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Journal of Alloys and Compounds 293–295 (1999) 237–242

Journal of ALLOYS AND COMPOUNDS

The influence of hydrogen on the magnetic properties and electronic structures of intermetallic compounds: YFe_2–D_2 system as an example

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Abstract

The magnetic and electronic properties of YFe_2D_x deuterides ($1.2 \leq x \leq 4.2$) have been studied by bulk magnetic measurements and X ray absorption spectroscopy (XAS). The magnetic moment increases with deuterium content whereas the Curie temperature increases from 560 K ($x=0$) to 720 K ($x=1.2$) then decreases almost linearly down to 300 K ($x=4.2$). For $x=1.2$, 1.75 and 1.9, structural transitions from distorted structures to C15 cubic structure occurs below the Curie temperature and are therefore related to deuterium ordering. The analysis of the XAS spectra at the Fe–K edge shows a progressive filling of the 4p-3d band with increasing deuterium content. The evolution of the magnetic and electronic properties of the YFe_2D_x deuterides are discussed in relation with the evolution of the Fe–Fe distances in these compounds and band structure calculations.

Keywords: Hydrides; Magnetism; Electronic structure; X-ray absorption

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1. Introduction

The magnetic properties and electronic structures of rare earth (R) and transition metal (M) intermetallic compounds are generally strongly modified by hydrogen absorption. It can lead to a change of magnetic order, of the magnitude of the total magnetic moment and of the ordering temperatures [1,2]. Hydrogen absorption also modifies the electronic structure of the host compound by the creation of new M–H bonding states, the shift of the Fermi level and the modification of the symmetry and the width of the bands [3,4]. The interesting magnetic properties of the RM_n intermetallic compounds are the results of the interaction of 4f and 3d moments. The magnetic moments of rare earth atoms arise from the 4f electrons and are well localized whereas those of transition metal atoms are due to the d conduction electrons which are more itinerant. The RM_n compounds are characterized by three kinds of magnetic interactions between R–R, R–M and M–M atoms respectively. The R–R and R–M interactions can be described with by RKKY interaction, which occurs via the 5d conduction electrons, or by indirect exchange. The M–M interactions, which are the strongest, lead to more controversial interpretations and a large variety of localized or band models have been developed to describe them [5]. Hydrogen absorption in the RM_n compounds leads to an increase of the cell volume and therefore to a change of the interatomic distances which will modify these magnetic interactions. When the 3d magnetism is dominant, hydrogen absorption generally leads to a reduction of the 3d moment for Ni and Co compounds, to an enhancement of the 3d moment for Fe compounds and to a change of magnetic order for Mn compounds. The magnetic ordering temperatures may vary in opposite directions upon hydrogen absorption depending on the host compounds. For example, in the case of Fe compounds a reduction of T_c is observed for RFe_2 hydrides [6] whereas it increases for R_6Fe_{23} [7] or R_2Fe_{17} hydrides [8]. Buschow [9] has explained these different variations of T_c upon H absorption in RFe_n compounds by a negative pressure effect. The sign of the variation of T_c in the hydrides was opposite to that observed under applying a pressure, which reduces the cell volume. According to the Stoner-Wohlfarth criterion the band width, the density of state and the intra-atomic exchange interaction are sensitive to pressure. The Curie temperatures of the ternary hydrides can be understood as a pressure dependent parameter of the Stoner-Wohlfarth model as it has been observed for R_2Fe_{17} hydrides, for which a linear relationship was established between $\Gamma = \delta \ln(T_c)/\delta \ln(V)$ and T_c^{-2} [8].

In order to have a better understanding of the influence of the hydrogen on the Fe–Fe interactions, we have undertaken a detailed study of the structural, magnetic and

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0925-8388/99/$ – see front matter
PII: S0925-8388(99)00323-0

electronic properties of the YFe_2 deuterides as a function of hydrogen content. YFe_2 is a Laves phase compound which crystallizes in the C15 type cubic structure ($a = 7.36$ Å) and is ferromagnetic with a Curie temperature of 560 K and a total magnetization of 2.9 μB/f.u [10]. It can absorb large amount of hydrogen up to 4.2 H/f.u. and several single phase hydrides were identified in the range $1.2 \leq x \leq 4.2$ [11,12]. These phases are characterized not only by a cell volume augmentation with increasing deuterium content but also by different structures derived from the cubic C15 structure of the parent YFe_2 compound.

