Synthesis, Structure, and Physical Properties of

YbNi₃Al_{9.23}

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Abstract

The physical properties of YbNi₃Al_{9.23}, including the crystal structure, magnetization, specific heat, valence, and electrical resistivity, are reported. Single crystal X-ray diffraction reveals that the compound crystallizes with the rhombohedral space group *R*32 and has unit cell parameters $a = 7.2443(3)$ Å and $c = 27.251(3)$ Å with some crystallographic disorder on an Al site. The compound orders antiferromagnetically at $T_N = 3$ K despite the presence of strong ferromagnetic correlations, accompanied by a spin flop-like transition to a moment-aligned state above 0.1 T. X-ray absorption spectroscopy and magnetic susceptibility measurements indicate a localized Yb³⁺ electronic configuration, while the Sommerfeld coefficient in the magnetically ordered state was determined to be approximately 135 mJ/mol-K², suggesting moderately heavy fermion behavior. Therefore, these data indicate a balance between competing Ruderman-Kittel-Kasuya-Yosida (RKKY) and Kondo interactions in YbNi₃Al_{9.23} with a somewhat dominant RKKY interaction that leads to a relatively high ordering temperature.

1. Introduction

A variety of interesting correlated electron phenomena can be found in the literature, such as unconventional superconductivity (β -YbAlB₄) [1], heavy fermion behavior (e.g., YbCo₂Zn₂₀) [2], mixed valent behavior (e.g., YbAl₃) [3], and quantum criticality (e.g., YbRh₂Si₂) [4]. This results from strong hybridization between the 4f electrons of the Yb local moments and ligand electrons much of which has attracted interest in recent years. The compounds β -YbAlB₄ and α -YbAlB₄ are the newest examples of correlated electron Yb compounds and, in particular, β -YbAlB₄ [1,5] is the first Yb-based heavy fermion superconductor with $T_c = 80$ mK and a Sommerfeld coefficient of 150 mJ/mol-K^2 [1]. β -YbAlB₄ appears to be in close proximity to a field-tuned quantum critical point, in which the physical properties show non-Fermi liquid behavior, such as a logarithmic divergence of specific heat and unusual powerlaw behavior of the electrical resistivity and magnetic susceptibility [6].

In our search for new strongly correlated Yb compounds, we studied YbNi₃Al_{9,23}, which crystallizes in the rhombohedral space group $R\bar{3}2$ (No. 155). It should be noted that Ohara and co-workers are working on similar compounds and have recently shown some comparable physical properties [7]. Single crystals of the compound were isolated through metal flux growth reactions in an excess of Al metal and single-crystal X-ray diffraction determined that the structure crystallized in a new structure type that is closely related to ErNi₃Al₉ and YNi₃Al₉ [8]. The physical properties, including magnetization, specific heat, and electrical resistivity, reveal YbNi₃Al_{9,23} is an antiferromagnet at $T_N = 3$ K in zero magnetic field and undergoes a spin-flop transition to a moment-aligned state at fields above 0.1 T at temperatures below T_N .

2. Experimental details

Single crystals of $\text{YbNi}_3\text{Al}_{9.23}$ were prepared from the respective elements using an excess of Al. The elements were loaded in a 2 cm^3 alumina crucible in the ratio $\text{Yb:Ni:Al}=1:2:20$ and sealed in an evacuated quartz tube. This vessel was placed in a high temperature furnace that was allowed to heat according to the following sequence: ramp to 1000°C (200°C/h) where it dwelled for 12 h, and finally cooled to 700°C (8°C/h). At this temperature, the reaction vessel was quickly removed and the molten Al flux was isolated through centrifugation resulting in small irregular-shaped crystals with shiny facets. The majority of the crystals measured 2 mm in length.

