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Olivier Isnard, V. Pop. Magnetic properties of the iron sublattice in the YFe_{12-x}M_x compounds (M = Ti, Mo or V; x = 1-3.5). *Journal of Physics: Condensed Matter*, 2009, 21, pp.6003. 10.1088/0953-8984/21/40/406003 . hal-00978117

HAL Id: hal-00978117

<https://hal.science/hal-00978117>

Submitted on 26 Apr 2014

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Magnetic properties of the iron sub-lattice in the $\text{YFe}_{12-x}\text{M}_x$ compounds ($M=\text{Ti, Mo or V}$; $x=1$ to 3.5)

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Abstract

The magnetic properties of the $\text{YFe}_{12-x}\text{M}_x$ compounds ($M=\text{Ti, Mo or V}$; $x=1$ to 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 4K 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 $\text{YFe}_{12-x}\text{M}_x$ compounds. All the $\text{YFe}_{12-x}\text{M}_x$ compounds ($M=\text{Ti, Mo or V}$; $x=1$ to 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. On the contrary, they exhibit a wide range of magnetic behavior: the Curie temperature varies from 63 to 539K 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 $\text{YFe}_{12-x}\text{M}_x$ 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 $R\text{-Fe}$ and ternary $R\text{-Fe-B}$ compounds.

PACS : 75.30.-m, 75.30.Cr, 75.30.Et

Keywords : Spin fluctuation, Paramagnetic susceptibility, Exchange interaction

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1. Introduction

It has been shown already for many decades that combining of 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 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 $R\text{Fe}_{12-x}M_x$. Nowadays, an extensive class of materials of the type $R\text{Fe}_{12-x}M_x$ (where $M=\text{Ti, Cr, V, Mo, W, Si...}$) is known to retain the ThMn_{12} structure [2-8]. The stability range of the $R\text{Fe}_{12-x}M_x$ compounds is highly dependent upon the nature of the substituting element M [2,4,6].

These series of $R\text{Fe}_{12-x}M_x$ intermetallics offer two main advantages as hard magnetic material: (i) a high Fe content that favor 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 been increased in the last decade since it has been shown that the magnetic properties of these $R\text{Fe}_{12-x}M_x$ compounds can significantly been improved by insertion of light elements such as H, C or N within the crystal lattice [9-17].

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

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 10 days.

