Magnetic behaviour of the intermetallic compound YbCo$_2$Si$_2$

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New Journal of Physics 13 (2011) 083024 (13pp)
Received 13 January 2011
Published 16 August 2011
Online at http://www.njp.org/
doi:10.1088/1367-2630/13/8/083024

Abstract. We report on a comprehensive study of the magnetic properties of YbCo$_2$Si$_2$ investigated using a single crystal. All data confirm a stable trivalent Yb state carrying a well-localized 4f moment, as well as a non-magnetic Co state. We observed a sizeable anisotropy in the magnetic susceptibility $\chi(T)$, with the moment in the basal plane twice as large as that along the $c$-direction. Combining this result with published neutron-scattering data puts strong constraints on the values of the crystalline electrical field (CEF) parameters. Our calculation yields one possible solution with a $\Gamma_7$ CEF ground state. A peak in the easy-plane susceptibility at low temperatures demonstrates that the previously reported magnetic ordering at $T_N \approx 1.65$ K is of antiferromagnetic (AFM) nature. In addition, we observed sharp anomalies in all measured properties, evidencing a further phase transition at $T_L \approx 0.9$ K, found to be first order, which corresponds to a change in the AFM structure. Both transitions are suppressed by a magnetic field $B = 2$ T. Below $T_L$, the specific heat, $C^{4f}$, shows a well-defined $T^3$ dependence, as expected for AFM spin-wave excitations. Surprisingly, the resistivity, $\rho(T)$, also shows a $T^3$ power law in the same temperature range, instead of the expected $T^5$. This discrepancy might be related to more complex magnetic interactions, as also evidenced by the multiple transitions. By extending the linear fit of $C^{4f}/T$ versus $T^2$ to $T = 0$, the Sommerfeld coefficient $\gamma_0 = 0.13$ J K$^{-2}$ mol$^{-1}$ is extracted. Although magnetism in YbCo$_2$Si$_2$ is dominated by the RKKY exchange interaction, the enhanced $\gamma_0$ and a weak Kondo-like increase in $\rho(T)$ indicate the presence of some interactions between the conduction electrons and the 4f moments.

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1. Introduction

The recent development of the metallic-flux technique for growing single crystals of high quality has broadened the number of compounds available for low-temperature investigations to those containing elements with a high vapour pressure at high temperatures (low boiling point) [1, 2]. A typical example is Yb-based heavy-fermion (HF) intermetallic materials with ytterbium evaporating at 1200 °C, and therefore below the melting point of many transition metals. These systems can be considered to be the 4f-hole analogue of the Ce ones [3] and the low-temperature physics is usually strongly dependent on the Yb valence; that is, they do not very often show magnetic ordering due to the fact that in many of these materials the Yb ion has a valence state close to 2+, resulting in a filled 4f shell with 14 electrons. In compounds with a valence close to 3+, with one hole in the f shell, the situation is different. As in Ce-based systems, the ground state properties are sensitive to the degree of hybridization between the 4f electrons (holes) and the conduction 3d electrons through the Kondo effect, which tends to screen the rare-earth magnetic moment, leaving a paramagnetic ground state with enhanced quasi-particle masses, and the Ruderman–Kittel–Kasuya–Yoshida (RKKY) exchange interaction, which causes magnetic ordering [4]. The interplay between these two phenomena is known to be the basis for understanding the physics of HF materials [5].

Among this class of compounds, a system of particular interest is YbRh$_2$Si$_2$, which has attracted considerable attention as its low-temperature properties are influenced by the presence of an unconventional quantum critical point (QCP) [6]. It shows weak antiferromagnetic (AFM) ordering at a very low temperature $T_N = 0.07$ K, since the 4f electrons are strongly hybridized with a Kondo temperature $T_K \approx 25$ K [7]. A detailed study of the magnetic fluctuations at the QCP is hindered by the lack of knowledge of the magnetically ordered structure, which is due to the very low $T_N$ and the unexpectedly small ordered moment ($10^{-3} \mu_B/\text{Yb}$) [8]. One way to overcome this problem is to stabilize the AFM order by applying hydrostatic or chemical pressure, thus enhancing $T_N$ and the size of the ordered moment. This has been successfully realized by substituting isoelectronic Co for Rh [9]: in fact, the whole series Yb(Rh$_{1-x}$Co$_x$)$_2$Si$_2$ crystallizes in the tetragonal ThCr$_2$Si$_2$ structure (space group I4/mmm) [10]. The complete substitution of Rh by Co eventually leads to the stoichiometric compound YbCo$_2$Si$_2$, which is easier to study, since its $T_N = 1.7$ K [11] with almost localized 4f electrons.
and $T_K \ll T_{\text{RKKY}}$ [10]. A knowledge of its magnetic ordered structure will possibly give some insights into the magnetic structure in YbRh$_2$Si$_2$, provided that the Yb Kramers-doublet ground state is the same in both compounds.

