systems [8]. In a field of 0.1 T, T^* = 0.4 K and $\chi(T \rightarrow 0)$ = 1.2 emu/mole (Fig. 2a). With increasing field the low-temperature susceptibility is suppressed and T^* gradually rises towards 12 K, the upper border of the region in which the susceptibility is field dependent. Such a behavior is, however, quite unexpected for Kondo systems, being instead rather typical for local moment paramagnets. Interestingly, as shown in Fig. 2b, for $B >$ 0.1 T the low-temperature susceptibility of $YbPd_2Sb$ scales very well to the formula $\chi T^{0.75} = f(B/T)$. Such behavior is appropriate for systems with strongly interacting spin fluctuations [9] like an archetypal non-Fermi liquid alloy $CeCu_{6-x}Au_x$ [10]. Departure from the universal behavior for $B =$ 0.1 T manifests some additional contribution to the susceptibility, possibly due to magnetic exchange interactions. Such a hypothesis seems corroborated by the results of ac susceptibility studies (Fig. 3). The dynamic susceptibility of $YbPd_2Sb$ exhibits a broad maximum at about 0.4 K which hardly alters with varying the frequency of the alternating field except for some little change in the shape of this peak for $T < T^*$. The observed anomaly is, however, strongly sensitive to external steady magnetic fields. Already in a field of 0.025 T one observes a reasonable shift of the peak towards lower temperatures accompanied by its strong broadening while in B = 0.25 T no maximum in $\chi_{ac}(T)$ is discernible in the entire temperature range investigated. It is worthwhile noting that such a strong sensitivity of the ac susceptibility to small magnetic fields is typical for spin glasses [11].

The electrical resistivity of $YbPd_2Sb$ (Fig. 4) is characteristic of systems exhibiting an interplay of Kondo and CF interactions. The absolute values of $\rho(T)$ are rather large but they are well reproducible for different polycrystalline samples. A broad hump at around 70 K manifests CF splitting while a sharp maximum at T^{LT}_{max} = 1.4 K originates probably from coherence effects. In the range 2 K – 9 K the resistivity rises with decreasing temperature in a logarithmic manner and below 1 K (down to 0.09 K) it is proportional to T. Upon applying a magnetic field the resistivity decreases in Kondo-like fashion. The magnetoresistivity $\Delta\rho/\rho$ reaches -7 % at T = 1.8 K and B = 14 T. With increasing temperature the effect of the magnetic field dimin-

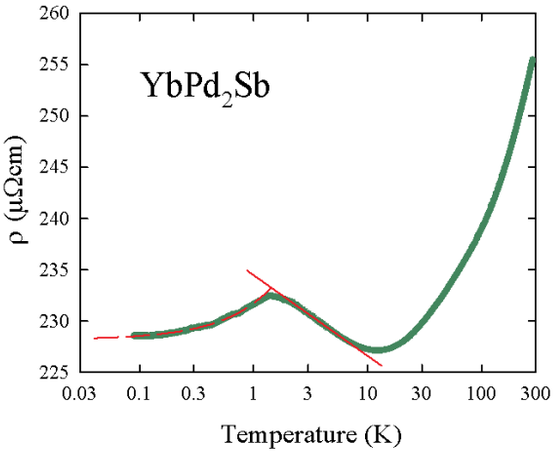

Fig. 4: Temperature dependence of the electrical resistivity of $YbPd_2Sb$. The solid line marks a Kondo-like behavior and the dashed curve emphasizes the proportionality $\rho \sim T$.