Single crystal and powder X-ray diffraction on the $\text{YbNi}_3\text{Al}_{9.23}$ crystals revealed that the crystal structure of the compound is rhombohedral with unit cell parameters $a = 7.2443(3)\text{ \AA}$, $c = 27.251(3)\text{ \AA}$. The details of the refinement are compiled in Table 1. After the initial structure solution it became apparent that the structure of the compound is closely related to that corresponding to both the ErNi_3Al_9 and YNi_3Al_9 types [8] with the space group $R\bar{3}2$ (Pearson symbol $hR78$). It should be noted that both structures are similar to one another, one being an ordered array of Al triangles (ErNi_3Al_9) and rare-earth atoms while the other is a disordered arrangement of the same units (YNi_3Al_9).

It is important to differentiate the two models in order to validate our refinement of $\text{YbNi}_3\text{Al}_{9.23}$. We therefore used both structure types for our initial refinements. The ErNi_3Al_9 structure contains one rare-earth site located at Wyckoff position $6c$, one Ni site, and six Al sites. The solution using the atomic coordinates from ErNi_3Al_9 resulted in high R values and negative thermal parameters on many of the sites. The YNi_3Al_9 structure on the other hand contains two Y sites located at Wyckoff sites $3b$ and $6c$, one Ni site, and seven Al sites. When the refinement was tried using this model a lot of crystallographic disorder was apparent which included the two Y sites that were partially occupied along with the Al5 and Al7 atoms. Fitting this model to the data results in a reasonable structural solution with R values less than 10% but the Al7 site has a very elongated thermal parameter. A better model for representing the

structure contains one Yb site, one Ni site, and seven Al positions of which only one site (Al5) is refined with a partial occupancy. This structure refinement has well behaved displacement parameters and R values of 2.25% and 6.09% for R_1 and wR_2 , respectively. The Al5 and Al7 atoms are close together in the structure with the arrangement of Al5 triangles with the Al7 at the center of this triangle and therefore results in short atomic distances. The reason for the slight excess of Al in the formula comes from the Al on the Al7 site. The ideal formula, therefore for the perfectly ordered structure would be YbNi_3Al_9 . Further crystallographic information including refinement parameters and atomic coordinates are tabulated in Tables 1 and 2.

The magnetization studies were made with a Quantum Design MPMS in the temperature range 350 K down to 2 K. Since the crystals were small, magnetization measurements were performed on a collection of randomly oriented single crystals. Specific heat measurements were performed on a Quantum Design PPMS from 0.4 K to 10 K in magnetic fields up to 9 T. For electrical resistivity measurements, contacts were made by spot welding 25- μm diameter Pt wires on a polished face of the crystals, and the resistivity was measured with an excitation current of 0.3 mA. In some cases, a small applied field (0.1 T) was used to suppress the superconducting transition of a small amount of Al flux. X-ray Absorption Near Edge Structure (XANES) data were collected for Yb L_{III} edge (8944 eV) at the Stanford Synchrotron Radiation Lightsource, beamline 4-1, using a half-tuned double crystal Si(220) monochromator with a slit height of 0.7 mm. The samples were ground to a fine powder in acetone, passed through a 30-micron sieve, and brushed onto adhesive tape. Two double layers were stacked so that the change in absorption at the Yb L_{III} edge, $\Delta\mu t$, was approximately unity, and the sample was loaded into a LHe flow cryostat. Three scans were collected for $T = 20$ and 300 K, respectively. An Yb_2O_3 reference sample was also measured simultaneously with the $\text{YbNi}_3\text{Al}_{9.23}$ sample to calibrate the edge energy (first inflection point set to 8943.0 eV) and to correct for the beam energy drift. The background from other absorption processes was extrapolated from the pre-edge region and subtracted from the data.

3. Results and Discussion

3.1 Crystal Structure

The crystal structure of $\text{YbNi}_3\text{Al}_{9.23}$ is shown in Figure 1 and is constructed from the layered arrangement of Ni and Al atoms with intercalated layers of Yb atoms stacked along the crystallographic c -axis. The Yb sublattice of the compound forms planar honeycomb-like nets similar to those found in graphite or in binary intermetallics crystallizing with the AlB_2 -type structure [8]. Centering each of these six-membered rings in the ab crystallographic plane are Al-atom triangles. The coordination of each Yb site in the crystal structure is a distorted 17-membered polyhedron composed of Al triangles and Ni triangles which surround the Yb cation. The Yb-Yb distance within the hexagonal sheets measured 4.183 Å, while the closest Yb-Yb distance between the layers corresponded to 9.093 Å.

