Investigation of $\text{Yb}_2\text{Pt}_6\text{Al}_{15}$ single crystals: heavy fermion system with a large local moment degeneracy

M Deppe$^1$, S Hartmann, M E Macovei, N Oeschler, M Nicklas and C Geibel

Max Planck Institute for Chemical Physics of Solids, Nöthnitzer Straße 40, 01187 Dresden, Germany
E-mail: Micha.Deppe@cpfs.mpg.de

*New Journal of Physics* 10 (2008) 093017 (7pp)
Received 14 July 2008
Published 15 September 2008
Online at [http://www.njp.org/](http://www.njp.org/)
doi:10.1088/1367-2630/10/9/093017

**Abstract.** We grew single crystals of $\text{Yb}_2\text{Pt}_6\text{Al}_{15}$ and investigated the magnetic properties of this compound by means of susceptibility $\chi(T)$, specific heat $C(T)$, resistivity $\rho(T)$ and thermoelectric power $S(T)$ measurements. While all properties follow in general the behavior typical for Kondo-lattice systems, $\chi(T)$ and $C(T)/T$ present broad maxima in the $T$ range 17–35 K, which matches nicely the prediction of the Coqblin–Schrieffer model for $J = 7/2$. A large degeneracy of the local moment is also supported by a reduced Kadowaki–Woods ratio. Thus, the analysis of all investigated properties evidences $\text{Yb}_2\text{Pt}_6\text{Al}_{15}$ to be a paramagnetic Kondo-lattice system with the whole $J = 7/2$ multiplet involved in the formation of the Kondo state, a Kondo temperature of the order of 60 K, and a heavy Fermi-liquid ground state with a Sommerfeld coefficient $\gamma_0 = 0.33 \text{ J (mol-Yb)}^{-1} \text{K}^{-2}$ corresponding to a mass enhancement of the order of 30.

---

$^1$ Author to whom any correspondence should be addressed.
1. Introduction

Systems at the crossover between a magnetic and a non-magnetic ground state are attracting considerable attention because of the observation of many interesting phenomena, e.g. the occurrence of quantum critical points (QCPs) or the formation of unconventional metallic and/or superconducting (SC) states [1, 2]. Intermetallic compounds based on Ce or Yb are ideal candidates for studies of these phenomena. The instability of the 4f-shell of these rare earth elements provides an easy way for tuning the magnetism. The onset of superconductivity at the disappearance of the magnetic ordered ground state has been observed quite often in Ce-based compounds [3], but never in Yb-based ones. On the other hand, since the observation of superconductivity with an enhanced $T_c$ in layered CeTIn$_5$ systems [4], it is widely believed that a strong two-dimensional (2D) character is favorable for the development of an unconventional SC state [5]. This suggests that Yb-based quasi 2D intermetallic compounds where Yb is close to a magnetic state are not only interesting systems for the study of 2D QCPs, but also prime candidates for finding an Yb analogue to the unconventional SC states in Ce compounds.

Therefore, we started an investigation of Yb$_2$Pt$_6$Al$_{15}$, since this compound exhibits a promising quasi-2D crystal structure. It is a new member of the rare-earth series RE$_2$Pt$_6$Al$_{15}$ [6], which crystallizes in the hexagonal structure type Sc$_{1.2}$Fe$_4$Si$_{9.8}$. This structure is formed by RE$_2$Al$_3$ layers separated by two Pt–Al layers leading to a large interlayer distance $d_{REL} = c/2 = 8.18\ \text{Å}$ and a large ratio $c/a \approx 4$. Some disorder is suggested in the stacking sequence of the ordering of the RE and Al atoms within the RE$_2$Al$_3$ layers along the hexagonal c-axis [6].

2. Crystal growth and experimental techniques

Single crystals of Yb$_2$Pt$_6$Al$_{15}$ were grown in Al$_2$O$_3$ crucibles using the Bridgman technique. Because the high vapor pressure of Yb can lead to a loss of the element, the Al$_2$O$_3$ crucible was embedded in a sealed tantalum crucible. The crucibles were heated up to 1450 °C and the growth was started with a pulling rate of the order of 1–2 mm h$^{-1}$. Single crystals were cleaved out of the batches and foreign phases were removed from the surface by polishing or cutting the crystals.

