Re-entrant ferromagnetism in the $ThFe_{3-x}Al_x$ system

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Abstract

The magnetic behaviour of $ThFe_{3-x}Al_x$ of which the parent compound displays re-entrant ferromagnetism, has been studied. Substitution of Al leads to an expansion of the lattice which is the largest in the basal plane. The Curie-temperature decreases monotonously from 428 K for $x = 0$ to 341 K for $x = 0.33$. The antiferromagnetic temperature range decreases and is completely suppressed for $x > 0.33$.

Keywords: Re-entrant ferromagnetism; Antiferromagnetism

All RFe_3 compounds with R = rare-earth, Y or Th crystallize in the rhombohedral Be_3Nb-structure [1], (space group $R\bar{3}m$, also called $PuNi_3$-structure) which has three inequivalent Fe-sites and two inequivalent R-sites. The Fe–Fe distances are much smaller than the Fe–R distances. The compound $ThFe_3$ is exceptional in its magnetic properties because it displays so-called re-entrant ferromagnetism [2]. This feature makes the series $ThFe_{3-x}Al_x$ interesting for studying the origin of antiferromagnetism and its dependence on interatomic distances in Fe-based intermetallics.

Samples $ThFe_{3-x}Al_x$ with $x = 0, 0.033, 0.083, 0.167,$ 0.333 were prepared by arc melting starting materials of at least 99.9% purity. After arc-melting the samples were wrapped in Ta-foil, sealed in an evacuated quartz tube and annealed for two weeks at 1000°C. The X-ray diffraction diagrams showed that the crystal structure of all samples corresponds to the rhombohedral Be_3Nb-structure. The samples with $x = 0.033$ and $x = 0.083$ were single phase while the parent compound showed a small ($\sim 5\%$) amount of Th_2Fe_7 (a ferromagnetic phase, also identified by its T_c of 570 K [3]). The samples with higher Al concentration showed a small amount of an unidentified second phase. In Fig. 1 the lattice constants are plotted versus the Al concentration. The expansion is monotonous in both directions and slightly larger in the basal plane than in the c-direction.
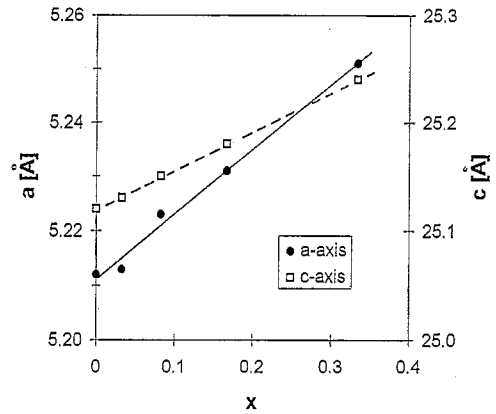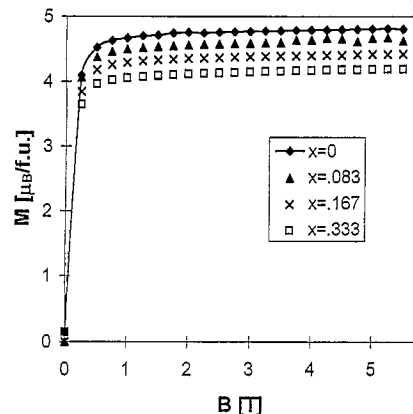

Fig. 1. Lattice constants of $ThFe_{3-x}Al_x$ compounds.

Fig. 2. Magnetic isotherms at 5.0 K for $ThFe_{3-x}Al_x$ compounds, taken in decreasing field. The solid line is a guide to the eye.

