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# Magnetic properties of the iron sublattice in the $YFe_{12-x}M_x$ compounds (M = Ti, Mo or V; $x = 1–3.5$)

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## Abstract
The magnetic properties of the $YFe_{12-x}M_x$ compounds (M = Ti, Mo or V; $x = 1–3.5$) have been determined in the ordered ferromagnetic state as well as in the paramagnetic state. The iron magnetic moment has been determined from 4 K up to the Curie temperature whereas the analysis of the paramagnetic region has led to the determination of the effective iron magnetic moment. The number of spins has been calculated below and above the Curie temperature in order to discuss the degree of itinerancy of the Fe magnetic behavior in the $YFe_{12-x}M_x$ compounds. All the $YFe_{12-x}M_x$ compounds (M = Ti, Mo or V; $x = 1–3.5$) have very similar crystalline properties: they crystallize in the same crystal structure and all the M elements used here are known to substitute for iron on the same crystal site. In contrast, they exhibit a wide range of magnetic behavior; the Curie temperature varies from 63 to 539 K and the mean magnetic moment per iron atom is also very dependent upon the M element used and its concentration. Furthermore the degree of itinerancy of the iron is not preserved along $YFe_{12-x}M_x$ compounds but is found to depend significantly upon the nature of the substituting element M and its concentration. The results are discussed and compared to earlier published results obtained on binary R–Fe and ternary R–Fe–B compounds.

## 1. Introduction

It has been known for many decades that combining rare-earth elements and Mn in a ratio 1:12 leads to intermetallic compounds of the $ThMn_{12}$ structure [1]. Corresponding Fe compounds do not form. Later investigations [2, 3] have shown that the formation of ternary compounds between rare-earths and Fe of crystal structure related to $ThMn_{12}$ is fairly general. The majority component of these compounds is Fe and their formula is described by $RFe_{12-x}M_x$. Nowadays, an extensive class of materials of the type $RFe_{12-x}M_x$ (where M = Ti, Cr, V, Mo, W, Si, ...) is known to retain the $ThMn_{12}$ structure [2–8]. The stability range of the $RFe_{12-x}M_x$ compounds is highly dependent upon the nature of the substituting element M [2, 4, 6].

These series of $RFe_{12-x}M_x$ intermetallics offer two main advantages as hard magnetic materials: (i) a high Fe content that favors a high magnetization, (ii) a relatively high Curie temperature, in particular for the Ti containing compounds. They have attracted much interest as potential candidate for high performance permanent magnet applications. This interest has increased over the last decade since it has been shown that the magnetic properties of these $RFe_{12-x}M_x$ compounds can be significantly improved by the insertion of light elements such as H, C or N within the crystal lattice [9–17].

Here we focus our attention on the $YFe_{12-x}M_x$ compounds in order to investigate the influence of the substitution on the magnetic properties of the iron sublattice in these phases. In particular, the magnetic behavior in the paramagnetic state will be analyzed and compared to the

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**Table 1.** Lattice parameters of the $YFe_{12-x}M_x$ compounds.

| Compound | $a$ (Å) | $c$ (Å) | $V$ (Å$^3$) |
|---|---|---|---|
| $YFe_{11}Ti$ | 8.503 (1) | 4.795 (1) | 346.7 |
| $YFe_{11}Mo$ | 8.519 (1) | 4.780 (1) | 346.9 |
| $YFe_{10}Mo_2$ | 8.553 (1) | 4.792 (1) | 350.6 |
| $YFe_{8.5}V_{3.5}$ | 8.508 (1) | 4.779 (1) | 345.9 |

behavior in the ordered region in order to determine the degree of localization of the Fe atoms. The results will be analyzed in the light of the self-consistent renormalization theory of spin fluctuations [18–20].

## 2. Experimental details

The alloys have been obtained by melting the high purity starting elements (better than 99.95%) in a high frequency induction furnace equipped with a water cooled copper crucible. The homogeneity of the ingot was subsequently achieved by annealing at 950 °C for ten days.