Concerning the magnetic properties of the YFe_2H_x hydrides controversial results has been published for the values of T_c in YFe_2H_4 with $T_c = 310$ K [10], 133 K [13] or 120 K [14]. However in a previous work we have observed that the evolution of the Curie temperature of YFe_2D_x deuterides was very sensitive to the change of crystal structure and deuterium content [15]. The change of electronic properties of YFe_2 by H absorption was studied by a resonant photoemission experiment, performed to measure the angular dependence of the valence band of YFe_2H_x [16]. As the hydrogen exposure time and temperature increase, the XPS spectra show a sharpening of a band located at 5.3 eV, the formation of a new structure at 10 eV and the progressive decrease of the d like states at the Fermi level E_F. The band located at 5.3 eV was attributed to the new s-p states due to hydrogen. However although these results give a qualitative evolution of the density of states at the Fermi level upon H absorption, the corresponding amount of deuterium cannot be determined. X-ray absorption experiments can be used to follow quantitatively the influence of hydrogen on the empty DOS. For example in the case of $LaNi_5$, $LaNi_{4.8}Sn_{0.2}$ and $La_{0.8}Ce_{0.2}Ni_{4.8}Sn_{0.2}$ compounds the modifications of the Ni K edge upon hydrogen absorption [17] were correlated with band structure calculations of M. Gupta on $LaNi_5$ and its hydride $LaNi_5H_6$ [18].

In this work, we will present results on the magnetic and electronic properties of single phases YFe_2D_x deuterides ($1.2 \leq x \leq 4.2$) studied by bulk magnetic measurements and by X-ray absorption spectroscopy. These results will be discussed in relation with neutron diffraction data, Mössbauer spectroscopy experiment and theoretical band structure calculations.

2. Experimental

The preparation and the characterization of the YFe_2 intermetallic compound and its deuterides are reported in Refs. [11,12]. The magnetic measurements were performed on a DSM8 magneto-susceptometer with an applied magnetic field up to 1.6 T. The evolution of the magnetization as a function of the temperature was measured with an applied field of 1 T and from 4.2 to 600 K. The measurements of the intrinsic magnetic properties of the deuterides was limited to 600 K, since they decompose above this temperature into YH_x and α-Fe. The X-ray absorption spectra have been registered by transmission at 13 K and 300 K at the Fe–K edge and Y–K edge on the XAS II spectrometer on the DCI ring of LURE (Orsay, France).

3. Experimental results

The structures of the single phase deuterides, were determined by X ray and neutron diffraction at 300 K [19] and their cell parameters are reported in Table 1. Three different structural behaviours were evidenced. For low deuterium contents ($x = 1.2$, 1.75 and 1.9) the deuterides are characterized by the existence of weak superstructure lines which arise from the displacement of Y and Fe atoms and deuterium ordering in preferential Y2Fe2 interstitial sites. For these three deuterides the superstructure lines disappear reversibly at $T_s = 460$ K, 442 K and 440 K, for

Table 1
Structural and magnetic results for YFe_2D_x compounds, M_s corresponds to the saturation magnetization at 4.2 K, T_s to the structural transitions and T_c to the Curie temperatures

Compound	Structures	Cell parameters (Å)	M_s (μB) at 4.2 K	T_s (K)	T_c (K)
YFe_2	cubic	$a = 7.36$	2.90(5)		560
$YFe_2D_{1.2}$	tetragonal	$a = 11.98$ $c = 7.61$	3.00(5)	460	720[a]
$YFe_2D_{1.75}$	cubic	$a = 15.34$	3.06(5)	442	650[a]
$YFe_2D_{1.9}$	tetragonal	$a = 12.15$ $c = 23.07$	3.30(5)	440	640[a]
$YFe_2D_{2.6}$	cubic	$a = 7.783$	3.20(5)		465
$YFe_2D_{2.9}$	cubic	$a = 7.828$	3.38(5)		435
$YFe_2D_{3.3}$	rhombohedral	$a = 5.508$ $\alpha = 61.43$	3.68(5)	350	363
$YFe_2D_{4.2}$	rhombohedral	$a = 5.518$ $\alpha = 62.75$	3.62(5)	100	300