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Table 1

Magnetic properties of $ThFe_{3-x}Al_x$. T_m is the minimum of the magnetization, $T_{1,2}$ are the points of steepest slope. For the compound with $x = 0$ a correction has been made for the amount of Th_2Fe_7

Compound	x				
	0	0.033	0.083	0.167	0.333
M_s at 5 K (μ_B /f.u.)	4.79	4.65	4.65	4.43	4.19
M_{Fe} (μ_B)	1.60	1.55	1.56	1.49	1.42
T_1 (K)	188	214	229		
T_m (K)	264	257	245	229	176
T_2 (K)	300	287	268		
T_C (K)	428	422	411	393	341

The magnetic measurements were made on powdered samples in a SQUID magnetometer in the temperature range 5–300 K in magnetic fields up to 5.5 T. Above 300 K, the measurements were made on polycrystalline material on a home-built Faraday balance. The magnetization in decreasing magnetic field is displayed in Fig. 2, and shows ferromagnetic behaviour for all samples at 5 K. Magnetic measurements on the parent compound using the neutron depolarization technique show ferromagnetic behaviour at 5 K in zero field [2], therefore we can conclude that the decrease in magnetization at low fields is due to domain formation. Substitution of Fe by Al causes a decrease in the saturation magnetization, which is due to the dilution of the Fe-sublattices as well as to a decrease in Fe-moment from 1.6 μ_B/Fe for $x = 0$ to 1.4 μ_B/Fe for $x = 0.33$ (see Table 1).

Fig. 3 displays the magnetization as function of temperature in a field of 0.1 T (two samples are left out for reasons of clarity). It shows a decrease in Curie-temperature with increasing Al concentration as can be explained by the dilution and the decrease in Fe-moment.
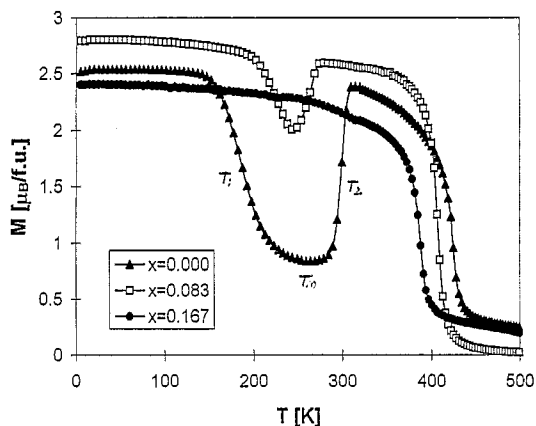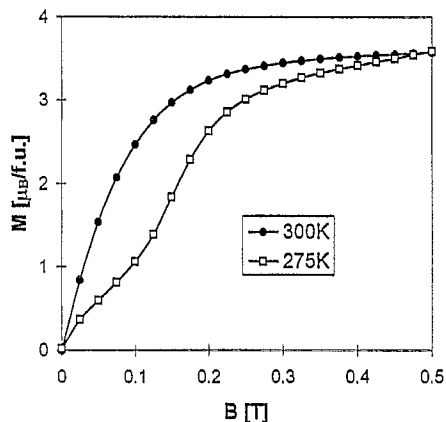

Fig. 4. Magnetic isotherms for $ThFe_3$. The lines are guides to the eye.

In the parent compound antiferromagnetism sets in at $T_2 = 300$ K and disappears at $T_1 = 188$ K. Fig. 4 shows the magnetic behaviour just below and above the second transition. The transitions are not accompanied by a structural change or sudden change in lattice constants [2]. At 275 K, a metamagnetic transition appears around 0.15 T and at 0.5 T the antiferromagnetic behaviour is completely suppressed.

Al substitution limits the temperature range in which antiferromagnetism appears and for $x = 0.167$, the antiferromagnetic behaviour is barely visible. The transition T_2 can tentatively be explained as a first-order antiferro–ferromagnetic transition, which is driven by the thermal expansion. Al substitution expands the lattice which causes a decrease in transition temperature.

The inset of re-entrant ferromagnetism (T_1) shifts to higher temperatures with increasing Al concentration, while the compounds $Th_{0.9}R_{0.1}Fe_3$ (R = Y, Gd or Lu) show antiferromagnetic behaviour down to 5 K [4,5]. Since neither Th nor Y or Lu have a magnetic moment, this indicates that the re-entrant ferromagnetism also critically depends on the Fe–Fe distances. More detailed explanation should await for neutron diffraction measurements, which are currently in progress.

Fig. 3. Temperature dependence of the magnetization of $ThFe_{3-x}Al_x$ compounds at 0.1 T.

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