The homogeneity of the sample was checked by conventional x-ray powder diffraction with iron K$\alpha$ radiation and, in all cases, less than about 5 vol% of $\alpha$-Fe was found in each of the samples. The indexation of the Bragg peaks was performed with a tetragonal unit cell compatible with the $I4/mmm$ space group.

The magnetic ordering temperatures have been determined with a Faraday type balance at heating and cooling rates of 5 K min$^{-1}$. A sample of about 50–100 mg was sealed under vacuum in a small silica tube in order to prevent oxidization of the sample during heating. The magnetization curves of the $YFe_{12-x}M_x$ compounds were recorded at 4 K by the extraction method [21] in a continuous field of up to 9 T. The spontaneous magnetization values have been derived from an extrapolation to zero field of the magnetization obtained in fields higher than 4 T. A detailed description of the experimental set up can be found elsewhere [21]. The same cryostat was used with another insert [22] enabling any temperature between 300 and 850 K to be regulated. This set up has been used to investigate the magnetic properties of the $YFe_{12-x}M_x$ compounds above room temperature and in particular in the paramagnetic region.

## 3. Results and discussion

According to the x-ray investigation, the samples were all found to retain the $I4/mmm$ space group and the $ThMn_{12}$ type structure. The lattice parameters determined by a least square refinement of all the observed Bragg reflections are listed in table 1. These parameters are in agreement with those reported earlier for isotype compounds [2, 6, 11, 23]. It is clearly seen that the unit cell volume of the Mo containing compounds increases by about 5 Å$^3$ per Mo atom.

In these compounds, the rare-earth is known to be located on the 2a crystallographic site, whereas the iron atoms are distributed over the three inequivalent crystal sites: 8i, 8j and 8f. In the $RFe_{12-x}M_x$ compounds, the Ti, Mo and V atoms are all known to show a strong preference for

occupying the 8i site [11, 23–27]. We can consider that in the $YFe_{12-x}M_x$ compounds the 8i site is randomly occupied by the iron and the M atoms whereas the 8j and 8f sites are fully occupied by iron. According to earlier published neutron diffraction investigations, all the compounds studied here exhibit a uniaxial alignment of the Fe magnetic moment along the $c$ axis of the crystal structure [11, 23, 26, 27].