3.2 Magnetization

The magnetic susceptibility data, represented as $\chi(T)$, are shown in Figure 2a. As shown in the top right inset of Figure 2a, the inverse susceptibility, $\chi^{-1}(T)$, exhibits Curie-Weiss behavior down to approximately to 10 K. A linear fit to these data above 150 K gives an effective moment $\mu_{\text{eff}} = 4.26 \mu_{\text{B}}$ (close to that expected for Yb^{3+} ; $\mu_{\text{eff}} = 4.54 \mu_{\text{B}}$) and a negative $\theta_{\text{CW}} = -9$ K, indicating a weak net antiferromagnetic interaction of the Yb moments. Trivalent Yb is also indicated from the XANES measurements (see below). At $T < 5$ K, the $\chi(T)$ data increase sharply, reaching a value nearly 2.5 emu/mol-Yb indicating a tendency towards ferromagnetism, and display a peak at 3 K (Figure 2), consistent with the onset of long range antiferromagnetic magnetic order. A plot of χT vs T (Figure 2b) shows an increase in this product below 7 K, which provides evidence for the presence of ferromagnetic correlations. The χT data also show a peak which reflects the point which the antiferromagnetic order sets in. We note that the $\chi(T)$ data were collected in a magnetic field of 0.1 T, very close to the spin-flop transition. Isothermal magnetization is plotted versus field in Figure 3a for temperatures above and below $T_{\text{N}} = 3$ K. The inset of Figure 3a is an expanded view of the low-field region that shows a step-like feature, indicative of a spin-flop transition, occurring at fields between 0.1 T - 0.2 T for temperatures of 2

K, 2.5 K, and 3 K in the Néel state. The M/H curves are similar to those of Yb_3Pt_4 , where strong antiferromagnetic interactions are observed but a small ferromagnetic component is seen at lower temperatures [9]. Arrott plots (M^2 vs H/M) constructed from the $M(H)$ data are shown in Figure 3b. An extrapolation of the linear region above 3 T suggests that the field-induced state is ferromagnetic-like with a Curie temperature $T_c = 3.5(5)$ K, determined from a fit of the $M_{\text{sat}}^2(T)$ data to obtain T_c where $M_{\text{sat}}^2 = 0$. Lastly, neutron diffraction measurements would be useful to determine the magnetic structure of $\text{YbNi}_3\text{Al}_{9.23}$.

3.3 XANES

To determine the valence of Yb in $\text{YbNi}_3\text{Al}_{9.23}$, X-ray absorption spectroscopy measurements were performed at the Yb L_{III} edge (8943 eV) as shown in Fig. 4. The variation between the Yb L_{III} -edge XANES data measured at $T = 20$ K and 300 K is very small, even in the extended x-ray absorption fine structure (EXAFS) region ($E \geq 50$ eV). The XANES data were fit between 8900 and 9040 eV to a sum of numerical functions, as no lutetium material was available to provide an analog to the Yb absorption lineshapes for the potential Yb(II) and Yb(III) contributions. Instead, as shown in Figure 4, two pseudo-Voigt functions are used to fit the white-line region, which is typically composed of a divalent (~ 8940 eV) and trivalent peak (~ 8946 eV), an integrated pseudo-Voigt (essentially an arctangent function, used to simulate the post-edge background absorption) and, in this case, a negative Gaussian peak (used to fit the dip caused by the EXAFS oscillation at ~ 8956 eV). Here we only show the data at $T = 20$ K with the fit, since they are almost identical for $T = 300$ K. These data indicate no visible divalent contribution. To obtain a quantitative estimate of the f -hole occupancy n_f , the position of the divalent peak in the fit function was held fixed, yielding $n_f = 0.96 \pm 0.04$, consistent with a (nearly) trivalent, localized Yb ground state in $\text{YbNi}_3\text{Al}_{9.23}$.

