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Magnetic properties of $\text{LaFe}_{13-x}\text{Al}_x\text{N}_y$ compounds

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Abstract

Interstitial nitrides of the type $\text{LaFe}_{13-x}\text{Al}_x\text{N}_y$ have been synthesized and their magnetic properties have been investigated. It is found that the magnetic properties are drastically modified by the introduction of interstitial nitrogen. The physical origin of the changes is discussed.

The magnetic properties of the $\text{LaFe}_{13-x}\text{Al}_x$ compounds have been investigated by Palstra et al. [1,2], who found that this series displays complex magnetic behaviour. The magnetic properties vary dramatically with the concentration of aluminum. When the Al concentration is higher than $x = 5$, the ferromagnetic state is unstable with respect to the Fe–Al–Fe indirect exchange in the low-iron-concentration regime, leading to mictomagnetism. When the Al concentration is lower than $x = 1.8$, the ferromagnetic state is destroyed by the direct antiferromagnetic exchange in the high-iron-concentration regime, leading to an antiferromagnetic state.

We have synthesized the interstitial compounds $\text{LaFe}_{13-x}\text{Al}_x\text{N}_y$ [3] and investigated the effect of the interstitial nitrogen atoms on the magnetic properties of the compounds. Nitrogenation of the compounds was carried out by heating the powder of the compounds in the temperature range from 500 to 650°C, during 3 to 30 h, depending on the compound. The nitrogen contents were determined by weighing the mass gain after the nitrogenation. The introduction of nitrogen becomes more difficult with increasing Al concentration. More impurity α -Fe was detected in the nitrides with higher x , whereas the X-ray diffraction patterns show that $\text{LaFe}_{11.5}\text{Al}_{1.5}\text{N}_{2.9}$ and $\text{LaFe}_{10.5}\text{Al}_{2.5}\text{N}_{2.7}$ are single phase. For $x > 4.5$, no nitrogen can be introduced. As a result of the decrease in nitrogen take-up with increasing Al concentration all compounds $\text{LaFe}_{13-x}\text{Al}_x\text{N}_y$ studied have approximately the same unit-cell volume.

We investigated the temperature dependence of magnetization of both the parent and the nitrogenated compounds from 4.2 K to room temperature in a low static field (0.04 T). Fig. 1 shows the results. The curves for $x = 1.5$ in Fig. 1 show that the magnetism of $\text{LaFe}_{11.5}\text{Al}_{1.5}$ has changed

upon nitrogenation, and that the compound has become ferromagnetic from its original antiferromagnetic state. The Curie temperature of the nitride is about 250 K, which is consistent with the ac-susceptibility result [3].

It has been pointed out in Ref. [2] that the antiferromagnetic state occurs in the compounds $\text{LaFe}_{13-x}\text{Al}_x$ with $1.0 \leq x < 1.8$, which is attributed to the very high Fe–Fe coordination number. In this concentration range, a considerable fraction of the Fe sites has an Fe–Fe coordination number approaching 12, which results in antiferromagnetic coupling between the two Fe sites [4]. It is reasonable to assume that the high Fe–Fe coordination number is reduced by the introduction of the interstitial nitrogen atoms upon the nitrogenation. The ferromagnetic state recovers upon this change. A more specific conclusion can only be drawn after the location of the interstitial nitrogen atoms has been established.

From the curves for $x = 2.5$, 3.5 and 4.5 in Fig. 1, it can be seen that the Curie temperatures of $\text{LaFe}_{10.5}\text{Al}_{2.5}$, $\text{LaFe}_{9.5}\text{Al}_{3.5}$ and $\text{LaFe}_{8.5}\text{Al}_{4.5}$ are about 230, 245 and 200 K respectively, which is consistent with the results obtained by Palstra [4]. For $\text{LaFe}_{10.5}\text{Al}_{2.5}\text{N}_{2.6}$, $\text{LaFe}_{9.5}\text{Al}_{3.5}\text{N}_{2.0}$ and $\text{LaFe}_{8.5}\text{Al}_{4.5}\text{N}_{1.7}$, the Curie temperatures have shifted to above room temperature. In order to determine the Curie temperatures of these three nitrides, we measured the temperature dependence of the magnetization up to 1000 K with a Faraday balance in an atmosphere of helium gas. The field strength used in the measurements was 0.05 T. Fig. 2 shows the M – T curves of $\text{LaFe}_{10.5}\text{Al}_{2.5}\text{N}_{2.6}$. To check whether the compounds have been oxidized or have decomposed, we measured the magnetization both with increasing and with decreasing temperature. From Fig. 2 the Curie temperature can be determined to be 825 K. The lower magnetization of the curve measured with decreasing temperature may be due to the outgas of nitrogen at high temperatures. A similar increase of the Curie temperatures has also been found in the nitrides with higher Al concentration [3].

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The increase in the Curie temperature of $\text{LaFe}_{10.5}\text{Al}_{2.5}\text{N}_{2.6}$ is about 600 K. This increase is even bigger than in $\text{R}_2\text{Fe}_{17}\text{N}_x$ compounds, where one finds that the Curie temperatures are increased upon nitrogenation by up to about 400 K, which may be connected with the expansion of the unit cells. It has been pointed out in Ref. [4] that for $\text{LaFe}_{13-x}\text{Al}_x$ with $x = 2.5$, the Fe–Fe distance is in the range where the exchange constant increases with increasing Fe–Fe distance. The expansion of the unit-cell volume in $\text{LaFe}_{13-x}\text{Al}_x$ upon nitrogenation is quite large. The expansion for the compound with $x = 2.5$ is 8%, which is

Fig. 1. M - T plots in a magnetic field of 0.04 T for $\text{LaFe}_{13-x}\text{Al}_x$ and $\text{LaFe}_{13-x}\text{Al}_x\text{N}_y$. The (■) represent the nitrides and the (□) represent the parent compounds. The relatively big change in magnetization of the nitrides at low temperatures (below 20 K) may be related to the motion of domain walls.

Fig. 2. M - T plots of $\text{LaFe}_{10.5}\text{Al}_{2.5}\text{N}_{2.6}$ measured in a Faraday balance. The arrows indicate whether the measurement has been performed with increasing or decreasing temperature. All the lines are guides to the eyes. The relatively big change in magnetization at the beginning of the measurements may be related to the motion of domain walls.

larger than the expansion in $\text{R}_2\text{Fe}_{17}\text{N}_x$ compounds. The large expansion results in a very strong increase of the Curie temperature. However, it is still not possible to be more specific about the increase of the Fe–Fe distance before we know the location of nitrogen in the complex cubic structure.

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