The spontaneous magnetization obtained for all the studied $YFe_{12-x}M_x$ compounds is summarized in table 2. As expected the largest magnetizations are obtained for the smallest M content. The largest magnetizations being 19.8 and 18.6 $\mu_B$/f.u. for $YFe_{11}Mo$ and $YFe_{11}Ti$ respectively. As can be seen from the $YFe_{12-x}Mo_x$ series of compounds, a large decrease of the magnetization is observed upon increasing the Mo content. A similar concentration dependence of the Fe magnetic moment has been reported for other M elements such as Ti, W, Cr, V [7, 8]. This magnetization decrease is in agreement with the strong decrease of the Curie temperature observed for these $YFe_{12-x}Mo_x$ compounds when the Mo concentration increases; see tables 2 and 3. Such magnetization decrease has been reported by Sun $et$ $al$ [28] for the $YFe_{12-x}Mo_x$ compounds. The spontaneous magnetizations obtained here are somewhat smaller than those reported in [23]. This most probably originates from the small magnetic fields used ($<1.6$ T), whereas in the present study a much better saturation can be obtained by using magnetic fields up to 9 T for the magnetization measurements. In this $RFe_{12-x}M_x$ structure the iron magnetic moments have been found to be very sensitive to the local atomic environment, the larger Fe moment being observed on the 8i site [29]. The saturation magnetization of the vanadium containing compounds is only about 10 $\mu_B$/f.u. which, assuming no magnetic moment on the M atoms, leads to a mean magnetic moment of 1.17 $\mu_B$ per Fe atom. This value is much lower than the 1.7 or 1.8 $\mu_B$/f.u. obtained for $YFe_{11}Ti$ and $YFe_{11}Mo$ respectively. This calculation probably underestimates the mean Fe magnetic moment value in $YFe_{8.5}V_{3.5}$. Indeed, it is well known that Fe–V hybridization leads to a significant negative polarization on the V atoms. This has been described extensively for the binary Fe–V system [30] but has also been reported from electronic structure calculations on $YFe_{10}V_2$ [31] where a negative polarization of about $-0.5$ $\mu_B$ per V has been obtained. More recently, this has been experimentally shown for the isotype $RFe_{10.5}V_{1.5}$ ternary compounds [26] where a negative polarization as high as 0.8 $\mu_B$ per V atoms has been obtained. The relatively low saturation magnetization of $YFe_{8.5}V_{3.5}$ compared to its large ordering temperature 410 K may indicate the contribution of vanadium atoms to the magnetization and the magnetic order. The 4, 300 and 380 K isothermal magnetization curves of $YFe_{8.5}V_{3.5}$ are shown in figure 1. The temperature dependences of the spontaneous magnetization for $YFe_{8.5}V_{3.5}$, $YFe_{11}Ti$ and $YFe_{11}Mo$ are shown in figure 2. It is noticeable that at 4 K, $YFe_{11}Ti$ has a smaller magnetization than $YFe_{11}Mo$, whereas the situation is reversed at room temperature. This larger magnetization at room temperature is clearly due to the much higher Curie temperature of $YFe_{11}Ti$. The 4 and 300 K saturation magnetizations are given in table 3 for all the studied

**Table 2.** Saturation magnetization $M_s$, Curie–Weiss constant $C$, paramagnetic effective moment $\mu_{eff}$, mean number of spins in the ordered $S_0$ or paramagnetic states $S_p$ and the corresponding ratio $r = S_p/S_0$ for the $YFe_{12-x}M_x$ compounds compared to that of the pure Fe.

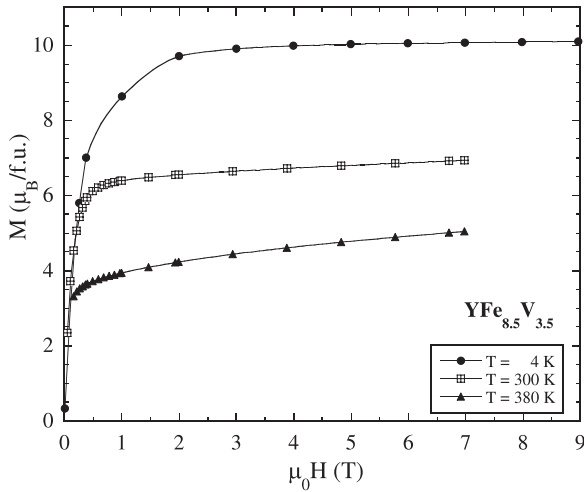| Compound | $M_s$ 300 K ($\mu_B$/f.u.) | $M_s$ 4 K ($\mu_B$/f.u.) | $M_{Fe}$ 4 K ($\mu_B$/atom) | $S_0$ | $\mu_{eff}$ ($\mu_B$/Fe atom) | $S_p$ | $r$ | $C$ ($\mu_B$ K/f.u. T) |
|---|---|---|---|---|---|---|---|---|
| $YFe_{11}Ti$ | 16.3 | 18.6 | 1.69 | 0.845 | 3.745 | 1.439 | 1.70 | 34.59 |
| $YFe_{11}Mo$ | 15.6 | 19.8 | 1.8 | 0.90 | 3.649 | 1.392 | 1.55 | 32.80 |
| $YFe_{10}Mo_2$ | 5.5 | 13.1 | 1.31 | 0.655 | 3.412 | 1.278 | 1.95 | 26.07 |
| $YFe_{8.5}V_{3.5}$ | 6.5 | 9.96 | 1.17 | 0.586 | 3.409 | 1.277 | 2.18 | 22.12 |
| $\alpha$-Fe | 2.17 | 2.22 | 2.22 | 1.11 | 3.18 | 1.17 | 1.05 | 2.26 |