3.4 Specific Heat

As shown in Figure 5a, the specific heat plotted as C/T as a function of T , shows a sharp anomaly near 3 K in zero field and is consistent with the peak observed in $\chi(T)$. The specific heat at T_N is large,

reaching nearly 2.70 J/mol-K^2 . In the paramagnetic state, the Sommerfeld coefficient (γ) obtained from a fit to the data in the form $C/T = \gamma + \beta T^2$ between $6 \text{ K} < T < 15 \text{ K}$ yields $\gamma = 180 \text{ mJ/mol-K}^2$ (Figure 5b) and $\beta = 1.407 \text{ mJ/mol-K}^4$, corresponding to a Debye temperature $\theta_D = (12\pi^4 R n / 5\beta)^{1/3} = 262 \text{ K}$, where R is the Gas constant and n is the number of atoms in the formula unit. Below T_N , the Sommerfeld coefficient $\gamma = 132 \text{ mJ/mol-K}^2$ is obtained from the zero-temperature limit of a linear fit of the C/T vs T^2 data below 0.7 K , where the leading order T^2 term is appropriate for antiferromagnetic magnons (Fig. 5b). The application of magnetic fields greater than the spin-flop field as plotted in Figure 5a shows that the specific heat anomaly is pushed to higher temperatures and somewhat broadened, consistent with a field-induced ferromagnetic-like state. To obtain the magnetic entropy in zero field, the lattice contribution (the high temperature βT^2 term) was subtracted from the measured C/T data and the result integrated from $T = 0$ to T , since a LuNi_3Al_9 analogue could not be synthesized. As shown in the inset of Figure 5a, the magnetic entropy (S_{mag}) associated with the zero-field magnetic transition amounts to about $80\% R \ln 2$ consistent with a doublet ground state. Such a doublet ground state is expected since the crystalline electric field will split the $J = 7/2 \text{ Yb}^{3+}$ multiplet into four doublets [10].

3.5 Electrical Resistivity

The electrical resistivity in zero field between 2 and 300 K of $\text{YbNi}_3\text{Al}_{9.23}$ is shown in Figure 6. The $\rho(T)$ data decrease approximately linearly from room temperature down to 75 K . A change in the slope can be observed near 100 and 25 K , possibly reflecting a change in scattering due to depopulation of excited crystal field levels. A sharp decrease in $\rho(T)$ at 3 K corresponds to the antiferromagnetic transition, observed in both the magnetic susceptibility and specific heat. The room temperature resistivity is $40 \mu\Omega\text{-cm}$, while the resistivity at 2 K is $\rho = 3 \mu\Omega\text{-cm}$, giving a residual resistivity ratio (RRR) value of 13 . Note that at $T = 0$, the extrapolated value would be even greater.

4. Discussion

There are many examples of ternary rare-earth transition metal aluminides in the literature [8]. Quite a number of these are composed of Al-atom triangles and are found in nine structure types including $\text{Gd}_{1.33}\text{Pt}_3\text{Al}_8$ [11], ErNi_3Al_9 [12], $\text{Er}_{1.33}\text{Pt}_3\text{Al}_8$ [13], $\text{Er}_4\text{Pt}_9\text{Al}_{24}$ [13], $\text{Ho}_2\text{Rh}_3\text{Al}_9$ [14], $\text{Sc}_{0.67}\text{Fe}_2\text{Si}_5$ [15], YNi_3Al_9 [12], $\text{Y}_4\text{Pt}_9\text{Al}_{24}$ [13], and $\text{Y}_2\text{Co}_3\text{Ga}_9$ [16]. It is also well documented that these compounds can contain significant disorder arising from stacking faults of the atomic layers. The “1-3-9” compounds with the ErNi_3Al_9 and YNi_3Al_9 types only have a few isostructural analogues formed with the mid and late rare-earth elements Gd and Er for ErNi_3Al_9 and Y and Dy for YNi_3Al_9 . It has been suggested by others that the reaction conditions play a significant role on minimizing the disorder in some of these compounds, e.g., longer cooling rates or annealing times for flux-grown crystals [13]. From our own efforts, we find no apparent difference in the crystallographic disorder in $\text{YbNi}_3\text{Al}_{9.23}$ for reactions which were allowed to dwell for an extended amount of time before cooling.