Small single crystals were crushed and used for x-ray powder diffraction. The x-ray powder diffraction pattern confirmed the Sc$_{1.2}$Fe$_4$Si$_{9.8}$-type structure with the space group $P6_3/mmc$. A lattice-parameter fit results in the values $a = 4.266(1)\ \text{Å}$ and $c = 16.361(2)\ \text{Å}$ with $c/a = 3.83$. The volume of the unit cell $V_{uc} = 257.88\ \text{Å}^3$ fits into the reported lanthanum
Figure 1. Magnetic susceptibility $\chi(T)$ of Yb$_2$Pt$_6$Al$_{15}$ versus log-$T$ for magnetic field $B = 1$ T applied in-plane and out-of-plane. (a) Inverse susceptibility $\chi^{-1}(T)$ for both directions. The solid lines represent a Curie–Weiss (CW) fit to the data. (b) Comparison between the experimental data and the prediction of the Coqblin–Schrieffer model for $J = 7/2$ and $T_0 = 120$ K plotted as $\chi_{\text{poly}}(T)/\chi_0$ versus $T/T_0$.

The contraction of the rare-earth series RE$_2$Pt$_6$Al$_{15}$ [6]. Microprobe analyses showed a ratio Yb : Pt : Al $\approx$ 8.6 : 25.3 : 66.1 for the main phase in good agreement with the theoretical value 8.7 : 26.1 : 65.2. The single crystals were oriented with Laue backscattering, which indicates that they grow mainly as platelets perpendicular to the $c$-axis.

Standard experimental techniques were used in the commercial equipment MPMS and PPMS (Quantum Design) for the measurements of the magnetic susceptibility $\chi_{dc}$, the specific heat $C_p$, the electrical resistivity $\rho$ and the thermoelectric power $S$.

3. Results and discussion

Figure 1 shows the temperature dependence of the magnetic susceptibility $\chi_{dc}(T)$ versus log-$T$ in the range $2 < T < 400$ K for a magnetic field $B = 1$ T applied along the $c$-axis and within the basal plane on two different single crystals. At high temperatures $\chi_{dc}^{-1}(T)$ reveals a CW behavior for both directions, described by an effective magnetic moment $\mu_{\text{eff}} = 4.08 \mu_B$ and a Weiss temperature $\Theta_p = -8$ K for $B \perp c$ and $\mu_{\text{eff}} = 4.12 \mu_B$, $\Theta_p = -30$ K for $B \parallel c$. The $\mu_{\text{eff}}$ values are close to that of free Yb$^{3+}$ ions ($\mu_{\text{eff}} = 4.54 \mu_B$) suggesting an Yb valence close to 3+. At low temperatures, a clear maximum develops in $\chi_{dc}(T)$ for both directions at $T_{\text{max}}(\parallel c) = 35$ K and $T_{\text{max}}(\perp c) = 25$ K. Toward lower temperature $\chi_{dc}(T)$ levels out with a value $\chi_0(\parallel c) = 2.9 \times 10^{-7}$ m$^3$ mol$^{-1}$ and $\chi_0(\perp c) = 4 \times 10^{-7}$ m$^3$ mol$^{-1}$ showing only a small anisotropy with $c$ as the hard magnetic direction. Such a maximum in $\chi_{dc}(T)$ is typical for a Kondo-lattice system with a larger characteristic energy, i.e. at the border to the intermediate-valent state. Then not only the crystal electric field (CEF) ground state doublet, but also some of the excited CEF states or even the whole multiplet is involved in the physics at low $T$.
Figure 2. Temperature dependence of the $4f$ specific heat $\Delta C_{4f}/T$ versus log-$T$. A hump develops at 17 K. Dotted line represents the prediction of the Coqblin–Schrieffer model for $J = 7/2$ and $T_0 = 120$ K. (a) $\rho(T)/\rho_{300}$K plotted in a log-$T$ scale down to 0.4 K with $I \parallel c$ and $I \perp c$. (b) Thermoelectric power $S(T)$ for heat current $q \perp c$ in the temperature range $2 < T < 200$ K.

leading to a maximum in $\chi(T)$ and in $C(T)/T$ [7]. For a more precise analysis of our data, we first calculated the powder average of the susceptibility $\chi_{\text{poly}}(T) = 2/3 \chi_{ab}(T) + 1/3 \chi_c(T)$ and then scaled it with the value at low $T$. We then compared this normalized $\chi_{\text{poly}}(T)/\chi_0$ with the theoretical results for the Coqblin–Schrieffer model of Rajan [7]. An excellent agreement was obtained assuming the whole multiplet $J = 7/2$ of Yb$^{3+}$ to be involved and a scaling energy $T_0 = 120$ K (figure 1, inset (b)). This scaling energy can be converted into a Kondo temperature (in its high $T$ definition) according to $T_K = T_0 \cdot \frac{2}{W_{7/2}} \cdot \frac{\pi}{(2J + 1)}$, which for $J = 7/2$, $W_{7/2} = 0.6783$ [8] gives $T_K = 64$ K.