YbCo$_2$Si$_2$ has already been investigated by several groups (see [11–14]), but all of these studies are based on polycrystalline material and do not focus on the low-temperature properties. The first evidence of magnetic order in YbCo$_2$Si$_2$ was given by Hodges in $^{170}$Yb Mössbauer spectroscopy experiments: Hodges found magnetic order below 1.7 K with the easy magnetization close to the basal plane and a saturated moment of $1.4 \mu_B$/Yb [11]. He also succeeded in explaining his results well, taking into account an Yb$^{3+}$ valence state that experiences a tetragonal crystalline electrical field (CEF), resulting in a Kramers doublet $\Gamma_7$ as the ground state. Inelastic neutron scattering (INS) experiments have unambiguously demonstrated that Yb is trivalent at high temperatures and that the doublet ground state is separated from the excited doublets at 4, 12.5 and 30.5 meV [14]. The authors of these measurements proposed a solution for the CEF scheme that does not reproduce the saturation magnetization. On the other hand, the solution provided by Hodges, although mostly correct, gives the second excited doublet at an incorrect energy of 18.4 meV. Finally, Kolenda et al measured the magnetic susceptibility up to 300 K, showing, by applying a Curie–Weiss law, that Co is not magnetic in this material [13]. This feature is a characteristic of the whole Yb(Rh$_{1-x}$Co$_x$)$_2$Si$_2$ series [10].

In the course of the synthesis of Yb(Rh$_{1-x}$Co$_x$)$_2$Si$_2$ single crystals, we were able to grow YbCo$_2$Si$_2$ single crystals of good quality. Recently, a second AFM phase transition was observed in these crystals at a temperature $T_L = 0.9$ K by means of magnetization and resistivity measurements and this transition seems to be of first order [15, 16]. Both transition temperatures are suppressed by a field larger than 2 T, and the $B$–$T$ magnetic phase diagram becomes very complex. Moreover, preliminary powder neutron diffraction studies, conducted in the intermediate and low-$T$ AFM states, have detected magnetic peaks and suggested that the magnetic propagation vector (not determined yet) changes its periodicity between the two phases [17].

In this paper, we present the crystallographic analysis and a complete study of the low-temperature properties, e.g. the magnetic susceptibility ($\chi = M/B$), electrical resistivity ($\rho$) and specific heat ($C^{4f}$), of such YbCo$_2$Si$_2$ crystals. We propose a correct solution for the CEF scheme of Yb ions, which matches all existing data as well as the magnetic anisotropy [16], and seems to be similar to that considered for YbRh$_2$Si$_2$ [18, 19]. The phase transition at $T_L$ is confirmed to be of first order by specific heat measurements which, together with the resistivity, show strong spin-wave excitations below $T_L$ and a partial hybridization of the 4f electrons.

2. Experimental techniques

The solvent growth technique with in flux was used for the synthesis of the YbCo$_2$Si$_2$ single crystals [1, 2]. The starting material with an atomic ratio of Yb : Co : Si : In of 3 : 2 : 2 : 24 was placed in an Al$_2$O$_3$ crucible, which was inserted in a second larger crucible of tantalum sealed in argon atmosphere. This prevented the evaporation of Yb during the crystal growth process at high temperatures. The crucible was heated to 1500 °C, which was followed by slow cooling to 1000 °C within 150 h. The single crystals were retrieved from the in flux using hydrochloric acid (HCl) for chemical etching since the crystals themselves showed no reactivity to HCl.
A crystallographic analysis of the single crystals was performed to compare the obtained results with already existing data about polycrystalline material. Laue backscattering reflection pictures, as well as x-ray powder diffraction spectra, were taken using a STOE-diffractometer system, Stadip MP, combined with the Polaroid XR-7 system for Laue x-ray photographs. Furthermore, several crystals of each batch were prepared by surface polishing in order to perform energy dispersive x-ray spectra on a scanning electron microscope (Philips XL30) with a Si(Li)-x-ray detector. This examination allowed firstly the detection of possible contaminating elements and secondly an accurate investigation of the actual stoichiometry of different single crystals.

