Magnetic Structure of $TbRu_2Al_{10}$

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Specific heat and magnetisation measurements show that $TbRu_2Al_{10}$ orders antiferromagnetically at 15 K. The magnetic structure has been determined from powder neutron diffraction to be sinusoidal along the *b*-axis with the moments parallel to the *c*-axis and a peak moment of 7.4(1) μ_B. The overall structure can be described as a 'zigzag' antiferromagnet.

1. Introduction

$TbRu_2Al_{10}$ belongs to a series of rare earth intermetallic compounds that crystallise in the $YbFe_2Al_{10}$ orthorhombic crystal structure [1]. There has been recent interest in members of this family due to the range of interesting magnetic properties they possess: there are multiple magnetic phase transitions and metamagnetism, e.g. in $TbFe_2Al_{10}$ [2-4] and anomalously high magnetic ordering temperatures in $CeRu_2Al_{10}$ [5,6], as well as the marked effects that variation of the lanthanide and transition metal used have on overall magnetic properties [7,8]. $TbRu_2Al_{10}$ is one of many compounds of this family that have yet to be fully studied and thus an understanding of its structure and properties will provide insight into trends in the behaviour of the wider series.

2. Experimental

Powdered samples of $TbRu_2Al_{10}$ were obtained from offcuts of single crystals which had been grown via an Al flux. Phase purity of the samples was tested using X-ray diffraction on a Panalytical Empyrean X-ray diffractometer. Neutron diffraction was carried out on both the *Wombat* and *Echidna* instruments at the OPAL reactor, *Lucas Heights* (neutron wavelengths of 2.421 Å and 2.439 Å, respectively were used). Bulk property measurements were carried out using a Quantum Design PPMS.

3. Results

3.1 Heat Capacity Measurements

The heat capacity measurements carried out between 2 K and 300 K on a polycrystalline sample of $TbRu_2Al_{10}$ indicate that there is only a single magnetic phase transition in this range. A subset of this data (2-60 K) is seen in Fig. 1 where the $TbRu_2Al_{10}$ result is compared with the isomorphous non-magnetic compound $LaRu_2Al_{10}$. Comparison with a non-magnetic iso-structural compound allows a determination of the magnetic contribution to both the heat capacity and total entropy, assuming that the temperature values are appropriately scaled according to the atomic weight of the differing rare earth atoms. This is done here using the many-Debye method [9], showing a significant contribution to the heat capacity from the magnetic phase transition. Integrating over this difference in the complementary Heat Capacity/Temperature vs Temperature data gives a value of 17.5(1) J $mol^{-1}K^{-1}$ for the magnetic entropy of $TbRu_2Al_{10}$, lower than the expected $R\ln(2J+1)$ value of 21.32 J $mol^{-1}K^{-1}$. This indicates that there may be crystal field splitting of the $2J + 1$ total angular momentum sublevels occurring.

3.2 Magnetisation and Susceptibility Measurements

Magnetisation and susceptibility measurements between 2 K and 300 K indicate a transition to an antiferromagnetic structure at 15 K (2 K to 200 K is shown in Fig. 2). Using the Curie-Weiss law, the curve of inverse susceptibility above the transition temperature was used to determine a Curie constant, calculated as 2.3 x 10^{-4} m^3 K kg^{-1}. This corresponds to a moment of 9.56 μ_B, close to the free ion value of 9.73 μ_B for Tb^{3+}.

