First-Order Magnetic Phase Transition in TmGa

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Recently [1], we determined the magnetic structure of the intermetallic compound TmGa using neutron diffraction and ^{169}Tm Mössbauer spectroscopy. TmGa shows two magnetic 'events' in its ac-susceptibility, at 15(1) K and 12(1) K. The upper transition is to an incommensurate antiferromagnetic structure that gives way to a dominant ferromagnetic structure at the lower transition. However, the Mössbauer results suggested that the 12 K transition is in fact first-order, with coexisting ferromagnetism and incommensurate antiferromagnetism. Here, we present neutron diffraction data that show a clear thermal hysteresis in the magnetic scattering intensity, confirming our suggestion of first-order magnetic ordering in TmGa.

1. Introduction

The series of intermetallic compounds RGa (R = rare earth) crystallise in the orthorhombic CrB-type *Cmcm* (#63) structure with one R site and one Ga site, both 4c. The RGa compounds were first prepared in the early 1960's and the dominant magnetic order in this series is ferromagnetic. The RGa compounds have recently attracted attention due to their potential for use in magnetocaloric-effect-based low-temperature refrigeration applications [e.g. 2]. We refer the reader to our previous paper [1] for a full description of the work on the RGa series of compounds.

The subject of this short note is TmGa which was reported to be a ferromagnet with a Curie temperature of 15(1) K by Gao et al. [3]. Specific Heat measurements on TmGa showed two magnetic transitions occurring at 15 K and 12.5 K, the upper transition being the Curie temperature and the lower transition ascribed to a spin-reorientation. Single-crystal magnetisation measurements at 4.2 K showed that the magnetic order is along the a-axis [3]. A substantial magnetisation along the a-axis was observed and a Tm magnetic moment at 2 K of 6.1 μ_B in an applied magnetic field of 7 T was reported. This is significantly smaller than the 'free-ion' value of 7 μ_B. It is also clear from the curvature of the magnetisation data presented by Gao et al. [3] that the magnetic structure of TmGa below the 'Curie' temperature is more complex than that of a simple ferromagnet.

Recently [1], we reported our determination of the magnetic structure of TmGa, based on high-resolution neutron powder diffraction and ^{169}Tm Mössbauer spectroscopy. We showed that the magnetism of TmGa is predominantly ferromagnetic but involves a coexistence of commensurate and incommensurate components, suggesting that the transition at 12 K is not a 'spin-reorientation' but rather a first-order transition from incommensurate antiferromagnetism to commensurate ferromagnetism. Here we present new neutron diffraction data that exhibit the clear thermal hysteresis signature of first-order behaviour.

2. Experimental Methods

All details of the sample preparation and characterisation procedures are given in our previous paper [1]. Briefly, the TmGa sample was prepared in an argon-arc furnace and its purity was confirmed by x-ray powder diffraction (Cu-K_α). Neutron diffraction data were obtained on the Echidna high-resolution powder diffractometer at the OPAL reactor in Sydney with a neutron wavelength of 2.4395(5) Å. All diffraction patterns were refined using the Rietveld method as implemented in the FullProf/WinPlotr program. The new diffraction patterns were collected on warming and cooling over the temperature range 7 to 15 K with a 1 K step. Temperature equilibration times of around half an hour were employed.

3. Results

The neutron diffraction pattern of TmGa obtained at 12 K and shown in our recent paper [1] exhibited two clear incommensurate magnetic peaks at scattering angles 2θ of around 43° and 47° spanning the (021) nuclear position. The propagation vector describing this incommensurate (IC) antiferromagnetism (AF) is **k** = [0 0.275(2) 0] with the Tm magnetic moments aligned along the orthorhombic a-axis. Upon cooling below 12 K we observed the growth of a strong ferromagnetic (FM) peak at the (021) position (around 45°), but the IC-AF peaks remained. The combination of the neutron diffraction and ^{169}Tm Mössbauer data implied a first-order magnetic ordering with coexisting IC-AF and FM structures.

In Fig. 1 we show new neutron diffraction data obtained by careful thermal scanning over the temperature range 7 to 15 K. This figure is centred on the (021) position. These data show a clear thermal hysteresis of 0.5 K, indicative of a first-order transition. This is most clearly seen in the behaviour of the strong FM (021) peak around 2θ = 45°.

The tilting of the two IC satellites is due to a weak temperature dependence of the IC propagation vector. Refinement of the 3 K pattern yields **k** = [0 0.271(3) 0].

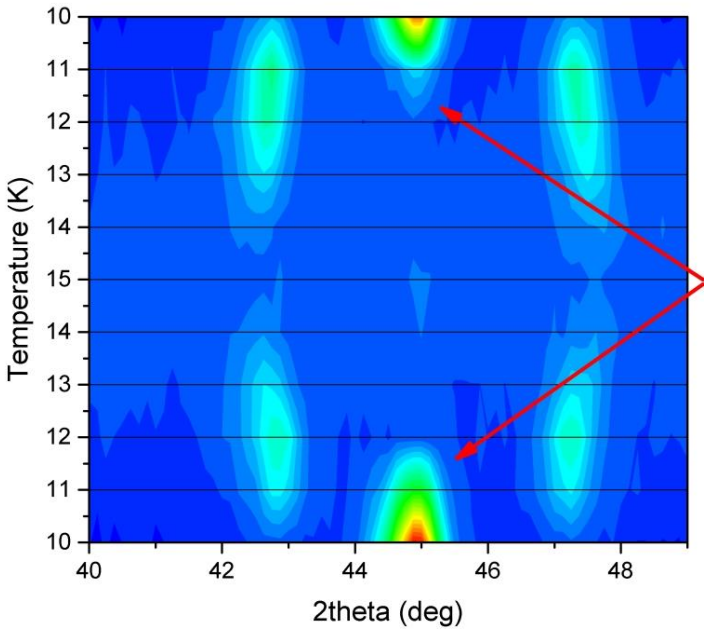

Fig. 1. Neutron powder diffraction thermal scan contours of TmGa, obtained over the temperature range 10 K to 15 K to 10K. The red arrows indicate the ferromagnetic (021) peak.

4. Conclusion

We have used a thermal scanning method to obtain neutron diffraction patterns of the intermetallic compound TmGa. These data confirm our earlier suggestion [1] that the magnetic behaviour of this compound is more complex than previously thought and involves first-order behaviour, rather than the previously suggested spin-reorientation [3].

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References

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