The antiferromagnetic ordering temperature of 3 K for $\text{YbNi}_3\text{Al}_{9.23}$ is relatively high for Yb-based intermetallics; high Néel temperatures have only been observed in a few compounds such as YbPtAl ($T_N = 5.8$ K) [17], YbNiAl ($T_N = 3$ K) [17,18], YbPtIn ($T_N = 5.8$ K) [19], and YbPtSn ($T_N = 3.5$ K) [20]. For a given crystal-field splitting, the ground state for Yb compounds is determined by a balance between the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, which promotes magnetic order, and the Kondo effect, which favors a heavy, Fermi-liquid paramagnetic ground state, as pointed out by Doniach some years ago [21]. Some Yb-based materials (e.g., YbRh_2Si_2 [4], YbAgGe [22]) appear to be at a boundary where the RKKY and Kondo interactions are comparable, which leads to the suppression of long-range magnetic order and the formation of a quantum critical point. Other Yb-based compounds with high ordering temperatures exhibit localized Yb^{3+} behavior [23] with little or moderate effective mass enhancement, suggesting they reside in the RKKY-dominated side of the Doniach diagram, or at most in the middle where the ordering temperature is maximal. Without a series of isostructural YbM_3Al_9 compounds, a quantitative analysis of the hybridization strength between local Yb moments and

conduction electrons that would place $\text{YbNi}_3\text{Al}_{9.23}$ precisely on the Doniach diagram is not possible; however, the physical properties of $\text{YbNi}_3\text{Al}_{9.23}$, including the XANES and susceptibility measurements, indicate (nearly) trivalent Yb, while the enhanced Sommerfeld coefficient and slightly reduced entropy for a doublet ground state suggest that electronic mass renormalization by the Kondo effect is important in $\text{YbNi}_3\text{Al}_{9.23}$. If this suggestion is correct, the application of pressure to $\text{YbNi}_3\text{Al}_{9.23}$ would be expected to move the system further to the left side of the Doniach diagram and suppress the Néel temperature.

5. Summary

We report the physical properties of $\text{YbNi}_3\text{Al}_{9.23}$ including the structure, magnetization, x-ray absorption near-edge structure, specific heat, and electrical resistivity. In zero field, the material exhibits antiferromagnetic order at $T_N = 3$ K and undergoes a spin-flop transition at fields greater than 0.1 T into a ferromagnetic-like ground state. Both the XANES and magnetic susceptibility data indicate a well localized Yb^{3+} magnetic moment, while the extrapolated Sommerfeld coefficient in the magnetically ordered state is ~ 135 mJ/mol-K², suggesting a dominant RKKY interaction with a significant Kondo interaction in this material.

6. References

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Table 1. Selected Single-Crystal Data and Structure Refinement Parameters for YbNi₃Al_{9.23}

Empirical formula	YbNi ₃ Al _{9.23(1)}
Formula weight	598.11 g/mol
Collection temperature	140 K
Radiation, wavelength (λ)	0.71073
Crystal system	Trigonal
Space group	<i>R</i> 32
Unit cell dimensions	$a = 7.2443(3) \text{ \AA}$ $c = 27.251(3) \text{ \AA}$
Unit cell volume, <i>Z</i>	1238.53(14) $\text{\AA}^3$, 6
Density (ρ_{calc})	4.811 g cm ⁻³
Absorption coefficient (μ)	18.856 mm ⁻¹
Crystal size (mm)	0.060 $\times$ 0.045 $\times$ 0.039
Reflections collected	4688
Unique reflections	676
GOF on F^2	1.106
Final R indices ^a ($I > 2\sigma(I)$) ^a	$R_1 = 0.0225$ $wR_2 = 0.0609$

^a $R_1 = \Sigma ||F_o| - |F_c|| / \Sigma |F_o|$; $wR_2 = [\Sigma [w(F_o^2 - F_c^2)^2] / \Sigma [w(F_o^2)^2]]^{1/2}$, and $w = 1/[\sigma^2 F_o^2 + (AP)^2 + BP]$, $P = (F_o^2 + 2 F_c^2)/3$.