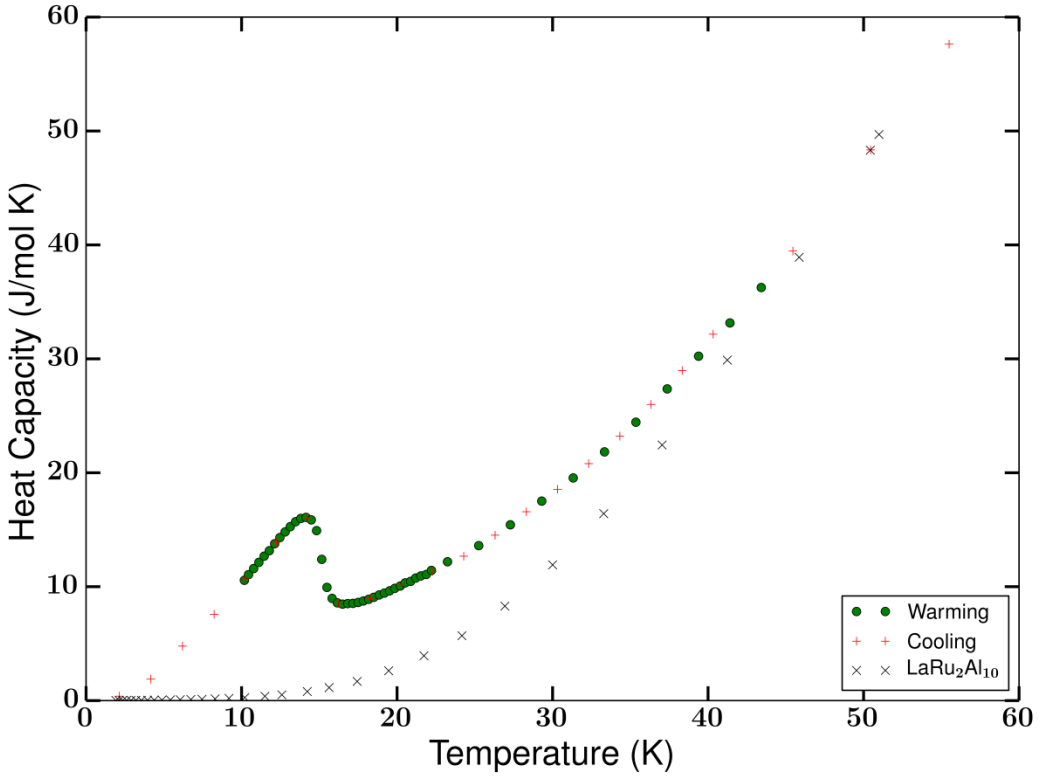

Fig. 1: Heat Capacity vs Temperature curves for $TbRu_2Al_{10}$ and $LaRu_2Al_{10}$ between 2 K and 60 K. Only a single magnetic transition is present at 15 K for the Tb compound with no further transitions seen in this temperature range. Both warming and cooling curves are indicated and show a significant contribution to the heat capacity attributable to the ordering of the magnetic moments within the material.

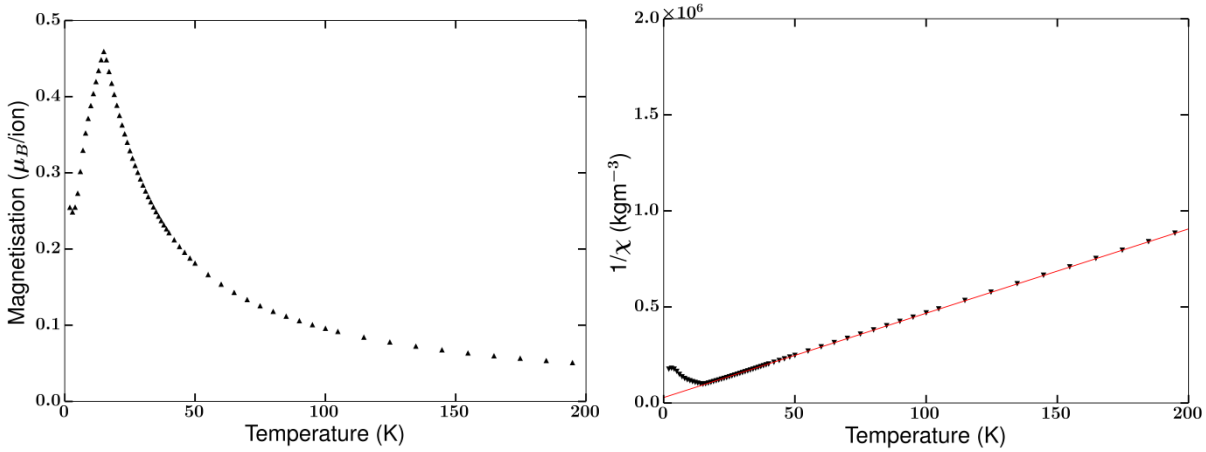

Fig. 2: Magnetisation vs Temperature (left) and Inverse Susceptibility vs Temperature (right). Both indicate antiferromagnetic ordering at T_N = 15 K. A fit using the Curie-Weiss law has been included on the inverse susceptibility graph, giving an effective magnetic moment of 9.56 μ_B, close to the free ion value of 9.73 μ_B for Tb^{3+}

