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Magnetic and structural properties of the $Lu_2Fe_{17}H_y$ hydrides

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ABSTRACT

The structural and magnetic parameters of the $Lu_2Fe_{17}H_y$, $y = 0–1.77$ hydrides were studied and compared with those of the $Lu_2Fe_{17-x}Mn_x$ system. The hydrides like their initial compound are ferromagnets at low temperatures and antiferromagnets at high temperatures. Spontaneous magnetization of the hydrides is virtually independent on hydrogen content. The lattice parameters of the $Lu_2Fe_{17}H_y$ hydrides increase relatively fast with y whereas their ferromagnetism, as compared with the $Lu_2Fe_{17-x}Mn_x$ system, strengthens slowly. Hence, there appears an indirect evidence that the unit cell volume increase is not the main reason for the surprising abrupt ferromagnetism strengthening observed in the $Lu_2Fe_{17-x}Mn_x$ system.

1. Introduction

Magnetic $T–x$ phase diagrams of the $R_2Fe_{17-x}Mn_x$ compounds (R—rare earth metal) are rather various. The Curie temperature T_C of the $R_2Fe_{17-x}Mn_x$ ferromagnets with R = Pr, Nd, Sm, Y and ferrimagnets with Gd, Tb, Er decrease monotonously as the Mn content increases (see Refs. in [1]). In the case of $Ce_2Fe_{17-x}Mn_x$, the ferromagnet-to-antiferromagnet transition temperature $\Theta_T(x)$ decreases so abrupt with x, that the ferromagnetism disappears already for $x = 0.5$ and the compounds with $x = 0.5–1$ are already helical antiferromagnets. However, the compounds with $x > 1$ are ferromagnets again [2–4]. The opposite behaviour has been observed in the $Lu_2Fe_{17-x}Mn_x$ system [1]. The transition temperature $\Theta_T(x)$ increases instead of decreasing with x.

The reasons of different changes of the ground magnetic state in the $R_2Fe_{17-x}Mn_x$ systems are not fully understood at that moment. It is commonly assumed that the magnetic state and Curie temperature of R_2Fe_{17} systems are determined by the competition of positive and negative exchange interactions between 3d atoms [5]. The positive interaction dominates if the distances between 3d atoms are higher than the critical one. It means that the role of unit cell volume is essential for magnetic ground state of these compounds. In the compounds doped with Mn, the Mn moments couple antiparallel to those of Fe. Insertion of hydrogen or nitrogen and carbon to the any metallic alloy lattice usually increases interatomic distances. When introduced they did the same in the R_2Fe_{17} compounds, and strengthening of positive exchange interactions between iron atoms are observed [6–9].

In this report, the magnetic and lattice parameters of the $Lu_2Fe_{17}H_y$ hydrides are presented and a comparison with the $Lu_2Fe_{17-x}Mn_x$ compounds is made [1].

2. Experimental results and discussion

The $Lu_2Fe_{17}H_y$ hydride samples with $y = 0–1.77$ were prepared in a volumetric apparatus through the direct absorption of hydrogen by metallic sample at 423 K. The hydrogen content y in the samples was determined with an accuracy of ±0.02 by measurements of the hydrogen pressure drop in a calibrated apparatus volume. According to X-ray analysis, all $Lu_2Fe_{17}H_y$ hydride samples had hexagonal crystal structure of the Th_2Ni_{17}-type. The lattice parameters were calculated using the DICVOL program. Magnetization measurements were performed with SQUID magnetometer. The temperature of magnetic phase transitions were determined from $M(T)$

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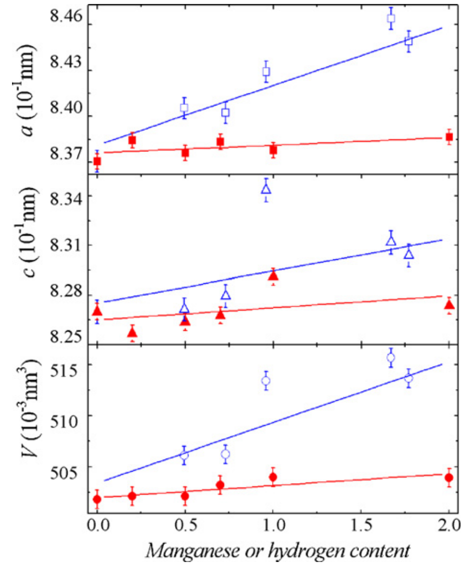

Fig. 1. (Color online) Composition dependence of the unit cell parameters in the $Lu_2Fe_{17-x}Mn_x$ (■), (▲), (●) [1] alloys and in $Lu_2Fe_{17}H_y$ (□), (△), (○) hydrides at 295 K.