**Figure 1.** The 4, 300 and 380 K isothermal magnetization temperature curves recorded for the $YFe_{8.5}V_{3.5}$ sample.
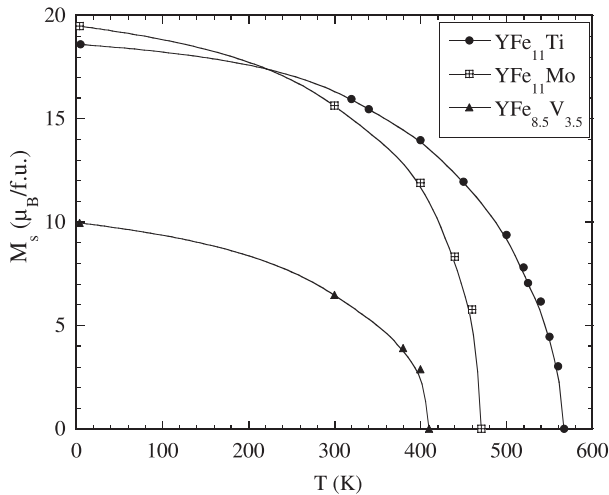


**Figure 2.** The temperature dependence of the spontaneous magnetization of $YFe_{11}Ti$, $YFe_{11}Mo$ and $YFe_{8.5}V_{3.5}$ compounds.



**Figure 3.** Arrott plots at the indicated temperature around the Curie temperature of $YFe_{11}Mo$.

**Table 3.** Curie temperature $T_C$, paramagnetic Curie temperature $\theta_p$, molecular field coefficient $n_{Fe-Fe}$ and exchange field $B_{ex}$ in the $YFe_{12-x}M_x$ compounds.

| Compound | Number of Fe atom/f.u. | $T_C$ (K) | $\theta_p$ (K) | $n_{Fe-Fe}$ ($\mu_0$) | $B_{ex}$ (T) |
|---|---|---|---|---|---|
| $YFe_{11}Ti$ | 11 | 563 | 565 | 232 | 290 |
| $YFe_{11}Mo$ | 11 | 469 | 500 | 213 | 283 |
| $YFe_{10}Mo_2$ | 10 | 340 | 384 | 275 | 171 |
| $YFe_{8.5}V_{3.5}$ | 8.5 | 410 | 372 | 275 | 185 |
| $\alpha$-Fe | 1 | 1043 | 1093 | 466 | 1023 |

compounds. It is noteworthy that the values obtained here are in good agreement with those reported earlier [6, 23].

The Curie temperatures have been determined by two different methods: (i) from the thermomagnetic analysis by plotting the temperature dependence of the square of the magnetization; (ii) from the Arrott plots [32] figure 3. The two methods are found to give results that are in excellent agreement. The Curie temperatures $T_C$ of the $YFe_{12-x}M_x$