3.3 Neutron Powder Diffraction

Neutron diffraction was carried out on polycrystalline $TbRu_2Al_{10}$ over a range of temperatures, both above and below the ordering temperature of 15 K. The nuclear reflections are found to be well described using the *$YbFe_2Al_{10}$* structure type (space group: Cmcm, #63) with lattice parameters as they appear in Table 1. The magnetic reflections appear to be incommensurate with the nuclear peaks in this compound, with a propagation vector of (0, 0.760(1), 0). Such incommensurate structures are often described using irreducible representations of the parent space group [10,11]. Using the program *BasIreps* in combination with *FullProf* [12] it is possible to refine a magnetic structure that is based off the irreducible representations obtained. For this material, the position of the magnetic moment bearing atoms are not fully described by the symmetry elements of the little group (symmetry elements that leave the propagation vector invariant) and as such the two atom positions at (x,y,z) and (-x,-y,-z) are separated into two magnetic sub lattices, each described by a separate set of basis vectors. From the magnetisation measurements, it is known that this material orders antiferromagnetically, hence the basis vectors of these two sub lattices are equal in magnitude but opposite in sign. Three different configurations were obtained as possible solutions, with the magnetic moments in each pointing along different crystallographic axes. However, the representation corresponding to moments parallel to the *c*-axis was the only satisfactory configuration, as refinements with the other two representations left several magnetic peaks unaccounted for. The refinement statistics and atomic parameters for the Rietveld refinement are shown in Table 2. Schematics of the magnetic structure are shown in Fig. 3.

Table 1: Lattice parameters for $TbRu_2Al_{10}$ at a range of temperatures.

Temperature (K)	***a***	***b***	***c***
6	9.054(1)	10.175(1)	9.094(1)
30	9.058(1)	10.185(1)	9.100(1)
300	9.066(1)	10.190(1)	9.108(1)

Based on these refinements, the magnetic structure appears to be sinusoidal along the *b*-axis with the moments parallel to the *c*-axis and this structure is maintained from the ordering temperature down to 6 K, with a peak moment value equal to 7.4(1) μ_B. This is in contrast to $TbFe_2Al_{10}$, in which the ordering is parallel to the *a*-axis and the structure shifts from sinusoidal to square wave modulated as the temperature is decreased, with a maximum moment of 8.33(7) μ_B[3,4]. However the overall magnetic structure of $TbRu_2Al_{10}$ does have similarities with that of the iron analogue in that there are zigzag double layers of ferromagnetically aligned moments that are themselves aligned antiferromagnetically with neighbouring double layers along the *b*-axis. The structural and magnetic differences seen between the two compounds may be due to the larger unit cell of $TbRu_2Al_{10}$ that is the result of including a larger ruthenium atom in place of iron. This increases the spacing between the magnetic Tb^{3+} ions and alters the Fermi surface, causing a change in the exchange energy associated with the RKKY interaction and hence an overall change in the structure.

Table 2: Atomic parameters and refinement statistics for $TbRu_2Al_{10}$ at 6 K

Atom	x	y	z
Tb	0	0.127(1)	0.25
Ru	0.25	0.25	0
Al1	0.221(3)	0.361(3)	0.25
Al2	0.356(2)	0.131(3)	0.25
Al3	0	0.159(2)	0.596(2)
Al4	0	0.382(3)	0.053(2)
Al5	0.225(2)	0	0