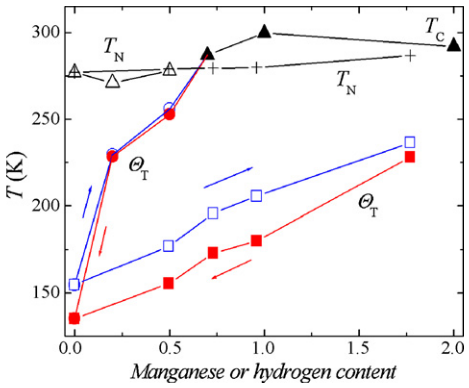

Fig. 2. (Color online) Composition dependence of the Néel temperature T_N (△ and +), the temperature of transition from ferromagnet to antiferromagnet Θ_T (○), (●) and (□), (■) for the $Lu_2Fe_{17-x}Mn_x$ [1] and $Lu_2Fe_{17}H_y$ compounds, respectively. Curves labeled (○), (□) and (●), (■) correspond to the heating and cooling procedure of measurement, respectively. The (▲) curve displays the Curie, T_C, temperature dependence for $Lu_2Fe_{17-x}Mn_x$ alloys.

dependences taken in the field 100 Oe in the temperature range of 2–400 K for polycrystalline samples. Because of hysteresis on $M(T)$ curves, two sets of Θ_T parameters were determined for the heating and cooling processes, respectively. The saturation magnetization $M_{sat.}$ was determined by extrapolation of experimental dependences $M(1/H)$ to $1/H \to 0$ measured for the free powder samples.

The lattice constants a, c and the unit cell volume V measured at room temperature for the $Lu_2Fe_{17}H_y$, $y = 0$–1.77 hydrides are shown in Fig. 1 as a function of y together with the lattice constants in the $Lu_2Fe_{17-x}Mn_x$ system in the range of $x = 0$–2 [1]. There is observed a much quick increase of the lattice constants with H content as compare to such changes with Mn content.

The T_N is virtually constant, whereas Θ_T increases by a factor of ~1.5 or ~2 in the case of $Lu_2Fe_{17}H_y$ and $Lu_2Fe_{17-x}Mn_x$ [1], respectively (Fig. 2). Ferromagnetism strengthening in the $Lu_2Fe_{17}H_y$ hydrides for $y = 0$–1.77 is analogous to those observed in the other $R_2Fe_{17}H_y$ systems and was interpreted as due to the increase of the 3d interatomic distances [6–9]. Spontaneous magnetization of the $Lu_2Fe_{17}H_y$ hydrides is virtually independent of hydrogen content.

Thus, the crystal unit cell volume of the initial Lu_2Fe_{17} compound enlarges visibly slowly with partial substitution of Fe by Mn (Fig. 1), whereas its ferromagnetism strengthens much stronger (Fig. 2) as compare with hydrogen insertion effect. This is an indirect evidence that the unit cell volume increase is not the main reason for the ferromagnetism strengthening in the $Lu_2Fe_{17-x}Mn_x$ system as it was proposed in recently published paper [1]. The strong enhancement of positive exchange interactions with the increasing of x in $Lu_2Fe_{17-x}Mn_x$ could be rather explained as a result of the local exchange interactions between 3d atoms, which are located in the crystal lattice defects [1]. According to Givord et al. [10] the Lu_2Fe_{17} crystal structure is not the ideal of Th_2Ni_{17}-type but rather is the disordered $LuFe_{9.5}$-type structure.

The magnetic parameters of the studied $Lu_2Fe_{17}H_y$ hydrides are very similar to those published in the paper [8], but they are different from that reported in the paper [9], where the hydride sample with $y = 0.8$ is already a ferromagnet. Apparently, also in this case the more or less distorted crystal structure of the initial Lu_2Fe_{17} compounds prepared by different authors [1,6,8–10] is probable responsible for the reported different magnetic parameters for their $Lu_2Fe_{17}H_y$ hydride samples.

In conclusion, owing to comparison of structural and magnetic parameters of the $Lu_2Fe_{17}H_y$ and $Lu_2Fe_{17-x}Mn_x$ systems, indirect evidence was established that the previously suggested unit cell volume increase is not the main reason for the abrupt ferromagnetism strengthening in the $Lu_2Fe_{17-x}Mn_x$ system.

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