[a] The values of T_c are calculated using $M_s(T)M_s(0) = f\ (T/T_c)$.

$x=1.2$, 1.75 and 1.9, respectively, leading to the cubic C15 structure above T_s. Then for $x=2.6$, 2.7 and 2.9, the deuterides crystallize in the cubic C15 structure with a statistical occupation of deuterium in Y2Fe2 interstitial sites. For $x>3$ the deuterides display a rhombohedral structure due to a contraction along the 111 axis of the cubic cell. At 350 K, the rhombohedral distortion of $YFe_2D_{3.5}$ disappears, but some superstructure lines due to a preferential occupation of deuterium in 2 over 3 Y2Fe2 sites are still observed [20]

The study of the local order ^{57}Mössbauer spectroscopy and EXAFS at 13 K and 300 K, has also revealed the existence of several Fe sites and a large distribution of interatomic distances for all the deuterides [21].

3.1. Magnetic measurements

The magnetization curves of YFe_2D_x compounds at 4.2 K are reported on Fig. 1. Up to $x=2.9$ the magnetization curves display a similar ferromagnetic behaviour, whereas for the rhombohedral phases $x=3.3$ and 4.2 the saturation is not reached at 1.6 T. The saturation magnetization M_s was extrapolated from the magnetization curves and are reported versus D content on Fig. 2 and in Table 1. Despite non saturation the values of M_s for the rhombohedral phases (3.7 μ_B) are in agreement with previous works [10,14]. For $x=1.2$ and 1.75, M_s increases only slightly, then more sharply up to 3.7 μB for $x=3.3$ and remains constant for $x=4.2$.

The evolution of the magnetization as a function of the temperature is reported on Fig. 3 for $0 \leq x \leq 2.9$. For $x=1.2$, 1.75 and 1.9, a weak increase of the magnetization is observed at 460 K, 442 K and 440 K respectively. These temperatures are the same than those observed for the structural order–disorder transitions T_s. The Curie temperatures of these three deuterides are higher than that of YFe_2 but due to the deuterides decomposition, they cannot be measured directly. In order to estimate the Curie temperatures T_c of these deuterides with $x=1.2$, 1.9 and 1.75 we have assumed that the reduced magnetization curves $M(T)/M_s(0)=f\ (T/T_c)$ were similar for all the deuterides ($1.2 \leq x \leq 3.3$). The values of T_c were adjusted in order to superpose all the M(T) curves from 4.2 to 600 K divided by M_s (4.2 K). According to this method the estimated Curie temperatures are 720±10 K for $x=1.2$, 650±10 K for $x=1.75$ and 640±10 K for $x=1.9$. For $x=2.6$ and 2.9 the thermal evolution of the magnetization curves shows that their Curie temperatures (465 K and 435 K) are lower than that of YFe_2 and decrease with deuterium content.

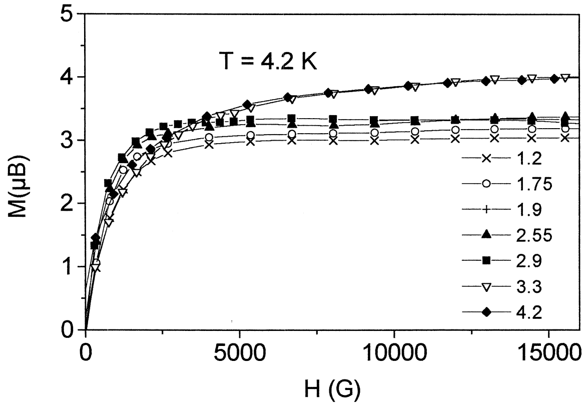

Fig. 1. Magnetization curves at 4.2 K of YFe_2D_x deuterides.