Measurements of the electrical resistivity and specific heat were carried out in a 9 T ³He physical properties measurement system (Quantum Design), allowing an experimental analysis in the temperature range $0.35 \leq T \leq 300$ K. For the electrical resistivity measurements, a standard four-terminal ac technique was utilized, whereas for the specific heat the $2\tau$-relaxation time method (see [20]) was used. Measurements of angle-dependent magnetic susceptibility were carried out in a 5 T SQUID (Quantum Design) with a rotator inset. For the low-temperature measurements (down to 0.5 K), we used a ³He option for the SQUID (iQuantum Corporation).

3. Results

3.1. Crystallographic analysis

Laue back-scattering reflection pictures and x-ray powder diffraction spectra were taken on the synthesized stoichiometric single crystals of YbCo₂Si₂. The obtained outcomes are shown in figure 1. On the top right of figure 1, the single crystal is displayed on millimetre paper to show its dimensions (volume $\approx 5$ mm³ and mass $\approx 40$ mg). Interestingly, almost all large crystals have a square shape with sides parallel to the crystallographic direction [100], which makes them easy to align.

The x-ray diffraction pattern obtained from the powder of three crushed single crystals is depicted on the bottom of the same figure: the numbers marking the different reflection peaks indicate that the single crystals show no indications of impurity phases. The crystal structure is confirmed to be tetragonal (ThCr₂Si₂-type) with space group I4/mmm. The least-square fitting procedure provides the crystallographic parameters $a = 3.852(1)$ Å, and $c = 9.689(2)$ Å, close to those reported by Rossi et al [12] and Kolenda and Szytula [13] for polycrystalline materials.

Another proof of the good quality of the synthesized single crystals is the Laue backscattering reflection picture. The reflection peaks are pin sharp rather than blurred, leading to the conclusion that the single crystal is not affected by disorder or impurity effects. However, a slight local disorder due to the statistical Co–Si transposition cannot strictly be ruled out. The experimental results on the physical properties presented in the following show, however, that possible local disorder can be neglected.

3.2. Magnetocrystalline anisotropy: crystalline electrical field

The magnetic properties of rare-earth compounds commonly exhibit a strong anisotropy due to the effect of the CEF on the orbital part of the total angular momentum $J$. In a tetragonal point symmetry, the $J = 7/2$ multiplet of the Yb$^{3+}$ ion (with $g_J = 8/7$) is split by the CEF into two
Kramers doublets, \( \Gamma_6 \) and \( \Gamma_7 \). In such a case, the CEF Hamiltonian can be written as

\[
H_{\text{CF}} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_4^4 O_4^4 + B_6^0 O_6^0 + B_6^4 O_6^4,
\]

where \( B_m^n \) are the CEF parameters and \( O_m^n \) are the Stevens operators [21]. Adding the Zeeman term, the global Hamiltonian becomes

\[
H = H_{\text{CF}} + g J \mu_B B,
\]

where \( \mu_B \) is the Bohr magneton and \( B \) the magnetic field. As mentioned in the introduction, two solutions to equation (1) have already been given in [11] and [14], but both are incorrect: the solution in [11] gives the second excited doublet at an incorrect energy of 18.4 meV, compared to what has been measured by INS (\( \Delta_1 = 4 \text{ meV}, \Delta_2 = 12.5 \text{ meV} \) and \( \Delta_3 = 30.5 \text{ meV} \)), whereas that in [14] provides an incorrect ground state with a saturation moment in the basal plane of \( 2.25 \mu_B \), substantially exceeding the value \( 1.4 \mu_B \) observed experimentally in Mössbauer [11] and magnetization [15] measurements. The fact of having a single crystal available offers the possibility of measuring directly the magnetic crystalline anisotropy, which gives a further constraint for solving equation (1); this strongly limits the allowed solutions.