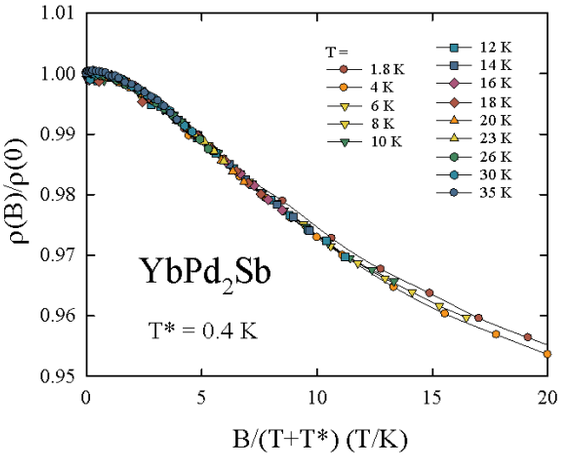

Fig. 5: Field variation of the magnetoresistivity of $YbPd_2Sb$, taken at several different temperatures, plotted as $\rho(B)/\rho(0)$ vs $B/(T+T^*)$ where $T^* = 0.4$ K.

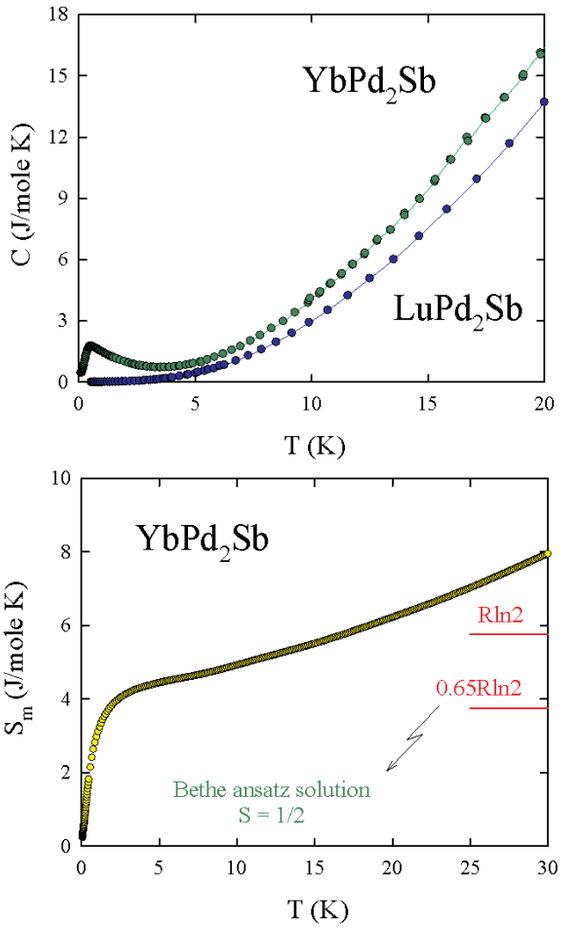

Fig.6: a) Temperature dependence of the specific heat of $YbPd_2Sb$ and $LuPd_2Sb$. (b) Low-temperature non-phononic specific heat and the entropy of $YbPd_2Sb$. The dashed curve represents the Kondo contribution for s = ½ and $T^* = 0.4$ K.

ishes. For $T > 2$ K a single-ion Kondo scaling [12] is fulfilled (Fig. 5) with the characteristic temperature $T^* = 0.4$ K, as found from the magnetic data.

The temperature variation of the specific heat of $YbPd_2Sb$ (Fig. 6) reveals a broad anomaly at $T^* = 0.4$ K but no feature at $T^{LT}_{max} = 1.4$ K. Approximating the phonon contribution $C_{ph}(T)$ by the measured specific heat of $LuPd_2Sb$ one obtains the non-phonon part, $\Delta C(T) = C_p - C_{ph}$, which can be qualitatively described by the Coqblin-Schrieffer model [8] for the effective spin $s = 1/2$ and $T^* = 0.4$ K. The Sommerfeld coefficient interpolated from the region 4 K – 10 K, i.e., where the high-temperature tail of the $T^* = 0.4$ K anomaly is negligible, is equal to 112 mJ/(K^2mole). At the lowest temperatures the ratio C_p/T strongly rises and attains a huge value of about 5 J/(K^2mole) at $T = 0.09$ K. The entropy derived from the $\Delta C(T)$ data is very small (Fig. 6). It shows some tendency to saturate between 2 K and 9 K, i.e., in the range where $\rho \sim \ln T$. This saturation value, however, is larger than the Bethe ansatz solution $s = 1/2$ but considerably reduced with respect to the value expected for a doublet ground state with no Kondo interactions.

