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Published in:
Hyperfine Interactions

DOI:
[10.1007/bf02147350](https://doi.org/10.1007/bf02147350)

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Document Version
Publisher's PDF, also known as Version of record

Publication date:
1983

[Link to publication in University of Groningen/UMCG research database](#)

Citation for published version (APA):

Kraan, A. M. V. D., Buschow, K. H. J., & Palstra, T. T. M. (1983). Magnetic Behaviour of the Cubic La(Fe,Al)₁₃ Compounds. *Hyperfine Interactions*, 16(1). <https://doi.org/10.1007/bf02147350>

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MAGNETIC BEHAVIOUR OF THE CUBIC $\text{La}(\text{Fe},\text{Al})_{13}$ COMPOUNDS

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The magnetic properties of the cubic NaZn_{13} type pseudobinary compounds $\text{LaFe}_x\text{Al}_{13-x}$ were studied in the temperature range $T=4.2 - 300$ K by means of ^{57}Fe -Mössbauer spectroscopy, magnetization and zero-field susceptibility measurements. The compounds $\text{LaFe}_x\text{Al}_{13-x}$ show a rather peculiar concentration dependence of the type of magnetic ordering as well as of the ordering temperature.

1. Introduction

LaCo_{13} , which is strongly ferromagnetic, is the only compound among 45 binary systems consisting of a rare earth element (R) and one of the metals Fe, Co or Ni (T) with the cubic NaZn_{13} type of structure. Kripyakevich et al. [1] showed that the cubic NaZn_{13} type of structure can be stabilized in other binary rare earth transition metal systems (R-T) by substitution of Si for part of the transition metal T in RT_{13} . Previously we have studied $\text{LaFe}_x\text{Si}_{13-x}$ [2] in the concentration region $10.5 \leq x \leq 11.5$. Only in this region single phase samples were obtained. In the present investigation we show that the structure stabilization occurs over a much wider concentration range when part of the transition metal T is substituted by Al. We have studied the magnetic properties of $\text{LaFe}_x\text{Al}_{13-x}$ by means of ^{57}Fe -Mössbauer spectroscopy, magnetization and temperature-dependent zero-field susceptibility measurements.

2. Experimental

The samples of the compounds $\text{LaFe}_x\text{Al}_{13-x}$ were prepared by argon arc melting of the appropriate amounts of the starting materials and vacuum annealed for about 10 days at $T=1200$ K. The samples were investigated by means of X-ray diffraction. Single phase samples of the cubic NaZn_{13} type of structure were obtained in the concentration range $6 \leq x < 12$. For $x < 6$ a contamination with compounds of the tetragonal ThMn_{12} structure is observed in the samples. Microscopic measurements indicate besides the NaZn_{13} type of structure a so far unidentified second phase for $x > 12$.

The magnetic properties of these samples were determined by means of an adapted Faraday method in the range $T=4.2 - 300$ K using magnetic field strengths up to 1.8 T. The temperature dependence of the zero-field susceptibility has been determined with a sensitive pendulum magnetometer in the same T-range.

The ^{57}Fe -Mössbauer spectra were obtained by means of a standard constant-acceleration type spectrometer by using a ^{57}Co -Rh source.

3. Results

The shape of the magnetization versus temperature curve for

$x \leq 11.2$ is typical for a ferromagnet. The values for the saturation moment per iron atom at $T=4.2$ K (μ_s) and the values of the Curie temperature (T_C) are listed for the various compounds in Table 1 and plotted in fig. 1.

Table 1

The lattice constant a , T_C and μ_s determined from magnetization measurements and T'_C and H_{eff} determined from Mössbauer spectrometry of the compounds LaFe_xAl_{13-x}.

x	a(Å)	T _C (K)	μ _s (μ _B /Fe)	T' _C (K)	H _{eff} (T)
6	11.93	-	0	65	12.0
7	11.86	-	0	55	12.0
7.5	-	-	-	70	-
8	11.80	138	1.02	126	14.5
9	11.74	237	1.47	238	19.0
10	11.67	250	1.66	250	22.0
11	11.61	195	2.00	190	26.5
11.8	-	-	0.1	-	-
12.0	-	-	1.0	235	26.0

The saturation moment increases from $\mu_s \approx 0 \mu_B$ for $x = 7$ to $\mu_s \approx 2.0 \mu_B$ for $x = 11$. For $x > 11$ the magnetization is strongly decreasing to about $\mu_s \approx 0.1 \mu_B/\text{Fe}$ for $x = 11.8$. However for $x = 12.0$ $\mu_s \approx 1 \mu_B/\text{Fe}$ is measured. The Curie temperature (T_C) determined from magnetization measurement increases with increasing x and after reaching a maximum for $x = 9.5$ it decreases. For $x > 11$ it was impossible to determine T_C in this way.