The uniform magnetization \( M \) has been measured at different fields between 0.5 and 300 K. The inverse of the magnetic susceptibility \( \chi^{-1} = B/M \) is shown in figure 2. The \( c \) crystallographic axis is clearly the magnetic hard axis of the ground state (cf the right

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**Figure 1.** Top right: photograph of an YbCo\(_2\)Si\(_2\) single crystal on millimetre paper. Bottom right: x-ray backscattering diffraction pattern from the powder diffraction of three crushed single crystals. Left: Laue backscattering reflection picture.
Figure 2. Left: inverse of the magnetic susceptibility $\chi = M / B$ with $B$ parallel (blue circles) and perpendicular (red squares) to the tetragonal $c$-axis plotted as a function of $T$. The straight black lines are corresponding fits with the Curie–Weiss law. Right: magnetic anisotropy in YbCo$_2$Si$_2$, due to CEF splitting, measured at 2 K in 0.1 T.

The system is paramagnetic for $T \geq 2$ K, and at high temperature, $\chi^{-1}$ follows a Curie–Weiss behaviour with effective moment $\mu_{\text{eff}} \approx (4.7 \pm 0.1) \mu_B$, close to that of the free Yb$^{3+}$ ion. Cobalt is indeed not magnetic in YbCo$_2$Si$_2$, or in the whole series Yb(Rh$_{1-x}$Co$_x$)$_2$Si$_2$ [10]. The paramagnetic Weiss temperatures $\theta_{ab}^W$ and $\theta_{c}^W$, along both the principal crystallographic axes, can be expressed on the basis of molecular field theory [22] as a function of the first CF parameter $B_2^0$, which is therefore a measure of the strength of the magnetocrystalline anisotropy:

$$B_2^0 = \left( \theta_{ab}^W - \theta_{c}^W \right) \frac{10k_B}{3(2J - 1)(2J + 3)}.$$  

Using the experimental values $\theta_{ab}^W = -4$ K and $\theta_{c}^W = -160$ K extracted from the fits shown in figure 2, we find that $B_2^0 = 0.75$ meV, a value slightly larger than that deduced by Hodges (0.71 meV),\(^3\) but very close to that proposed by Goremychkin and Osborn (0.76 meV) [14]. From equation (1) we can derive the following two equations,

$$\begin{vmatrix} A & E \\ E & D \end{vmatrix} \Gamma_6 = \epsilon_6 \Gamma_6,$$

$$\begin{vmatrix} B & F \\ F & C \end{vmatrix} \Gamma_7 = \epsilon_7 \Gamma_7,$$

where the diagonal energies $A$, $B$, $C$ and $D$ are linear combinations of the parameters $B_2^0$, $B_4^0$ and $B_6^0$, and the off-diagonal energies $E$ and $F$ are linear combinations of $B_4^4$ and $B_6^4$. The

\(^3\) We adapt, to the common definition of $B_n^m$, the factor $\alpha_J = 2/(9 \times 7)$, which differs from the definition used by Hodges [11].
Figure 3. Schematic crystal-field level scheme proposed for YbCo$_2$Si$_2$.

The values of $\alpha$ and $\alpha'$ can be extracted from magnetization [15, 23] or ESR [24] experiments: these measurements lead to very similar results for the anisotropic $g$-factor of the CEF ground state doublet, with $g_{ab} \approx 2.8$ and $g_c \approx 1.4$. This restricts the solutions of the possible CEF ground state wave functions to one $|\Gamma_6\rangle$ with $\alpha \approx 54^\circ$ or one $|\Gamma_7\rangle$ with $\alpha' \approx 27^\circ$. Assuming $|\Gamma_7\rangle$ to be the ground state and a CEF scheme, as displayed in figure 3, we can derive $S_7$, $M_7$ and $F$ from equations (5), taking into account the CEF excitation energies determined in neutron-scattering experiments

$$\Delta_1 = 4 \text{ meV}, \quad \Delta_2 = 12.5 \text{ meV}, \quad \Delta_3 = 30.5 \text{ meV},$$