The homogeneity of the sample was checked by conventional X-ray powder diffraction with iron K_α radiation and, in all cases, less than ca. 5 volume percent of α -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 per minute. A sample of c.a. 50 to 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 $\text{YFe}_{12-x}M_x$ compounds were recorded at 5 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 $\text{YFe}_{12-x}\text{M}_x$ 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 that reported earlier for isotype compounds [2,6,11,23]. It is clearly seen that the unit cell volume of the Mo containing compounds increases of about 5 \AA^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 $\text{RFe}_{12-x}\text{M}_x$ 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 $\text{YFe}_{12-x}\text{M}_x$ 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 investigation, all the compounds studied here are exhibiting 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 $\text{YFe}_{12-x}\text{M}_x$ compounds are summarized in table 2. As expected the largest magnetization are obtained for the smallest M content. The largest magnetization being 19.8 and 18.6 μ_B /formula unit for YFe_{11}Mo and YFe_{11}Ti respectively. As can be seen from the $\text{YFe}_{12-x}\text{Mo}_x$ series of compounds a large decrease of the magnetization is observed upon increasing the Mo content. 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 $\text{YFe}_{12-x}\text{Mo}_x$ compounds when the Mo concentration increases –table 2-. Such magnetization decrease has been reported by Sun et al. [28] for the $\text{YFe}_{12-x}\text{Mo}_x$ compounds. The spontaneous magnetization obtained here are somewhat smaller than that reported in reference [23]. This most probably originates from the small magnetic fields used ($<1.6\text{T}$), whereas in the present study a much better saturation can be obtained by using a magnetic fields up to 9 T for the magnetization measurements. In this $\text{RFe}_{12-x}\text{M}_x$ 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 μ_B /formula unit which, assuming no magnetic moment on the M atoms, leads to a mean magnetic moment of 1.17 μ_B per Fe atom. This value is much lower than the 1.7 or 1.8 μ_B per formula unit obtained for YFe_{11}Ti and YFe_{11}Mo respectively. This calculation probably underestimates the mean Fe magnetic moment value in $\text{YFe}_{8.5}\text{V}_{3.5}$. 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 also reported from electronic structure calculation on $\text{YFe}_{10}\text{V}_2$ [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 $R\text{Fe}_{10.5}\text{V}_{1.5}$ 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 $\text{YFe}_{8.5}\text{V}_{3.5}$ compared to its large ordering temperature 410K may indicate the contribution of vanadium atoms to the magnetization and the magnetic order. The 4, 300 and 380K isothermal magnetization curves of $\text{YFe}_{8.5}\text{V}_{3.5}$ are shown in Fig. 1. The temperature dependence of the spontaneous magnetization for $\text{YFe}_{8.5}\text{V}_{3.5}$, YFe_{11}Ti and YFe_{11}Mo are shown in Fig. 2. It is noticeable that at 4K, 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 4K and 300K saturation magnetization are given in table 3 for all the studied compounds. It is noteworthy that the values obtained here are in good agreement with that 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 temperature T_c of the $\text{YFe}_{12-x}\text{M}_x$ compounds studied here are summarized in table 3. It is noteworthy that the Curie temperatures obtained here are in good agreement with that earlier reported in [2,4,33]. The Curie temperature value of $471 \pm 5\text{K}$ obtained by thermomagnetic analysis for YFe_{11}Mo agrees with the value of $475 \pm 4\text{K}$ obtained from the Arrott plots given in Fig. 3. The T_c values of the $\text{YFe}_{12-x}\text{Mo}_x$ compounds are in excellent agreement with that reported by Sun et al [28]. The molecular field coefficient $n_{\text{Fe-Fe}}$ and exchange field B_{ex} have been derived from the Curie temperature of the $\text{YFe}_{12-x}\text{M}_x$ 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 $n_{\text{Fe-Fe}}$ and exchange field B_{ex} are given in table 3.

$$B_{ex} = \frac{M T_c}{C} \quad (1)$$

$$n_{\text{Fe-Fe}} = \frac{T_c}{C} \quad (2)$$

The composition dependence of the exchange field in the $\text{YFe}_{12-x}\text{Mo}_x$ 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 interactions than the Mo isotype compound. The $n_{\text{Fe-Fe}}$ exchange interactions determined here are of the same magnitude than that reported recently in $\text{ThFe}_{11}\text{C}_x$ [34] but much larger than what is usually found for the $R_2\text{Fe}_{17}$ 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 $\text{YFe}_{12-x}\text{M}_x$ compounds its magnetic influence most likely will affect the determination of the intrinsic susceptibility of the $\text{YFe}_{12-x}\text{M}_x$ compounds. In order to avoid such an influence of the magnetic ordered iron impurities on the susceptibility values χ , the latter were obtained from field-dependent measurements according to Honda-Owen plots [36]:

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

by extrapolating to $H^{-1} \rightarrow 0$. 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 $\text{YFe}_{12-x}\text{M}_x$ compounds. At high temperature, the $\text{YFe}_{12-x}\text{M}_x$ compounds are exhibiting a Curie-Weiss behavior. The thermal evolution of the reciprocal susceptibilities is plotted in Fig. 4 for all the studied compounds. A linear fit of the data according to a Curie-Weiss law,

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

leads to the values reported in table 2 and 3.