The presence of a Kondo lattice with a large ground-state degeneracy is confirmed by the specific heat results. In figure 2, we plot $C_4(T)/T$ versus log-$T$, where $C_4(T)$ was obtained by subtracting $C(T)$ of La$_2$Pt$_6$Al$_{15}$ from the measured $C(T)$ of Yb$_2$Pt$_6$Al$_{15}$. $C_4(T)/T$ increases with decreasing $T$ below 50 K, passes through a broad but well-defined maximum at 20 K and then merges below 6 K in a constant value, corresponding to a total Sommerfeld coefficient $\gamma_0 = 0.33$ J (mol-Yb)$^{-1}$ K$^{-2}$ after addition of $\gamma_{\text{La}} = 12$ mJ (mol-La)$^{-1}$ K$^{-2}$ as determined for the non-f-electrons in La$_2$Pt$_6$Al$_{15}$. We again compare our experimental data with the prediction of Rajan for the Coqblin–Schrieffer model for $J = 7/2$ and $T_0 = 120$ K (as determined from the analysis of $\chi(T)$). Both data show a very similar $T$ dependence, the theoretical curve being slightly lower than the experimental one. The difference at low $T$ suggests that the real characteristic energy scale of Yb$_2$Pt$_6$Al$_{15}$ is slightly lower. The difference at higher $T$ has to be attributed to a slight overestimation of the phonon contribution, because the entropy obtained by integrating $C_4(T)/T$ reaches 18.7 J (mol-Yb K)$^{-1}$ at 50 K, which is already slightly larger than the high-$T$ limit $R \ln 8 = 17.3$ J (mol K)$^{-1}$ expected for a $J = 7/2$ state. This evidences a slight overestimation of $C_4$ at high $T$. Nevertheless, this comparison also gives strong evidence for a Kondo-lattice system where the full Yb$^{3+}$ multiplet is involved and with a quite large...
characteristic energy. A further estimate for the characteristic energy can be gained from the entropy, by taking twice the temperature $T = 21 \text{K}$ at which half of the full entropy $R \ln 8$ is recovered, which leads to $T^* = 42 \text{K}$, slightly smaller than $T_K = 64 \text{K}$ estimated from the analyses of $\chi(T)$ and $C_{4f}(T)$ using the Coqblin–Schrieffer model. However, because of the overestimation of $C_{4f}(T)$ mentioned above, this $T^*$ value underestimates the characteristic energy.

The involvement of the full Yb $J = 7/2$ multiplet in the low-$T$ physics despite $T_K$ being not so large implies a rather weak CEF splitting, which seems to be in contradiction with the quasi-2D crystal structure. However, a closer inspection of the structure evidences a quite isotropic local surrounding of Yb [6]. The coordination polyhedra of Yb is formed by 9 Al at almost the same distance, 3 in the basal plane and 6 in the planes above and below, and further 2 Al and 4 Pt at a slightly larger distance also above and below the basal plane. With such a large and isotropic coordination, a weak CEF is not surprising.

The temperature dependence of the resistivity also supports the presence of Kondo-type interactions with a larger characteristic energy scale. In figure 2(a), the temperature dependence of the normalized electrical resistivity $\rho(T)/\rho_{300K}$ is plotted versus log-$T$. While the absolute values of $\rho$ are quite anisotropic, with $\rho_{\parallel\text{c}}(300 \text{K}) = 235 \mu\Omega\text{cm} \gg \rho_{\perp\text{c}}(300 \text{K}) = 50 \mu\Omega\text{cm}$, the $T$ dependencies are similar. Below 300 K $\rho(T)$ decreases with $T$ for both current $(I)$ directions. Then the onset of Kondo-type scattering leads to a slight increase for $I$ in the basal plane while for $I$ along $c$ one observes only a pronounced shoulder. With further decreasing $T$ the coherent Kondo state forms leading to a pronounced drop in $\rho(T)$. Below 10 K for $I \perp c$ and 5 K for $I \parallel c$, we observe Fermi liquid behavior with $\rho(T) = \rho_0 + A \cdot T^2$. Fits to the low-$T$ data give $A = 0.069 \mu\Omega\text{cm} K^{-2}$ and $\rho_0 = 1.65 \mu\Omega\text{cm}$ for $I \perp c$ and $A = 0.59 \mu\Omega\text{cm} K^{-2}$ and $\rho_0 = 10.2 \mu\Omega\text{cm}$ for $I \parallel c$, respectively. Thus, while the absolute values of $\rho(T)$ are quite anisotropic, the residual resistivity ratio $\rho_{300K}/\rho_0 = \text{RRR} = 30$ for $I \perp c$ and RRR $= 23$ for $I \parallel c$ are similar. The rather large RRR values along both directions indicate a quite high perfection of the structural lattice, in contrast to the disorder suggested in earlier reports for other RE$_2$Pt$_6$Al$_{15}$ systems. The higher absolute values along the $c$-direction despite a similar RRR are therefore likely related to poor conducting small angle grain boundaries within stacks of platelets.