$$\Delta_6 = \Delta_3 - \Delta_1 = 2\sqrt{S_6^2 + E^2} = 26.5 \text{ meV},$$

$$\Delta_7 = \Delta_2 = 2\sqrt{S_7^2 + F^2} = 12.5 \text{ meV},$$

$$M_7 = \frac{\Delta_7}{2} - \frac{\Delta_1 + \Delta_2 + \Delta_3}{4} = -5.5 \text{ meV},$$

where

$$|\Gamma_6,1\rangle = \cos(\alpha)|\pm 1/2\rangle + \sin(\alpha)|\mp 7/2\rangle$$

$$|\Gamma_6,2\rangle = \sin(\alpha)|\pm 1/2\rangle - \cos(\alpha)|\mp 7/2\rangle$$

$$|\Gamma_7,1\rangle = \cos(\alpha')|\pm 3/2\rangle - \sin(\alpha')|\mp 5/2\rangle$$

$$|\Gamma_7,2\rangle = \sin(\alpha')|\pm 3/2\rangle + \cos(\alpha')|\mp 5/2\rangle.$$
Table 1. Crystal field parameters for YbCo$_2$Si$_2$ in meV.

| $B_0^2$ | $B_0^4$ | $B_0^6$ | $B_4^2$ | $B_4^4$ | $B_4^6$ |
|---------|---------|---------|---------|---------|---------|
| 0.75    | $6.162 \times 10^{-3}$ | $5.022 \times 10^{-2}$ | $-1.159 \times 10^{-4}$ | $2.242 \times 10^{-4}$ |

and therefore the $B_0^2$, $B_0^4$ and $B_0^6$ parameters. The $B_4^2$ and $B_4^4$ parameters can be calculated from the $E$ and $F$ values. In summary, the measured $g$ factors, the $B_0^2$ derived above and the three CEF excitation energies give a sufficient number of constraints for equation (1) to fix all $B_m^n$ CEF parameters. We found no solution with a $\Gamma_6$ as the ground state. The CEF parameters corresponding to this solution are listed in table 1. The ground state wave function is

$$|\Gamma_{1,1}\rangle = 0.891|\pm 3/2\rangle - 0.454|\mp 5/2\rangle.$$  \hspace{1cm} (7)

This ground state doublet leaves saturation moments of $\mu_{ab} = 1.6 \mu_B$ and $\mu_c = 0.77 \mu_B$ along the basal plane and the $c$-axis, respectively. This requires a small negative polarization of the Co 3d electrons to fit exactly the experimental results [15, 23]. A similar ground state is currently considered in YbRh$_2$Si$_2$ [18, 19, 25].

3.3. Magnetic susceptibility at low temperatures

The temperature dependence of the magnetization $\chi(T) = M(T)/B$ reveals clear anomalies below 2 K, indicative of AFM ordering [15]: on the left of figure 4, $\chi(T)$ is plotted for external fields 0.1 and 2 T, perpendicular to the magnetic hard direction $c$, whereas on the right side $\chi(T)$ is plotted for $B \parallel c$. With $B = 0.1$ T the susceptibility displays a clear feature at $T_{N}^{ab} = 1.65$ K and $T_{N}^{c} = 1.64$ K (very close to the Néel temperature found by Hodges), indicating the onset of
AFM order. Further decrease of temperature reveals a second anomaly at $T_L = 0.91$ K, in the form of a sharp drop in the susceptibility with $B$ along both directions. This behaviour points to a possible first-order phase transition into a second AFM phase. Direct evidence of the first-order character of the transition arises from the observation of latent heat in the specific heat measurements presented in section 3.5. The Mössbauer data of Hodges seemingly did not reveal any transition at $T_L$. Preliminary neutron diffraction experiments indicate that the ground state of YbCo$_2$Si$_2$ below $T_L$ is AFM with a commensurate propagation vector $Q$, whereas between $T_L$ and $T_N$ the magnetic reflections point to a longer periodicity of $Q$ [17, 26], suggesting a more complex magnetic structure. Moreover, detailed magnetization and magnetoresistance measurements on crystals of the same batch have been carried out in all three crystallographic directions [100], [110] and [001]. These show a rather complex $B$–$T$ phase diagram and a pronounced basal plane anisotropy [16, 23].