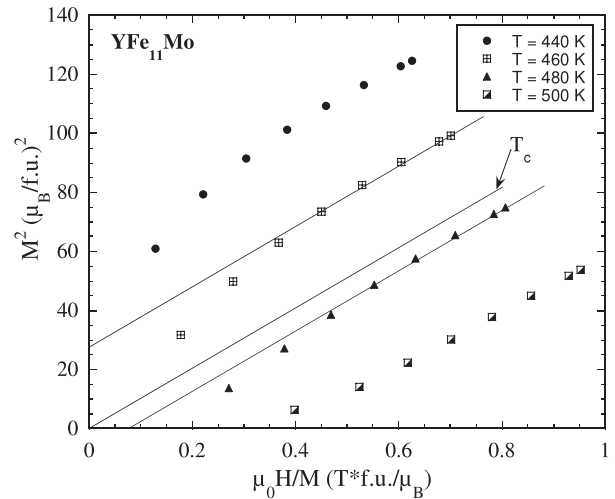compounds studied here are summarized in table 3. It is noteworthy that the Curie temperatures obtained here are in good agreement with those reported earlier in [2, 4, 33]. The Curie temperature value of 471 ± 5 K obtained by thermomagnetic analysis for $YFe_{11}Mo$ agrees with the value of 475 ± 4 K obtained from the Arrott plots given in figure 3. The $T_C$ values of the $YFe_{12-x}Mo_x$ compounds are in excellent agreement with those reported by Sun *et al* [28]. The molecular field coefficient $n_{Fe-Fe}$ and exchange field $B_{ex}$ have been derived from the Curie temperature of the $YFe_{12-x}M_x$ compounds assuming that the $T_C$ results only from Fe–Fe exchange interactions. The equations used are given below where $M$ and $C$ correspond to the magnetization and the Curie Weiss constant respectively. The corresponding values of the molecular field coefficient $n_{Fe-Fe}$ and exchange field $B_{ex}$ are

given in table 3.

$$B_{ex} = \frac{MT_C}{C} \qquad (1)$$

$$n_{Fe-Fe} = \frac{T_C}{C}. \qquad (2)$$

The composition dependence of the exchange field in the $YFe_{12-x}Mo_x$ compounds exhibits a dramatic decrease versus the Mo content. It is clear that the Mo for Fe substitution leads to a dramatic decrease of the exchange field. For the same composition, the Ti containing compound exhibit a similar but slightly higher exchange interaction than the Mo isotype compound. The $n_{Fe-Fe}$ exchange interactions determined here are of the same magnitude as those reported recently in $ThFe_{11}C_x$ [34] but much larger than what is usually found for the $R_2Fe_{17}$ compounds [35].

Magnetic measurements have been undertaken at high temperature for all the studied compounds in order to investigate the magnetic behavior of Fe in the paramagnetic state. Since the iron impurity orders at a much higher temperature than the $YFe_{12-x}M_x$ compounds its magnetic influence most likely will affect the determination of the intrinsic susceptibility of the $YFe_{12-x}M_x$ compounds. In order to avoid such an influence of the magnetic ordered iron impurities on the susceptibility values $\chi$, the latter were obtained from field-dependent measurements according to Honda–Owen plots [36]:

$$\frac{M}{H} = \chi + \frac{cM_s}{H} \qquad (3)$$

by extrapolating to $H^{-1} \to 0$. The impurity content is represented by $c$ whereas $M_s$ corresponds to the saturation magnetization of the impurity phase.

The thermal variation of the reciprocal susceptibility has been measured for all $YFe_{12-x}M_x$ compounds. At high temperature, the $YFe_{12-x}M_x$ compounds exhibit a Curie–Weiss behavior. The thermal evolution of the reciprocal susceptibilities is plotted in figure 4 for all the studied compounds. A linear fit of the data according to a Curie–Weiss law

$$\chi = \frac{C}{T - \theta_p} \qquad (4)$$

leads to the values reported in tables 2 and 3.

The determined paramagnetic temperatures $\theta_p$ are slightly larger than the Curie temperatures deduced from the thermomagnetic measurements, which is reminiscent of short-range magnetic interactions above $T_C$. As can be seen from table 3, $\theta_p$ is about 30–40 K larger than $T_C$ for the $YFe_{11}Mo$, $YFe_{11}Ti$, $YFe_{11}Mo_2$ compounds. The case of $YFe_{8.5}V_{3.5}$ is peculiar since the $\theta_p$ value is slightly lower than $T_C$. This may be indicative of the presence of antiferromagnetic interactions, thus confirming the negative polarization on the vanadium as deduced above from the magnetization analysis.