Upon applying an external magnetic field the broad peak in $C_p(T)$ shifts to higher temperatures, thus reflecting an increase in the Zeeman splitting

of the CF ground state. From the field dependence of this anomaly one concludes that the lowest energy CF level in $YbPd_2Sb$ is the Γ_7 doublet. Another effect of the magnetic field is the suppression of the low-temperature tail in $C_p(T)$. In high fields (B > 4 T) a Fermi liquid behavior of the specific heat is observed with strongly enhanced values of $\gamma(0)$.

In summary, despite the presence of rather large magnetic moments in $YbPd_2Sb$ (carried by almost trivalent ytterbium ions) no long-range magnetic order has been found down to 40 mK. Instead, some features characteristic of short-range antiferromagnetic correlations are observed in the magnetic, electrical and thermodynamic properties which define altogether the characteristic energy scale T^* = 0.4 K. The electrical resistivity of $YbPd_2Sb$ is governed by the interplay of crystal field and Kondo interactions. Likewise, the low-temperature specific heat can be interpreted in terms of Kondo effect. At the lowest temperatures the compound exhibits clear deviations from the Fermi liquid behavior, typical for systems being close to Doniach's magnetic-nonmagnetic instability. On-going investigations attempt to address questions relevant with respect to the nature of the electronic ground state in $YbPd_2Sb$. This includes the issue of the role played by possible atomic disorder that is commonly present in Heusler-type phases.

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References

[1] ***M. Ishikawa, J. L. Jorda and A. Junod,*** in Superconductivity in d- and f-Band Metals, W. Buckel and W. Weber (eds.), Kernforschungszentrum, Karlsuhe, 1982, p. 141;
C***. L. Seaman, N. R. Dilley, M. C. de Andrade, J. Herrmann, M. B. Maple and Z. Fisk,*** Phys. Rev. B **53**, 2651 (1996).

[2] H***. A. Kierstead, B. D. Dunlap, S. K. Malik, A. M. Umarji and G. K. Shenoy,*** Phys. Rev. B **32**, 135 (1985).

[3] ***R. N. Shelton, L. S. Hausermann-Berg, M. J. Johnson, P. Klavins and H. D. Yang,*** Phys. Rev. B **34**, 199 (1986).

[4] ***Y. Aoki, H. R. Sato, H. Sugawara and H. Sato,*** Physica C **333**, 187 (2000).

[5] ***D. Kaczorowski, A. Leithe-Jasper, T. Cichorek, K. Tenya, J. Custers, P. Gegenwart, and Yu. Grin,*** Proc. SCES'2002, Acta Phys. Polon. B, in press.

[6] ***D. Kaczorowski, A. Leithe-Jasper, T. Cichorek, K. Tenya, J. Custers, P. Gegenwart, and Yu. Grin,*** to be published.

[7] ***P. Riani, D. Mazzone, G. Zanicchi, R. Marazza, and R. Ferro,*** Z. Metallkd. **86**, 450 (1995).

[8] ***V. T. Rajan,*** Phys. Rev. Lett. **51**, 308 (1983).

[9] ***A. Tsvelik, and M. Reizer,*** Phys. Rev. B **48**, 9887 (1993).

[10] ***A. Schröder, G. Aeppli, R. Coldea, M. Adams, O. Stockert, H.v. Löhneysen, E. Bucher, R. Ramazashvili, and P. Coleman,*** Nature **407**, 351 (2000).

[11] ***J. A. Mydosh,*** Spin Glasses: an Experimental Introduction, Taylor and Francis, London, 1993.

[12] ***P. Schlottmann,*** Phys. Rep. **181**, 1 (1989).