At an external magnetic field of 2 T the AFM order is completely suppressed (at least down to $T = 0.5$ K in the results presented here). The dc-susceptibility remains at very high values since the magnetization is close to its saturation value.

3.4. Electrical resistivity

The temperature dependence of the normalized electrical resistivity $\rho(T)/\rho_{300K}$ of YbCo$_2$Si$_2$ with current $j \parallel ab$ and $B = 0$ T is plotted in figure 5 on a logarithmic scale down to 0.35 K. The two phase transitions are clearly visible at $T_N = 1.65$ K and $T_L = 0.91$ K. These temperatures correspond quite well to those found in the susceptibility. While at $T_N$ the resistivity starts decreasing towards lower values, at $T_L$ it shows a steep drop. Below $T_L$ a striking $T^3$ dependence of the resistivity is observed (see the inset of figure 5). For AFM magnons one expects a $T^3$ dependence of the specific heat (as it is indeed observed, see below), but a $T^5$ power law in the
Figure 6. (a) Electronic specific heat of YbCo$_2$Si$_2$ in zero field, showing the first-order phase transition at $T_L \approx 0.91$ K and the AFM onset at $T_N \approx 1.65$ K (black circles). At $B = 2$ T the transitions are suppressed (blue rectangles). Below $T_L$, $C^{4f}(T)$ follows a $T^3$ dependence, shown in the inset. (b) Temperature versus time relaxation curve across the first-order phase transition at $T_L$. The changes of slope depicted by arrows are due to latent heat. This curve was used to directly calculate the specific heat around $T_L$ (red line in (a)). (c) Electronic entropy $S(T)$ calculated by integrating $C^{4f}/T$ over $T$.

resistivity. The origin of this discrepancy is not yet clear. It indicates a more complex picture than a textbook one, in agreement with the multiple AFM transitions. A fit of the data below $T_L$ gives a residual resistivity $\rho_{0K} = 13 \mu\Omega\text{cm}$, leaving a residual resistivity ratio $\rho_{300K}/\rho_{0K} \approx 2$ for this sample.

Looking at the high-temperature region in figure 5, an arrow points to a broad and not very pronounced hump. A possible explanation for this weak anomaly is the excitation into the first CEF doublet, which is located at 4 meV. The overall behavior of the electrical resistivity in the high-temperature region can be described in the frame of the single-ion Kondo effect. The first drop in resistivity for $100 \leq T \leq 300$ K is followed by a steady increase, resulting from additional scattering processes of the conduction electrons with the local 4f moments. At $T_N$ this evolution is interrupted by the onset of the AFM order before coherent scattering can develop, in contrast to what has been observed in YbRh$_2$Si$_2$ [7]. This behaviour is a key indicator, which shows that the dominating exchange interaction in YbCo$_2$Si$_2$ is the RKKY interaction and not the Kondo one.

3.5. Specific heat

Specific heat measurements of the YbCo$_2$Si$_2$ single crystal down to 0.35 K are presented in figure 6(a). The black circles show the 4f electronic contribution $C^{4f}/T$ to the specific heat with $B = 0$ T, whereas the blue rectangles show a measurement made at 2 T with $B \perp c$. The temperature is plotted in the logarithmic scale. The phononic specific heat has been subtracted.
using the non-magnetic reference compound LuRh$_2$Si$_2$ instead of LuCo$_2$Si$_2$, which has not been synthesized yet. Since Co is lighter than Rh, which is expected to lead to harder phonons, this procedure should provide a slight overestimation of the phonon contribution and thus an underestimation of the 4f contribution. This shall affect the results only above 4 K (see [27, 28]). The steep increase of $C^{4f}/T$ at $T_N = 1.61 \pm 0.03$ K reflects the onset of the AFM order in zero field. The second anomaly at $T_L = 0.91 \pm 0.03$ K appears in the form of a sharp peak. A detailed analysis around the temperature region of the peak indicates that the magnetic phase transition is of first order. Consequently, the latent heat contribution leads to inaccurate values of $C^{4f}/T$ if the standard PPMS relaxation technique is used. Nevertheless, on the basis of the work of Lashley et al (see [20]), it is possible to calculate specific heat values from the time derivative of the relaxation curve presented in figure 6(b). The non-exponential temperature evolution in time during the heating and the cooling procedure is direct evidence of a first-order phase transition (see arrows in figure 6(b)). The obtained $C(T)$ curve across $T_L$ is plotted as a red line in figure 6(a). From the area under the peak in $C(T)$, one can estimate the latent heat associated with the transition, which amounts to about 0.5 J mol$^{-1}$. Below $T_L$, $C^{4f}/T$ follows a $T^2$ dependence, as expected for AFM spin-waves contributions with dispersion $\epsilon(q) \sim q$. Assuming this behaviour for $T \to 0$ (see the inset of figure 6(a)), a Sommerfeld coefficient $\gamma_0 = 0.13 J K^{-2} mol^{-1}$ for YbCo$_2$Si$_2$ can be extracted. Although YbCo$_2$Si$_2$ is dominated by the RKKY exchange interaction and no strong enhancement of the density of states at the Fermi energy is expected, the relatively large $\gamma_0$ indicates that the local 4f moments and the conduction electrons are partially hybridized.