R_p	R_{wp}	R_{exp}	R_{Bragg}	R_{Mag}	χ^2
7.46	7.94	4.36	2.39	4.48	**3.32**

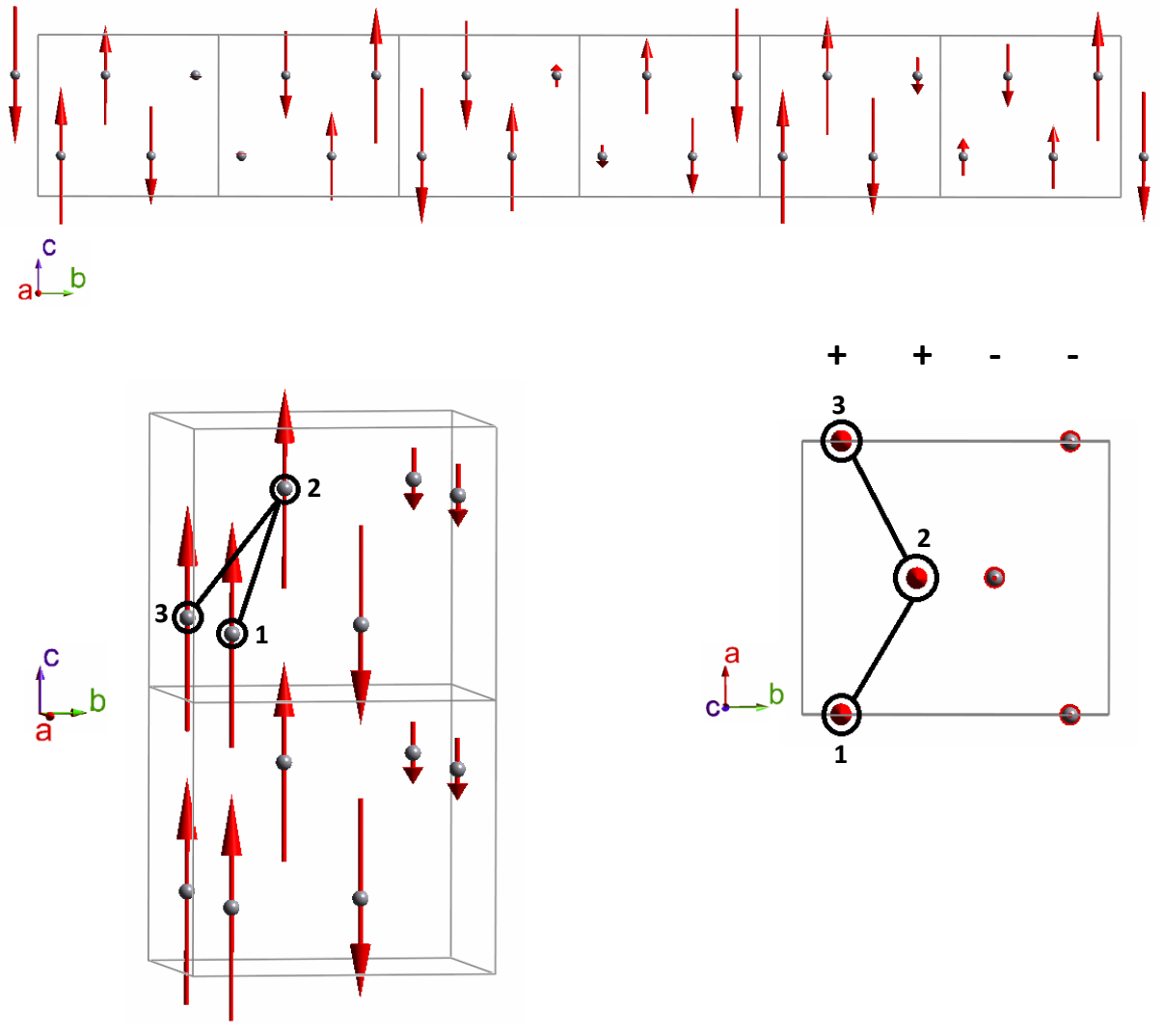

Fig. 3: Schematics of the magnetic structure of $TbRu_2Al_{10}$. The top image shows how the magnetic moments of the Tb^{3+} ions change along the *b*-axis direction. The bottom left image is made up of two arbitrary unit cells showing how the magnetic moments within the material form ferromagnetic double layers. Viewing the moments along the *c*-axis (the bottom right image) shows the zigzag pattern formed by the arrangement of the atoms, with the (+-) motif indicating the direction of the spins. Those atoms circled are the same in both images.

4. Conclusion

$TbRu_2Al_{10}$ has been found to have only a single magnetic transition at 15 K, at which point the material adopts a sine-wave modulated structure along the *b*-axis, with an overall zigzag antiferromagnetic character, similar to that previously observed in $TbFe_2Al_{10}$. Further studies of this material will include magnetisation measurements along each crystallographic axis as well as neutron diffraction in applied fields using a single crystal in order to study the metamagnetic phase transitions present.

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