Magnetic and Mössbauer measurements have been made on the intermetallic compound Tm$_2$Al from 1.4 to 300 K. The susceptibility data show Curie--Weiss behaviour with an effective magnetic moment of 7.7(1) μ$_B$ per atom, cf. the free ion value of 7.56 μ$_B$. No evidence of magnetic ordering is found down to 1.4 K. By way of contrast, however, the $^{169}$Tm Mössbauer data at 1.4 K reveal two fully split six-line spectra, with differing magnetic and quadrupole hyperfine parameters. By 4.2 K, the weaker of the two sub-spectrum has collapsed to form a peak in the centre of the Mössbauer spectrum. At higher temperatures the intensity of the split sub-spectrum slowly disappears, while retaining a virtually temperature-independent magnetic splitting. Both the Tm sites in Tm$_2$Al therefore exhibit typical paramagnetic relaxation behaviour, with little or no cross-talk between the two Tm species. As with both TmAl and Tm$_3$Al$_2$, Tm$_2$Al exhibits unusually long relaxation times for a metallic material.

1. Introduction

It is now well established that many Tm intermetallic compounds exhibit unusually slow paramagnetic relaxation effects. Such behaviour has been witnessed (by Dixon et al. [1]) in TmAl and TmCu, and in Tm$_3$Al$_2$ by Bowden et al. [2]. In Tm$_3$Al$_2$, for example, the $^{169}$Tm Mössbauer spectrum possesses a well resolved fully split hyperfine spectrum some 30 K above its Néel temperature of 6.0(3) K.

In this paper, we report similar behaviour for the orthorhombic Ni$_2$Si type (Pnma) intermetallic compound Tm$_2$Al. However, unlike the compounds previously investigated, Tm$_2$Al does not exhibit magnetic ordering, at least above 1.4 K. It is also shown that there is little or no cross-relaxation between the two Tm crystallographic sites in this compound.
2. Experimental details and results

The Tm$_2$Al sample was prepared by melting together appropriate quantities of 5N purity aluminium and 3N purity thulium in an argon arc furnace. The sample was turned and re-melted several times to ensure homogenisation. Because thulium possesses a large vapour pressure at elevated temperatures, the estimated loss of thulium was allowed for in weighing out the starting materials. The final composition was within $\approx 0.5\%$ of the ideal. However, as we shall see below, a small amount of Tm$_3$Al$_2$ was present in the sample. The resultant ingot was annealed at a temperature of 800°C for one week. Subsequent X-ray analysis using CuK$_\alpha$ radiation revealed that the indexed peaks were in accord with the crystal structure reported by Buschow and van der Goot [3]. Microscopic examination of the sample revealed that the sample contained less than 2% of impurity phases.

The $^{169}$Tm Mössbauer spectra were obtained using a conventional Mössbauer spectrometer equipped with a xenon proportional counter. More details of the radioactive source used in these experiments, together with the computer routines used to fit the Mössbauer data, are given by Bowden et al. [2] and Bleaney et al. [4]. The resultant spectra can be seen in figs. 1(a) and 1(b).

The analysis of the Mössbauer spectra in the presence of electronic relaxation was based on the phenomenological model of Wickman [5] and Wickman and Wertheim [6]. In this model, relaxation processes involving electronic levels, other than the $|J_z = \pm J\rangle$ ground state, are ignored. The uncertainty concerning the crystal field calculations rules out a more detailed treatment at this time. In the temperature range 4–8 K, the computed relaxation time $\tau$ was found to vary in the range $10^{-10}$ to $10^{-11}$ s. However, these figures should be viewed with some caution, for the reasons given above.

The magnetic measurements were made using a Faraday balance, and the results are summarised in figs. 2 and 3. From an examination of the inverse susceptibility versus temperature curve of fig. 2, it will be seen that there is little evidence for magnetic ordering above 1.4 K. However, the magnetization curves shown in fig. 3, obtained in small applied fields, reveal a small peak at 6 K. Since this is the ordering temperature of Tm$_3$Al$_2$, we believe that the sample is contaminated by a small amount of this material. This conclusion may be further strengthened following and examination of the Er-Al and Y-Al phase diagrams given by Buschow and van Vucht [7]. Both Er$_2$Al and Er$_3$Al$_2$, and their Y counterparts, melt peritectically with Er$_3$Al$_2$ precipitating from the melt first. In practice, therefore, it will be very difficult to prepare a sample of Tm$_2$Al free from contamination. However, we do not believe that the small amount of Tm$_3$Al$_2$ has unduly affected our results. Both the Mössbauer spectra and the magnetization results of Tm$_3$Al are significantly different from those of Tm$_3$Al$_2$.

From an examination of fig. 1, it will be seen that the susceptibility of Tm$_2$Al exhibits Curie, or free-ion-like behaviour, above 70 K. A straight line fit through the