The determined paramagnetic temperature θ_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, θ_p is about 30 to 40 K larger than T_C for the YFe_{11}Mo , YFe_{11}Ti , $\text{YFe}_{11}\text{Mo}_2$ compounds. The case of $\text{YFe}_{8.5}\text{V}_{3.5}$ is peculiar since θ_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 $\text{YFe}_{12-x}\text{M}_x$ compounds may be analyzed using the self-consistent renormalization theory of spin fluctuations [18-20]. In this model two extremes cases are described : i) local moment and ii) exchange enhanced paramagnetism. These two extreme cases are characterized by different value 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 $\text{YFe}_{12-x}\text{M}_x$ 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 renormalisation 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 component 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 $\text{YFe}_{12-x}\text{M}_x$ 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- reveals that the iron magnetic moment is in the similar degree of localization in $YFe_{12-x}M_x$ than in $Th_2Fe_{17}C_x$ compounds [35] and slightly less localized than in the $ThFe_{11}C_x$ phases [36]. It is worth to mention that none of these phases are as localized than elemental α -Fe ($r = 1.05$).

In the RFe_2 compounds the effective magnetic moment per Fe atom is found to be about 3.0 to 3.4 μ_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 that 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 interaction 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 plot is given in Fig. 5 evolution 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 have been neglected the agreement is reasonable [38]. This confirms that the major contribution to the magnetic properties arise from the Fe atoms.

Recent investigation of the ternary Th-Fe-C interstitial compounds has shown that 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 interpretation can be proposed to account for the magnetic properties of the carbides and nitrides. Further study on 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 to analyze the magnetic properties in a wide temperature range in the paramagnetic state.

4. Conclusion

The magnetic properties of the $\text{YFe}_{12-x}\text{M}_x$ compounds are found to dramatically dependent upon the M element nature and concentration. The iron magnetic moment as well as the Curie temperature and the exchange interactions are significantly reduced upon M for Fe substitution ($M=\text{Ti}, \text{Mo}, \text{V}$). The present study has shown that the spin fluctuations can vary from a rather localized to a strongly delocalized type for $\text{YFe}_{12-x}\text{M}_x$ 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 $\text{YFe}_{8.5}\text{V}_{3.5}$ respectively. This shows a much more delocalized magnetism than in iron even more than in the binary YFe_2 compounds. The $\text{YFe}_{12-x}\text{M}_x$ compounds are thus offering the unique opportunity to cover a wide range of magnetic behavior over the Rhodes and Wolfarth curves for compounds keeping the same crystal structure.

ACKNOWLEDGMENTS

V. P. thanks the ‘University Joseph Fourier, Grenoble, France’ for the grant of ‘Maître de conférence invité’ at the Faculty of physics. O. I. would like to thank E. Burzo for interesting discussions.

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Figure caption

Figure 1. The 4, 300 and 380 K isothermal magnetization curves temperature recorded for the $\text{YFe}_{8.5}\text{V}_{3.5}$ sample.

Figure 2. The temperature dependence of the spontaneous magnetization of YFe_{11}Ti , YFe_{11}Mo and $\text{YFe}_{8.5}\text{V}_{3.5}$ compounds.

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

Figure 4. The thermal evolution of the reciprocal magnetic susceptibilities for YFe_{11}Mo , $\text{YFe}_{10}\text{M}_2$, YFe_{11}Ti and $\text{YFe}_{8.5}\text{V}_{3.5}$ compounds.

Figure 5. The evolution of the $r = \mathcal{S}_p / \mathcal{S}_0$ values as function of the exchange fields. The $\text{YFe}_{12-x}\text{M}_x$ compounds are drawn in black circles, the others points are taken from reference [37].