The thermoelectric power $S(T)$ presented in figure 2(b) was measured with the heat current $q$ perpendicular to the $c$-axis. It exhibits large absolute values typical for heavy-fermion systems with a negative sign over the whole investigated temperature range as expected for an Yb-based Kondo-lattice system. A broad maximum develops at a temperature of $T \approx 35 \text{K}$ with a magnitude of $S(T) \approx -70 \mu\text{V}\text{K}^{-1}$. A single maximum in $S(T)$ of a Kondo-lattice system is expected when the Kondo energy scale is close to the CEF splitting. The temperature of the maximum in $S(T)$ is compatible with a Kondo scale of the order of 60 K. Thus, this result again supports the picture of a Kondo lattice with a large characteristic energy and with the whole multiplet being involved in the low-$T$ physics. At low $T$, the thermoelectric power $S(T)$ extrapolates to zero with an enhanced initial slope $S/T = -3.2 \mu\text{V}\text{K}^{-2}$ in accordance with a heavy Fermi-liquid ground state.

4. Conclusions

We have grown single crystals of Yb$_2$Pt$_6$Al$_{15}$ and investigated the magnetic properties of this compound by means of susceptibility, specific heat, resistivity and thermopower measurements.

New Journal of Physics 10 (2008) 093017 (http://www.njp.org/)
While at high $T$ the weakly anisotropic $\chi(T)$ follows a CW law with an effective moment close to that of a free Yb$^{3+}$ ion, at lower $T$ we observe broad maxima around $T = 35$ K for field along the hard $c$-axis and $T = 25$ K for field in the easy basal plane. A related maximum is also observed in $C_{4f}(T)/T$ around $T = 17$ K, while $C(T)/T$ levels out below 6 K in a constant Sommerfeld coefficient $\gamma_0 = 0.33$ J (mol-Yb)$^{-1}$ K$^{-2}$. This corresponds to a mass enhancement of about 30 when comparing with $\gamma_0 = 12$ mJ (mol-La)$^{-1}$ K$^{-2}$ in La$_2$Pt$_6$Al$_{15}$. The $T$ dependence as well as the maxima in $C_{4f}(T)$ and $\chi(T)$ can be very well described by the Coqblin–Schrieffer model for $J = 7/2$ and a scaling temperature $T_0 = 120$ K corresponding to a Kondo temperature $T_K = 64$ K. The resistivity presents a $T$ dependence typical of a Kondo-lattice system, with a maximum and a shoulder in $\rho(T)$ at $T \cong 50$ K for $I$ in the basal plane and along $c$, respectively, followed by a strong decrease toward low $T$ ending in a power law $\rho(T) = \rho_0 + A \cdot T^2$ at lowest $T$. We did not observe any evidence for a magnetic phase transition.