Table 2. Atomic Coordinates and Equivalent Isotropic Displacement Parameters (U_{eq}^a) for $YbNi_3Al_{9.23}$ (140 K)

Atom	Wyckoff site	x	y	z	U_{eq}
Yb	$6c$	0	0	0.166837(10)	0.0068(2)
Ni	$18f$	0.3332(2)	0.0056(2)	0.08517(5)	0.0034(2)
Al1	$6c$	0	0	0.38720(10)	0.0045(5)
Al2	$6c$	0	0	0.28137(10)	0.0036(5)
Al3	$6c$	0	0	0.05235(10)	0.0040(5)
Al4	$9d$	0.3339(3)	0	0	0.0063(7)
Al5 ^b	$9e$	0.2103(4)	0	$\frac{1}{2}$	0.0027(10)
Al6	$18f$	0.3337(4)	0.3358(4)	0.09950(10)	0.0021(5)
Al7 ^c	$3b$	0	0	$\frac{1}{2}$	0.0120(10)

^a U_{eq} is defined as one-third of the trace of the orthogonalized U_{ij} tensor. ^b Refined as a partial occupancy of 82%. ^c Al7 is refined with isotropic thermal parameters.

Figure 1. The crystal structure of $\text{YbNi}_3\text{Al}_{9.23}$ with the unit cell outlined. The Yb, Ni, and Al atoms are depicted as green, red, and blue spheres, respectively.

Figure 2. The magnetic susceptibility data measured on a collection of single crystals of $\text{YbNi}_3\text{Al}_{9.23}$ in a field of $H=0.1$ T. (a) The $\chi(T)$ data measured from 2 K to 350 K. Left inset: $\chi(T)$ below 10 K. Right inset: inverse susceptibility $\chi^{-1}(T)$. (b) Magnetic susceptibility, plotted as χT vs T .

Figure 3. (a) Magnetization M vs magnetic field H of $\text{YbNi}_3\text{Al}_{9.23}$. The inset shows the low field isotherms up to 0.5 T. (b) An Arrott plot, M^2 vs H/M , of the magnetic isotherms from 2 K to 4 K with the solid lines showing linear fits to the data.

Figure 4. Yb L_{III} -edge XANES data of $\text{YbNi}_3\text{Al}_{9.23}$ at 20 K (open circles) and fit. The various absorption features are modeled with pseudo-Voigt functions as described in the text. These data indicate a nearly trivalent state for Yb in this compound ($n_f=0.96\pm0.04$). The trivalent Yb contribution is the dominant peak at 8946 eV (blue line), the possible divalent contribution is centered at 8940 eV (green line), and the first EXAFS peak is near 8956 eV (purple line).

Figure 5. (a) The specific heat divided by temperature, C/T vs T , of $\text{YbNi}_3\text{Al}_{9.23}$ in various magnetic fields up to 9 T. Inset: The magnetic contribution to the entropy, S_{mag} , determined by subtracting the lattice contribution (βT^2) from the measured C/T . (b) C/T vs T^2 . The red line is a linear fit to the data yielding the Sommerfeld coefficient in the paramagnetic state. Inset: A linear fit to the data well below the ordering temperature yielding γ in the ordered state.

Figure 6. The electrical resistivity $\rho(T)$ measured within the temperature range between 2 K and 300 K. Inset: $\rho(T)$ below 20 K.

Figure 1 (Fig1_YbNi3Al9.tif)

Figure 2 (Figure 2_YbNi3Al9.tif)

Figure 3 (Figure 3_YbNi3Al9.tif)

Figure 4 (Figure4_YbN3A19.tif)

Figure 5 (Figure 5_YbNi3Al9.tif)

Figure 6 (Figure 6_YbNi3Al9.tif)

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