Table 1. Lattice parameters of the $\text{YFe}_{12-x}\text{M}_x$ 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
$\text{YFe}_{10}\text{Mo}_2$	8.553 (1)	4.792 (1)	350.6
$\text{YFe}_{8.5}\text{V}_{3.5}$	8.508 (1)	4.779 (1)	345.9

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

Compound	M_s 300K ($\mu_B/\text{f.u.}$)	M_s 4K ($\mu_B/\text{f.u.}$)	M_{Fe} 4K (μ_B/atom)	S_0	μ_{eff} ($\mu_B/\text{Fe atom}$)	S_p	r	C ($\mu_B^*\text{K}/\text{f.u.}*T$)
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
$\text{YFe}_{10}\text{Mo}_2$	5.5	13.1	1.31	0.655	3.412	1.278	1.95	26.07
$\text{YFe}_{8.5}\text{V}_{3.5}$	6.5	9.96	1.17	0.586	3.409	1.277	2.18	22.12
Fe α	2.17	2.22	2.22	1.11	3.18	1.17	1.05	2.26

Table 3. Curie temperature T_c , paramagnetic Curie temperature θ_p , molecular field coefficient $n_{\text{Fe-Fe}}$ and exchange field B_{ex} in the $\text{YFe}_{12-x}\text{M}_x$ compounds

Compound	Number of Fe atom/f.u.	T_c (K)	θ_p (K)	$n_{\text{Fe-Fe}}$ (μ_0)	B_{ex} (T)
YFe_{11}Ti	11	539	565	232	290
YFe_{11}Mo	11	469	500	213	283
$\text{YFe}_{10}\text{Mo}_2$	10	340	384	275	171
$\text{YFe}_{8.5}\text{V}_{3.5}$	8.5	410	372	275	185
Fe α	1	1043	1093	466	1023