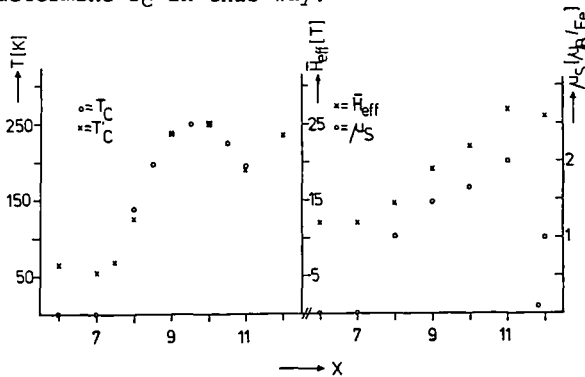


Fig.1. Concentration dependence of the Curie temperature, saturation moment μ_s and mean magnetic hyperfine field H_{eff} in LaFe_xAl_{13-x}

In fig.2 we have plotted the temperature dependence of the zero-field susceptibility for $x = 11.2, 11.5, 11.8$ and 12.0 respectively. The shape of the curve as found for $x = 11.2$ is characteristic for $x < 11.2$, indicating a sharp transition from ferromagnetic to paramagnetic behaviour. However a drastic change in the temperature dependence of the susceptibility is observed for $x > 11.2$.

The Mössbauer spectra obtained at $T=4.2$ K are shown in fig.3. The contribution of the unidentified second phase in the spectrum of LaFe₁₂Al is clearly observed and the outermost lines of this contribution are indicated by arrows in fig.3. The unidentified phase

($\approx 40\%$ of the sample) remains magnetically ordered at $T=238$ K as is shown in fig.4, while the other part of the sample is in the paramagnetic region. It is obvious that the spectrum of the sample with a nominal composition LaFe_4Al_9 really belongs to the compound LaFe_4Al_8 which crystallizes in the ThMn_{12} type of structure [3]. The mean hyperfine fields deduced are listed in Table 1 and have been plotted in fig.1. The mean hyperfine field shows the same concentration dependence as the saturation magnetization from $x = 11$ to $x = 8$. However, for lower values of x , the mean hyperfine field levels off to the value observed for LaFe_4Al_8 (ThMn_{12} structure), which compound orders antiferromagnetically [3]. The values of the magnetic ordering temperature (T'_C) defined as the temperature at which the hyperfine splitting of the Mössbauer spectrum disappears are listed in Table 1 and also plotted in fig.1. A remarkable increase of T'_C for $x = 12$ has been observed after a decrease from $T'_C = 250$ K for $x = 10$ to $T'_C = 190$ K for $x = 11$.