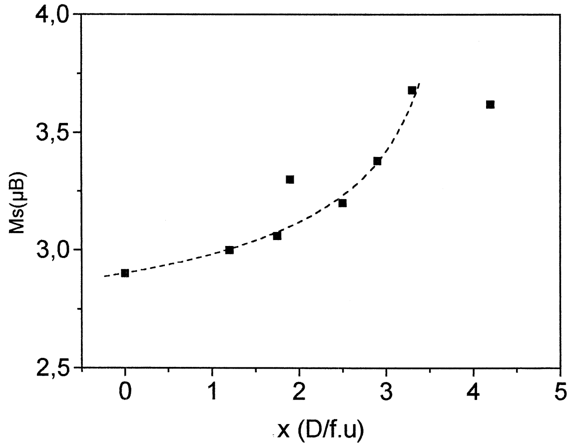

Fig. 2. Evolution of the saturation magnetization of YFe_2D_x at 4.2 K as a function of D content.

For the rhombohedral phases the evolution of the magnetization versus temperature are reported on Fig. 4. For $YFe_2D_{3.3}$ the Curie temperature is 340 K close to that measured previously for $YFe_2D_{3.5}$ [15]. The small bump at 350 K can be attributed to a structural transition from the rhombohedral to the cubic structure in agreement with the previous neutron diffraction study on $YFe_2D_{3.5}$ [20]. The evolution of the magnetization of $YFe_2D_{4.2}$ indicates a sharp transition around 100 K, then the magnetization decreases down to room temperature. Two anomalies

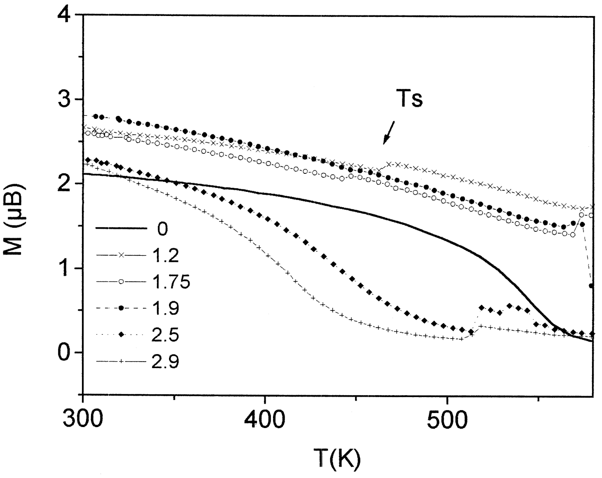

Fig. 3. Magnetization versus temperature for YFe_2D_x deuterides ($0 \leq x \leq 2.9$).

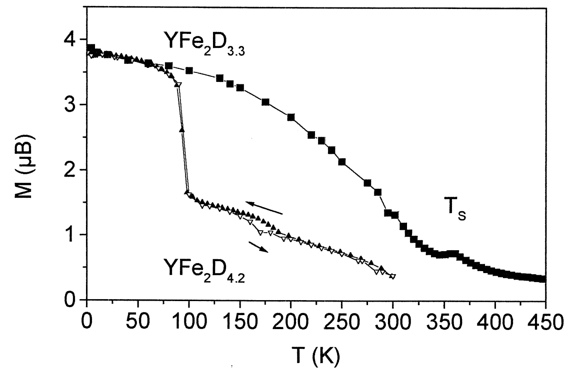

Fig. 4. Magnetization versus temperature for $YFe_2D_{3.5}$, $YFe_2D_{4.2}$.