Figure 1

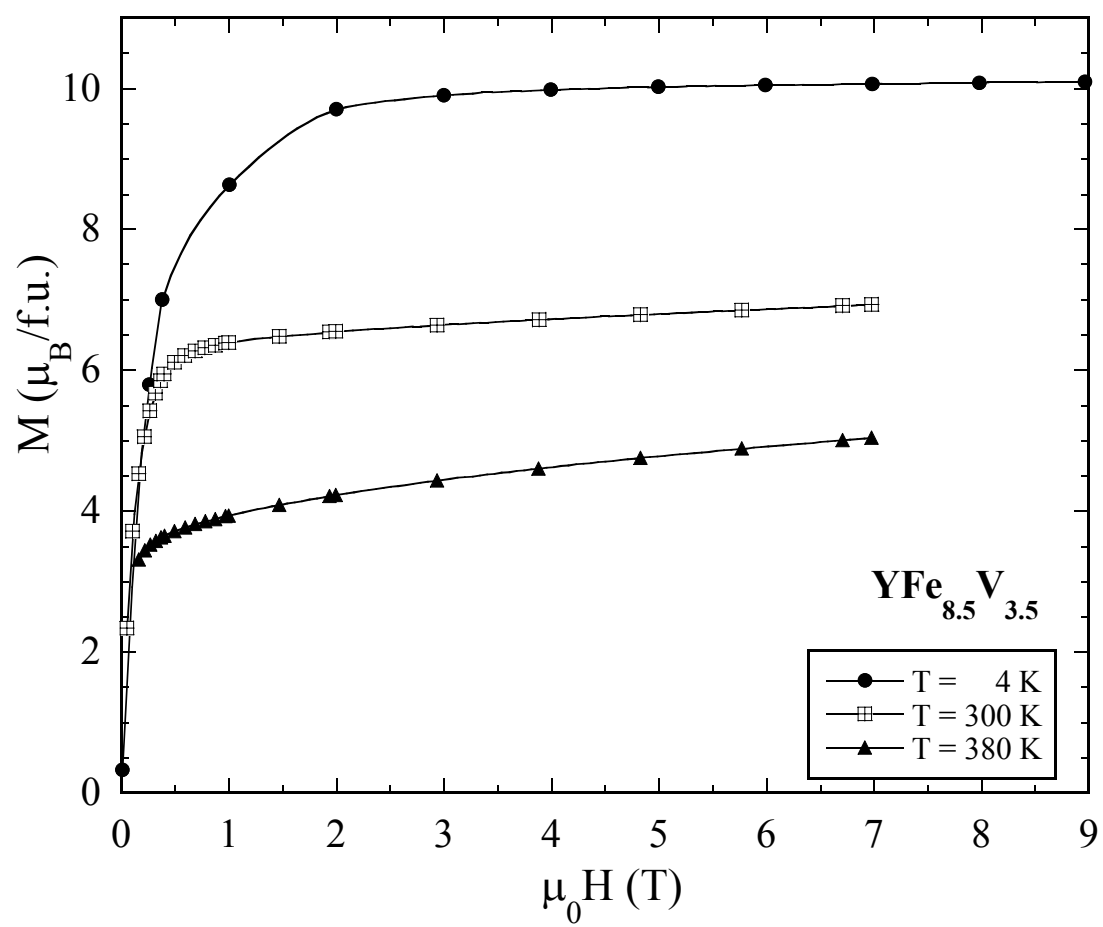


Figure 2

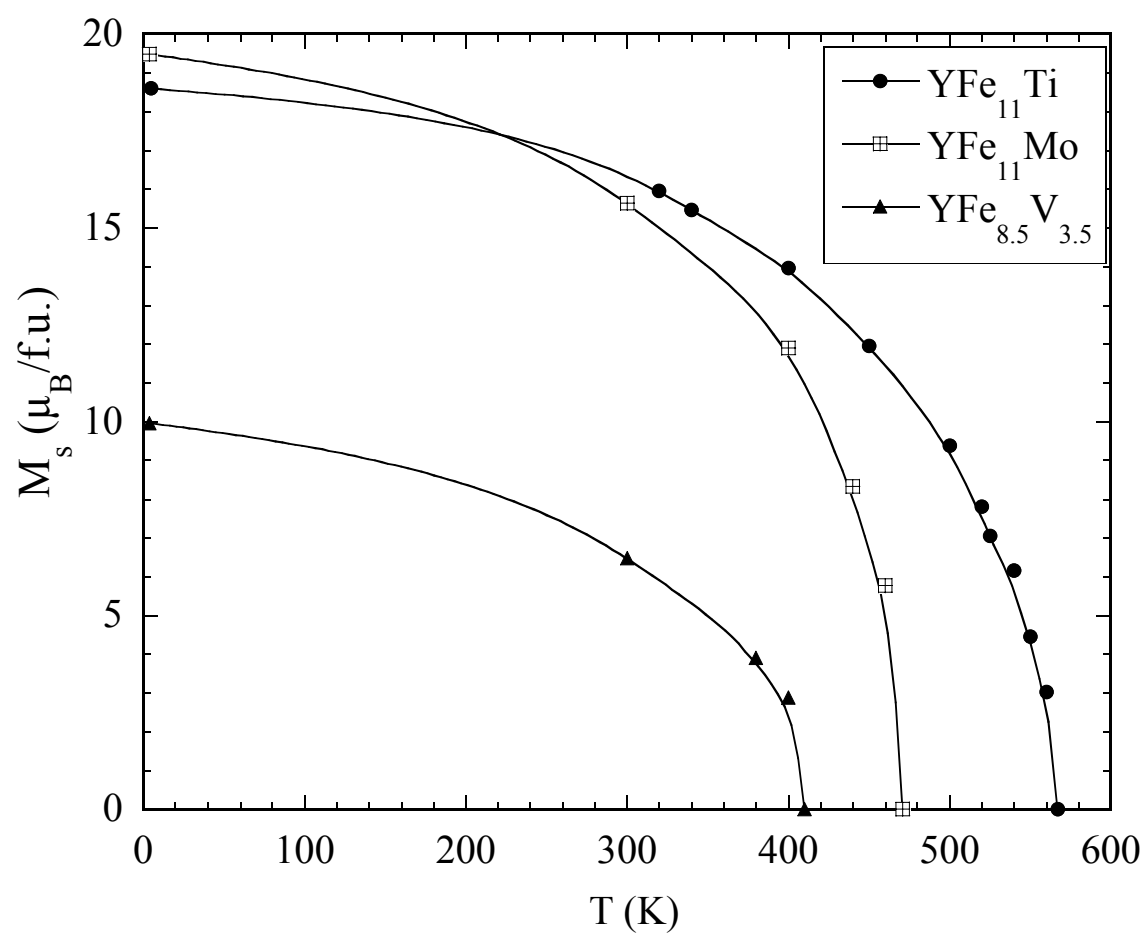