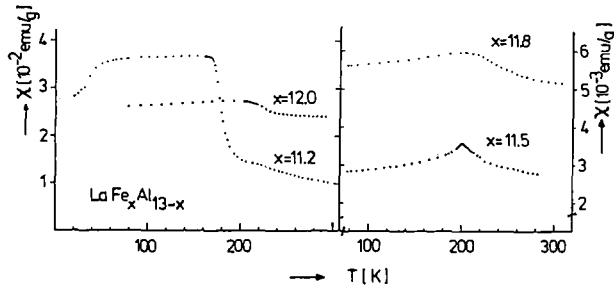


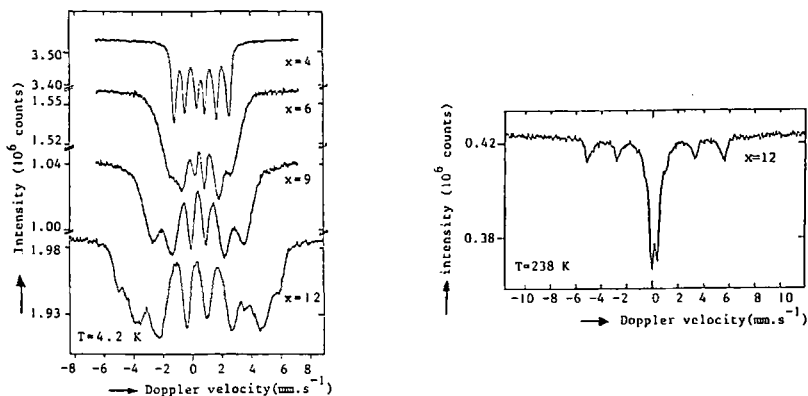
Fig.2. Temperature dependence of the zero field susceptibility χ_0 in $\text{LaFe}_x\text{Al}_{13-x}$.

4. Discussion

The heat of alloying between La and Fe is positive [4]. As a consequence a stable La-Fe intermetallic does not exist. A minimum amount of Al will be required to lead to a pseudo-binary compound of a negative heat of formation and in addition form a suitable structure to accommodate the size of each of the three constituent atomic species.

In the compounds $\text{LaFe}_x\text{Al}_{13-x}$ the moment per Fe atom decreases with decreasing Fe concentration and behaves according to expectation for $8 \leq x \leq 11$. However, for $x \leq 7$ no net magnetization has been found at $T=4.2$ K while a magnetic hyperfine splitting is still observed in the Mössbauer spectrum. We believe that the loss of the net magnetization for $x \leq 7$ is due to formation of a spin glass, which means that for decreasing Fe concentration the ferromagnetic exchange interaction is decreasing faster than the antiferromagnetic one. For $x < 6$ the ThMn_{12} type of structure becomes favoured. The composition of the compound formed is LaFe_4Al_8 , which orders antiferromagnetically [3]. It is interesting that at the low Fe concentration region not only the antiferromagnetic exchange interaction becomes relatively strong, as in LaFe_4Al_8 , but also the magnetic hyperfine field approaches the value observed for LaFe_4Al_8 [3].

In the high Fe concentration region the susceptibility measurements indicate that the pseudo binary compound is changing from entirely ferromagnetic ordering for $x = 11.2$ to an antiferromagnetic ordering for $x \geq 11.5$. This picture is qualitatively consistent with the very low magnetic moment observed for $x = 11.8$ while still a large hyperfine field is observed at the Fe-site. The increase of the

Fig.3. Mössbauer spectra at T=4.2 K for LaFe_xAl_{13-x}.Fig.4. Mössbauer spectrum at T=238 K for LaFe₁₂Al₁.

magnetic moment for $x = 12$ is due to the large contamination of the sample with the so far unidentified second phase.

The opposite concentration dependencies of T_C and magnetic moment (as observed for $10 \leq x \leq 11.2$) were claimed to be associated with the anomalous thermal expansion below T_C found in Invar type alloys[5] and is also observed earlier by us in the compounds LaFe_xSi_{13-x}[2]. In these latter compounds we have observed a cusp like anomaly near T_C in their temperature dependence of the resistivity. Similar measurements on the present samples are in progress. The weakening of the overall ferromagnetic coupling for the compounds in the range $10 \leq x \leq 11.2$ can be understood in terms of the site occupation of the Fe atoms in the NaZn₁₃ crystal structure in conjunction with the lattice constant decreasing with increasing iron content. Denoting the two Fe sites by Fe⁽¹⁾ and Fe⁽²⁾ in the hypothetical compound LaFe₁₂⁽¹⁾Fe₁⁽²⁾, an increasing Fe concentration in LaFe_xAl_{13-x} will lead to a larger occupation of both sites and hence to a larger occupation of the Fe⁽²⁾ site. This site is characterized by a rather low Fe-Fe nearest neighbour separation and by a nearest neighbour configuration consisting of 12 Fe atoms. The Fe⁽²⁾ nearest neighbour configuration is in several respects not much different from that of Fe atoms in γ -Fe. In the latter antiferromagnetism prevails and this may explain why one can expect an increasing antiferromagnetic interaction by filling more of the Fe⁽²⁾ sites in LaFe_xAl_{13-x}.

The increase of the ordering temperature for $x > 11.2$ is not yet understood.

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