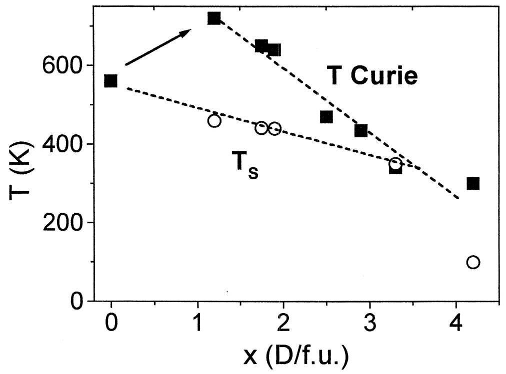

Fig. 6. Evolution of the Curie temperature T_c and of the structural transition T_s as a function of D content.

accompanied by thermal hysteresis effects are also observed around 170 and 260 K. At 300 K the magnetization is very weak, indicating that we are in the vicinity of the Curie temperature. Magnetization curves measured around 100 K clearly indicate that this sharp transition is field dependent with large hysteresis effects (Fig. 5). Such a magnetic behaviour has been already observed for $Sc_{0.25}Ti_{0.75}Fe_2$ but with much larger applied magnetic field (5–15 T) and has been explained by a ferromagnetic–ferromagnetic transition [22].

The evolution of the temperatures corresponding to the order–disorder transitions T_s and to the Curie temperatures T_c of the deuterides as a function of D content is reported on Fig. 6 and in Table 1. First we can notice that the structural transitions observed for $x=1.2$, 1.75 and 1.9 are below the Curie temperatures and cannot be explained by a magnetostrictive behaviour as previously proposed [21]. The weakness of the magnetic anomalies indicates that below their structural transition temperatures, these deuterides have a mean ferromagnetic structure close to that of YFe_2 in agreement with neutron diffraction results at 300 K [19]. The structural transitions should be therefore related to the deuterium ordering inside the lattice, producing a displacement of Fe and Y atoms. The second point is that upon hydrogen absorption the Curie temperature first increases compared to that of YFe_2, then decreases almost linearly as a function of the cell volume.

3.2. X ray absorption

The XAS spectra of the YFe_2D_x compounds have been registered for the Y–K edge and Fe–K edge at 13 K and 300 K. However for a given compound the same near edge structure was observed at 13 K and 300 K.

The XAS spectra of YFe_2 and its hydrides at the Y–K edge show weak changes in the first part of the absorption edge i.e. a decrease of the pre-edge intensity, but the experimental resolution is poor at this energy (16 965 eV) due to the hole life time (Fig. 7) and details are difficult to observe. More significant changes are seen at the Fe–K edge (7115 eV) (Fig. 8). On the spectra of YFe_2 a thin peak centered at 7110 eV is observed in the edge step. As the deuterium content increases the intensity of this peak progressively decreases. The variation of the normalized integrated intensity of this peak is reported on Fig. 9. The same evolution is observed at 13 K and 300 K showing a progressive decrease of the intensity with deuterium content.

Due to the dipolar selection rules, the K edge corresponds to an electronic transition from the 1s core state to

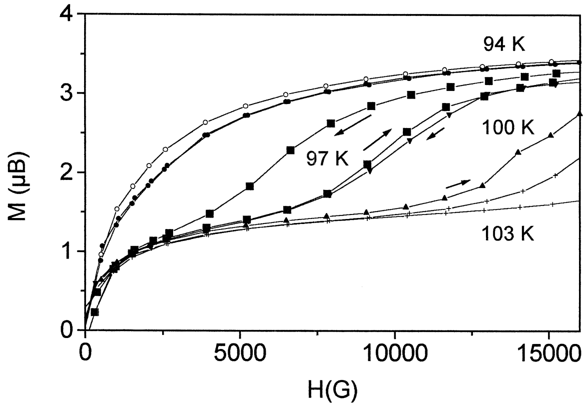

Fig. 5. Magnetization curves for $YFe_2D_{4.2}$ around the transition at 100 K.