The magnetic behavior of the $YFe_{12-x}M_x$ compounds may be analyzed using the self-consistent renormalization theory of spin fluctuations [18–20]. In this model two extreme cases are described: (i) local moment and (ii) exchange enhanced paramagnetism. These two extreme cases are
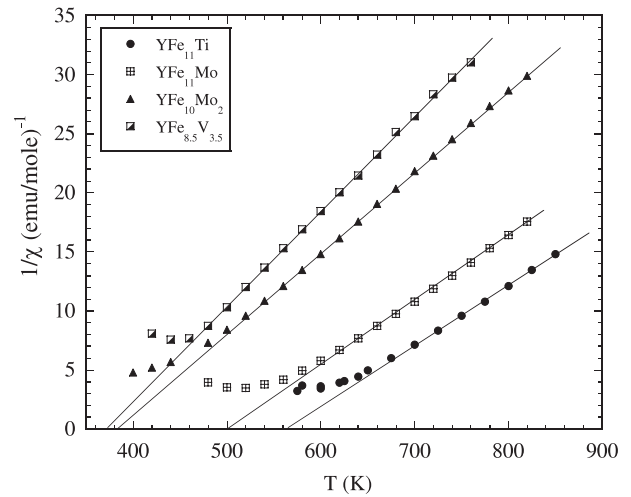


**Figure 4.** The thermal evolution of the reciprocal magnetic susceptibilities for $YFe_{11}Mo$, $YFe_{10}M_2$, $YFe_{11}Ti$ and $YFe_{8.5}V_{3.5}$ compounds.

characterized by different values of the ratio $r = S_p/S_0$ between the number of spins determined from Curie constants ($S_p$) and the saturation magnetization ($S_0$). For a local moment we have $r = 1$ while for longitudinal spin fluctuations $r = \infty$. The ratio $r$ has been calculated for all the $YFe_{12-x}M_x$ compounds and is reported in table 2. A significant increase of the ratio $r$ is observed upon increasing the M content. This bears witness to a progressive delocalization of the Fe magnetic moment [37]. According to the renormalization theory of spin fluctuations, when the amplitude of the local spin fluctuation is large and fixed there is the local moment limit where only the transverse components of local spin fluctuations are important. On the contrary, for a nearly ferromagnetic alloy, the longitudinal spin fluctuations dominate. The substitution induced increase of the $r$ ratio observed for the $YFe_{12-x}M_x$ compounds suggests that the contribution of the longitudinal spin fluctuations increases as the M content increases. The $r$ values obtained for the $YFe_{12-x}M_x$ compounds—see table 2 —reveal that the iron magnetic moment has a similar degree of localization in $YFe_{12-x}M_x$ as in $Th_2Fe_{17}C_x$ compounds [35] and is slightly less localized than in the $ThFe_{11}C_x$ phases [36]. It is worth mentioning that none of these phases are as localized as elemental $\alpha$-Fe ($r = 1.05$).

In the $RFe_2$ compounds the effective magnetic moment per Fe atom is found to be about 3.0–3.4 $\mu_B$/Fe atom in $YFe_2$ and $GdFe_2$ respectively. For these compounds the ratio $r$ between the number of spins obtained from the Curie constants and those determined from saturation magnetization measurements has been reported to be constant in the $Gd_xY_{1-x}Fe_2$ system with a value of 1.5 [37]. This value is very similar to that obtained for $YFe_{11}Mo$, showing that the Fe sublattice is in the same degree of localization in both compounds. Burzo [38] reported a $r$ value of 1.30 and 1.24 for the $Gd_xY_{1-x}Fe_3$ and $R_2Fe_{14}B$ compounds respectively which are known to have exchange interactions similar to the one obtained for the $YFe_{12-x}M_x$ compounds.

