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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$_3$Nb-structure [1], (space group R3m, 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$_3$Nb-structure. The samples with $x = 0.033$ and $x = 0.083$ were single phase while the parent compound showed a small ( ~ 5%) amount of Th$_2$Fe$_7$ (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.

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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.
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$_2$Fe$_7$.

<table>
<thead>
<tr>
<th>Compound</th>
<th>$x$</th>
<th>0</th>
<th>0.033</th>
<th>0.083</th>
<th>0.167</th>
<th>0.333</th>
</tr>
</thead>
<tbody>
<tr>
<td>$M_s$ at 5 K ($\mu_B$/f.u.)</td>
<td>4.79</td>
<td>4.65</td>
<td>4.65</td>
<td>4.43</td>
<td>4.19</td>
<td></td>
</tr>
<tr>
<td>$M_{Fe}$ ($\mu_B$)</td>
<td>1.60</td>
<td>1.55</td>
<td>1.56</td>
<td>1.49</td>
<td>1.42</td>
<td></td>
</tr>
<tr>
<td>$T_1$ (K)</td>
<td>188</td>
<td>214</td>
<td>229</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$T_m$ (K)</td>
<td>264</td>
<td>257</td>
<td>245</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$T_2$ (K)</td>
<td>300</td>
<td>287</td>
<td>268</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$T_C$ (K)</td>
<td>428</td>
<td>422</td>
<td>411</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

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-subslettes as well as to a decrease in Fe-moment from 1.6 $\mu_B$/Fe for $x = 0$ to 1.4 $\mu_B$/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.

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.

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References