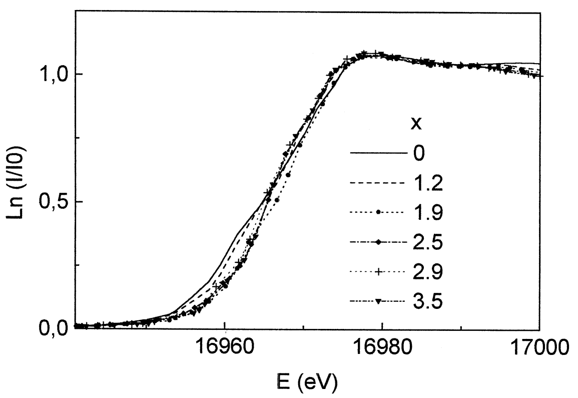

Fig. 7. XAS spectra at the Y–K edge of YFe_2D_x deuterides at 13 K.

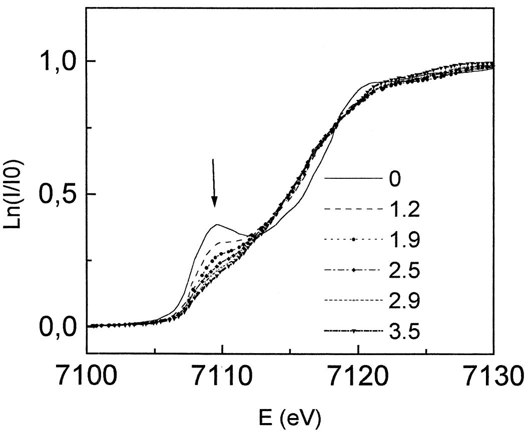

Fig. 8. XAS spectra at the Fe K edge of YFe_2D_x deuterides at 13 K. The arrow indicates the peak due to the transition to empty p-d states.

empty p states and the XAS near edge structure probes the empty projected local electronic density of p states (*D* (E)). However for Fe in YFe_2 the 4p states are hybridized with 3d states which are partially empty and transitions to the mixed empty p-d states can be observed. Therefore the structure at 7110 eV can be related to empty 4p and 3d states. Since band structure calculations show that in YFe_2 the Fe 3d states are dominant at the Fermi level [23], the intensity decrease may indicates a reduction of the number of 3d holes due to the shift of the 3d band below the Fermi Level upon hydrogen absorption. The same effect should occur at the Y–K edge due to the decrease of the pre-edge intensity, but as the intensity of 4d band of Y is weak in the DOS at the Fermi level, it may also explain that weaker changes are observed at the Y K edge.

4. Discussion

The evolution of the magnetic properties of YFe_2D_x deuterides as a function of deuterium content shows that the Fe moment progressively increases up to 3.7 μB for $x=3.3$ and remains almost constant for larger x values whereas the Curie temperature increases from 560 K for YFe_2 to 720 K for $YFe_2D_{1.2}$ then decrease down to 300 K for $YFe_2D_{4.2}$. In addition XAS experiments at the Fe K edge have shown a progressive filling of the 4p-3d band of Fe upon H absorption.

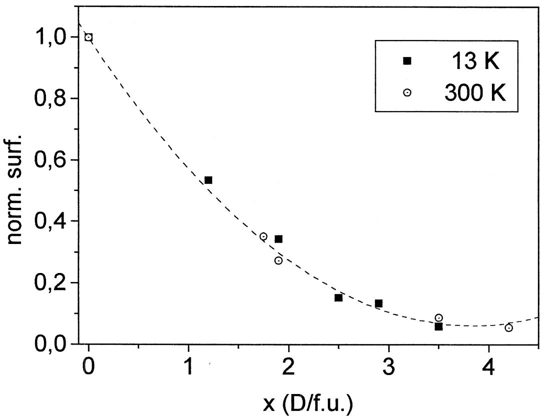

Fig. 9. Evolution of the normalized surface of the peak at 7110 eV at the Fe–K edge for YFe_2D_x compounds.

The increase of the Fe moment with hydrogen content, is generally explained by the increase of the Fe–Fe distances, which leads to a better localization of the Fe moments and therefore to a narrowing of the 3d band. A dependence of the band width on the interatomic distance $W\approx(d_{M-M})^5$ with $N(E)\approx W^{-1}$ was established [24]. Assuming that the deuterides crystallize in the C15 structure, band structure calculations show a progressive increases of the Fe moment as the deuterium content and the cell volume increase [23].