It has been shown that for many families of compounds, the $r$ ratio varies linearly versus the exchange fields. Such a

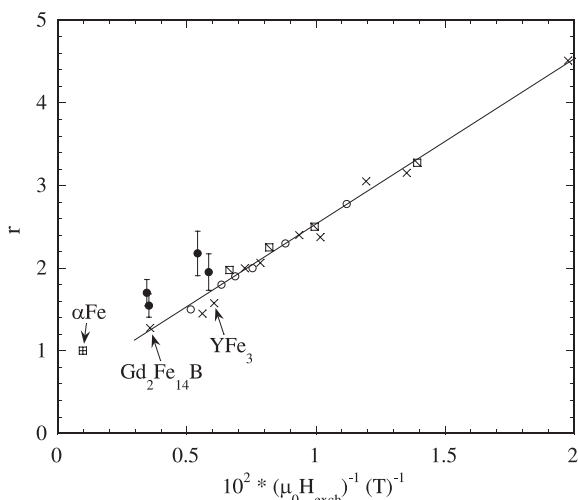**Figure 5.** The evolution of the $r = S_p/S_0$ values as a function of the exchange fields. The $YFe_{12-x}M_x$ compounds are drawn in black circles, the others points are taken from [37].

plot is given in figure 5, taking into account the $YFe_{12-x}M_x$ compounds studied here. Keeping in mind that the contribution of the M atoms to the exchange interactions has been neglected, the agreement is reasonable [38]. This confirms that the major contribution to the magnetic properties arises from the Fe atoms.

Recent investigation of the ternary Th–Fe–C interstitial compounds has shown a pronounced reduction of the $r$ value upon carbon insertion in the lattice [34, 35]. This change of the ratio between the ordered and the effective spin numbers ($r = S_p/S_0$) is slightly modified upon interstitial element insertion showing that the spin fluctuations change with the interstitial concentration. This reduction of the $r$ ratio observed for the $ThFe_{11}C_x$ and $Th_2Fe_{17}C_x$ series of compounds, shows that the carbon addition induces an evolution towards a local Fe magnetic moment.

Similar behavior can be expected for the $YFe_{12-x}M_xX_y$ (X = H, C, N) compounds whose Curie temperature is known to increase significantly upon interstitial element insertion [6, 11, 25, 28]. According to the Rhodes and Wolfarth curve [39], one can expect that the magnetic properties of the $YFe_{12-x}M_xX_y$ (X = H, C, N) compounds evolved towards a more localized-like magnetism. Such an interpretation can be proposed to account for the magnetic properties of the carbides and nitrides. Further study of the $YFe_{12-x}M_xX_y$ compounds would be needed to confirm this expectation and to quantify this evolution of the $r$ ratio. This is rather difficult to confirm experimentally since the $YFe_{12-x}M_xX_y$ compounds are metastable. They can partially decompose upon heating while performing the magnetic investigation in the high temperature paramagnetic state. This is particularly true for the nitrides which form rare-earth nitride and $Fe_4N$ [24] or in the case of the hydrides for which hydrogen is released upon heating [29], thus hampering any analysis of the magnetic properties over a wide temperature range in the paramagnetic state.

## 4. Conclusion

The magnetic properties of the $YFe_{12-x}M_x$ compounds are found to be dramatically dependent upon the nature and concentration of the element M. The iron magnetic moment as well as the Curie temperature and the exchange interactions are significantly reduced upon M for Fe substitution (M = Ti, Mo, V). The present study has shown that the spin fluctuations can vary from a rather localized to a strongly delocalized type for $YFe_{12-x}M_x$ compounds. In particular, the degree of itinerancy is dramatically increased upon increasing the Mo concentration. The ratio $r$ between the number of spins obtained from the Curie constants and that determined from saturation magnetization is found to vary from 1.5 to 2.2 for $YFe_{11}Mo$ and $YFe_{8.5}V_{3.5}$ respectively. This shows a much more delocalized magnetism than in iron even more than in the binary $YFe_2$ compounds. The $YFe_{12-x}M_x$ compounds are thus offering a unique opportunity to cover a wide range of magnetic behavior over the Rhodes and Wolfarth curves for compounds keeping the same crystal structure.

## Acknowledgments

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