Evidence of the strong local character of the Yb atoms is given by the entropy $S(T)$ shown in figure 6(c). More than 75% of the ground state entropy $S = R \ln 2$ is found below $T_N$. Since the measurements of the specific heat have been carried out just down to 0.35 K, the extrapolation to $T = 0$ and the integral of the area below the fit around $T_L$ were taken into account. However, this additional part leads only to a small contribution to the whole $S = R \ln 2$, which is reached at about 9 K. On the basis of the entropy, the Kondo temperature $T_K$ can be estimated to be below 1 K. Since the AFM ordering sets in at a higher temperature $T_N$, the compound YbCo$_2$Si$_2$ is clearly dominated by the RKKY interaction, in good agreement with the resistivity measurements, which do not show any sign of Kondo-lattice coherence effects.

In an external magnetic field of 2 T, both magnetic phase transitions are suppressed and $C^{4f}/T$ displays a broad maximum: it is due to the quasi-Schottky contribution that originated from the Zeeman splitting of the doublet ground state.

4. Conclusion

A comprehensive analysis of the magnetic properties of a single crystal of the intermetallic compound YbCo$_2$Si$_2$ has been presented. An investigation of the crystallographic parameters, magnetic anisotropy and low temperature properties studied using the magnetic susceptibility, resistivity and specific heat is included. The CEF level scheme has been determined and a Kramers doublet $\Gamma_7$ proposed as the ground state, similar to what is currently considered for the compound YbRh$_2$Si$_2$.

The results indicate that in YbCo$_2$Si$_2$ AFM order sets in at a temperature $T_N \approx 1.65$ K, followed by another AFM, but first-order, phase transition at $T_L \approx 0.9$ K. The two magnetic phase transitions can be suppressed by an external magnetic field $B = 2 T$ applied in both the easy magnetic direction ($B \perp c$) and the hard one ($B \parallel c$). Below $T_L$, $C^{4f}$ follows the $T^3$
behaviour expected for AFM magnons, but in $\rho(T)$ we did observe a striking $T^3$ power law rather than the expected $T^5$ one. This might be related to more complex magnetic interactions, as is also suggested by the multiple phase transitions. The fact that about 75% of the ground state entropy has been found below $T_N$ indicates that this system is dominated by the RKKY exchange interaction. Nevertheless, a Kondo-like increase of the resistivity below 100 K down to $T_N$ and an enhanced Sommerfeld coefficient $\gamma_0 = 0.13 J K^{-2} mol^{-1}$ indicate a partial hybridization of the local 4f moments with the conduction electrons, but with $T_K$ well below $T_N$.

Owing to our analysis and the fact that Yb seems to have a similar $\Gamma_7$ ground state in both YbCo$_2$Si$_2$ and YbRh$_2$Si$_2$, we highlight the unique opportunity for a direct comparison between these two compounds. In particular, neutron diffraction measurements on our YbCo$_2$Si$_2$ single crystals might be the key experiments to clarify the magnetic structure and magnetic excitations of YbRh$_2$Si$_2$.

Acknowledgment

We acknowledge the collaboration with K Kaneko, N Mufti, L Pedrero and O Stockert.

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