Experimentally, it is observed that the Fe moment is not increasing linearly with D content. For example M_s increases only slightly for low D content and saturates above 3 D/f.u. The evolution of the average hyperfine field (HFF) measured by Mössbauer spectroscopy [21] indicates also a non linear behaviour with D content: HFF increases only slightly up to 2 D/f.u (21.2 to 22 T), then linearly up to 2.9 D/f.u (22 to 26.5 T) and decrease again for $x=4.2$ D/f.u. (21 T). These results show that the mean Fe moment is very sensitive to the distribution of Fe–Fe distances and to the changes of crystal structure. For low D content, a large distribution of Fe–Fe distances is observed (2.4 Å $<d_{Fe-Fe}<$3.0 Å for $x=1.2$ and 1.75) and μ_{Fe} remains close to the value obtained for YFe_2. Then for $2.6\le x\le 2.9$, the deuterides have a C15 cubic structure and HFF increases linearly with x. For $x\ge 3.3$, a rhombohedral distortion occurs and a distribution of Fe–Fe distances should again be considered. For $x=4.2$, the saturation magnetization M_s is similar to that obtained for $x=3.3$ whereas HFF strongly decrease. This discrepancy for $x=4.2$ can be explained by the fact that the increase of M_s below 100 K depends of the applied magnetic field and that in Mössbauer experiments no external field was applied.

Considering the evolution of the Curie temperatures with D content they cannot be only explained by the increase of the average cell volume, since in this case they should all be lower than that of YFe_2 according to the negative pressure model. For low D content, the increase of T_c compared to YFe_2 can results from the presence of significant larger Fe–Fe distances. For Y_6Fe_{23} and Y_2Fe_{17} or $Y_2Fe_{14}B$, the increase of T_c resulting from H absorption, was related to the increase of Fe–Fe distances and of the average Fe moments [25]. As the deuterium content increases, the average cell volume increases and the distribution of Fe–Fe distances becomes thinner. This means that the amount of large Fe–Fe distances decreases and that the influence of the negative pressure dominates leading to a decrease of T_c. The linear decrease of T_c

observed for larger D content is in agreement with that observed for other RFe_2 hydrides (R = Ho, Er, Tm and Lu) [6], since the same variation of $T_c = f(a/a_0)$ is obtained. The magnitude of $\Gamma = \delta\mathrm{Ln}(T_c)/\delta\mathrm{Ln}(T_c) = -6.8$ is larger than that obtained from external pressure $\Gamma = -2.5$ [9].

Concerning the electronic structure of the YFe_2D_x deuterides, the evolution of the XAS spectra at the Fe–K edge can be correlated with the previous results obtained by resonant photoemission [16]. The progressive decrease of the d like states at E_F with H content observed by XPS is in agreement with the progressive filling of the 3d states observed by XAS. The continuous decrease of the intensity of the peak at 7110 eV as a function of D content is an indication that the filling of the 3d band occurs continuously with D content.

Spin polarized band structure calculations performed on YFe_2 hydrides have shown that the shift of the Fermi level decreases the DOS of the spin up d band but increases that of the spin down d band, which results in an increase of the total Fe magnetization [23].

5. Conclusion

The evolution of the magnetic and electronic properties of YFe_2H_x deuterides has been studied by bulk magnetic measurements and by XAS at the Y and Fe K edge. The influence of hydrogen absorption on the magnetic Fe–Fe interactions has been studied. It was observed that the magnetic moment and the Curie temperature of the YFe_2D_x deuterides are not varying linearly with the cell volume whereas the 4p-3d band of Fe is progressively filled as the D content increases. In order to interpret these results it is necessary to take into account the crystallographic structure of the deuterides which show for the low deuterium content (1.2–1.9) a large Fe–Fe distance distribution including very large Fe–Fe distances.

Band structure calculations taking into account the true structure of the deuterides should be performed, specially for the deuterides with low D content showing tetragonal distortion and deuterium ordering.

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