The enhanced, temperature independent $C(T)/T$, $\chi(T)$ and $S(T)/T$ observed at low $T$ as well as $\rho(T)$ increasing with $T^2$ indicate a well-defined heavy Fermi-liquid ground state. With $\gamma_0 = 0.33$ J (mol-Yb)$^{-1}$ K$^{-2}$, $\chi_{poly}(0) = 3.7 \times 10^{-7}$ m$^3$ (mol-Yb)$^{-1}$ and the experimentally determined $\mu_{\text{eff}} = 4.1 \mu_B$, the Wilson ratio $R_W = \chi_0/\gamma_0 \cdot (\pi \cdot k_B/\mu_{\text{eff}})^2/\mu_0$ amounts to $R_W = 1.15$ which matches exactly the value $8/7$ expected for a $J = 7/2$ Kondo system. Taking for $A$ the more reliable value for the current along the basal plane gives a Kadowaki–Woods ratio $A/\gamma^2 = 6.3 \times 10^{-7} \mu\Omega$ cm (mol-Yb K mJ$^{-1}$)$^2$. This is a factor of 16 smaller than the value $R_{KW} \cong 1.0 \times 10^{-5} \mu\Omega$ cm (mol K mJ$^{-1}$)$^2$ expected for a standard Fermi liquid [9], but in good accordance with the value $R_{KW}/(N(N-1)/2)$ proposed by H Kontani [10] for Kondo systems with a large degeneracy $N$. For a $J = 7/2$ state, $R_{KW}/(N(N-1)/2)$ amounts to $3.6 \times 10^{-7} \mu\Omega$ cm (mol K mJ$^{-1}$)$^2$, just a factor 1.7 smaller than our experimental $A/\gamma^2$ value for Yb$_2$Pt$_6$Al$_{15}$. The relation $A/\gamma^2 = R_{KW}/(N(N-1)/2)$ was confirmed by Tsujii et al [11] who performed a general analysis of a large variety of heavy-fermion compounds with degeneracy ranging from $N = 2$ to 8. They observed a clear dependence of the $A/\gamma^2$ ratio on the degeneracy $N$ and proposed a universal scaling for Kondo-lattice systems with a modified Kadowaki–Woods ratio $A/\tilde{\gamma}^2 \approx 1 \times 10^{-5} \mu\Omega$ cm (mol K mJ$^{-1}$)$^2$ where the coefficients $A = A/[1/2 N(N-1)]$ and $\tilde{\gamma} = \gamma/[1/2 N(N-1)]$ are normalized with respect to the degeneracy. Recently a whole class of Yb-based heavy Fermion compounds with composition YbT$_2$Zn$_{20}$ and showing a large orbital degeneracy was also confirmed to follow this general scaling [12, 13].

Thus, all the results evidence Yb$_2$Pt$_6$Al$_{15}$ to be a paramagnetic Kondo-lattice system with the whole $J = 7/2$ multiplet involved in the formation of the Kondo state, a Kondo temperature of the order of 60 K, and a heavy Fermi-liquid ground state with $\gamma_0 = 0.33$ J (mol-Yb)$^{-1}$ K$^{-2}$ corresponding to a mass enhancement of the order of 30.

Since application of pressure shifts an Yb system toward a full trivalent magnetic state, it will be interesting to investigate how in Yb$_2$Pt$_6$Al$_{15}$ the high local moment degeneracy and the 2D character of the structure, which shall become strongly relevant for the intersite magnetic exchange, affect the onset of magnetic order at the (quantum) critical point.

Acknowledgments

We thank Dr U Burkhardt, Dr R Ramlau and Ms P Scheppan for microprobe studies. Additional thanks go to Dr Y Prots for his stimulating discussions about the structural properties. SH, MEM, MN and CG acknowledge support from COST P16 action.
References

[1] Gegenwart P, Si Q and Steglich F 2008 Nat. Phys. 4 186
[2] Stewart G R 2001 Rev. Mod. Phys. 73 797
[3] Yuan H Q, Grosche F M, Deppe M, Geibel C, Spann G and Steglich F 2003 Science 302 2104
[4] Movshovich R, Jaime M, Thompson J D, Petrovic C, Fisk Z, Pagliuso P G and Sarrao J L 2001 Phys. Rev. Lett. 86 5152
[5] Monthoux P and Lonzarich G G 2001 Phys. Rev. B 63 054529
[6] Niermann J and Jeitschko W 2004 Z. Anorg. Allg. Chem. 630 361
[7] Rajan V T 1983 Phys. Rev. Lett. 51 308
[8] Rasul J W and Hewson A C 1984 J. Phys. C: Solid State Phys. 17 2555
[9] Kadowaki K and Woods S B 1986 Solid State Commun. 58 507
[10] Kontani H 2004 J. Phys. Soc. Japan 73 515
[11] Tsujii N, Kontani H and Yoshimura K 2005 Phys. Rev. Lett. 94 057201
[12] Torikachvili M S, Jia S, Mun E D, Hannahs S T, Black R C, Neils W K, Martien Dinesh, Bud’ko S L and Canfield P C 2007 Proc. Natl Acad. Sci. USA 104 9960
[13] Canfield P C, Jia S, Mun E D, Bud’ko S L, Samolyuk G D and Torikachvili M S 2008 Physica B 403 844