Figure 3

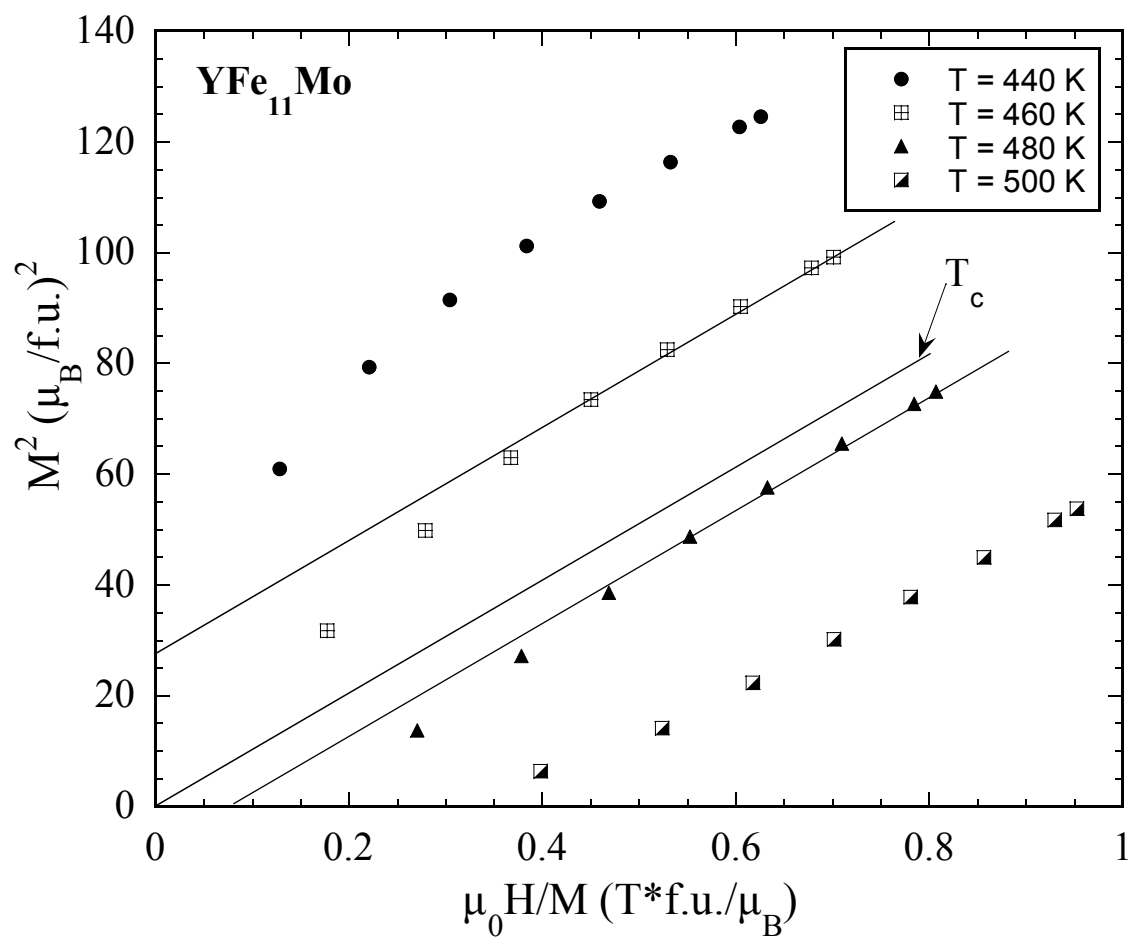


Figure 4

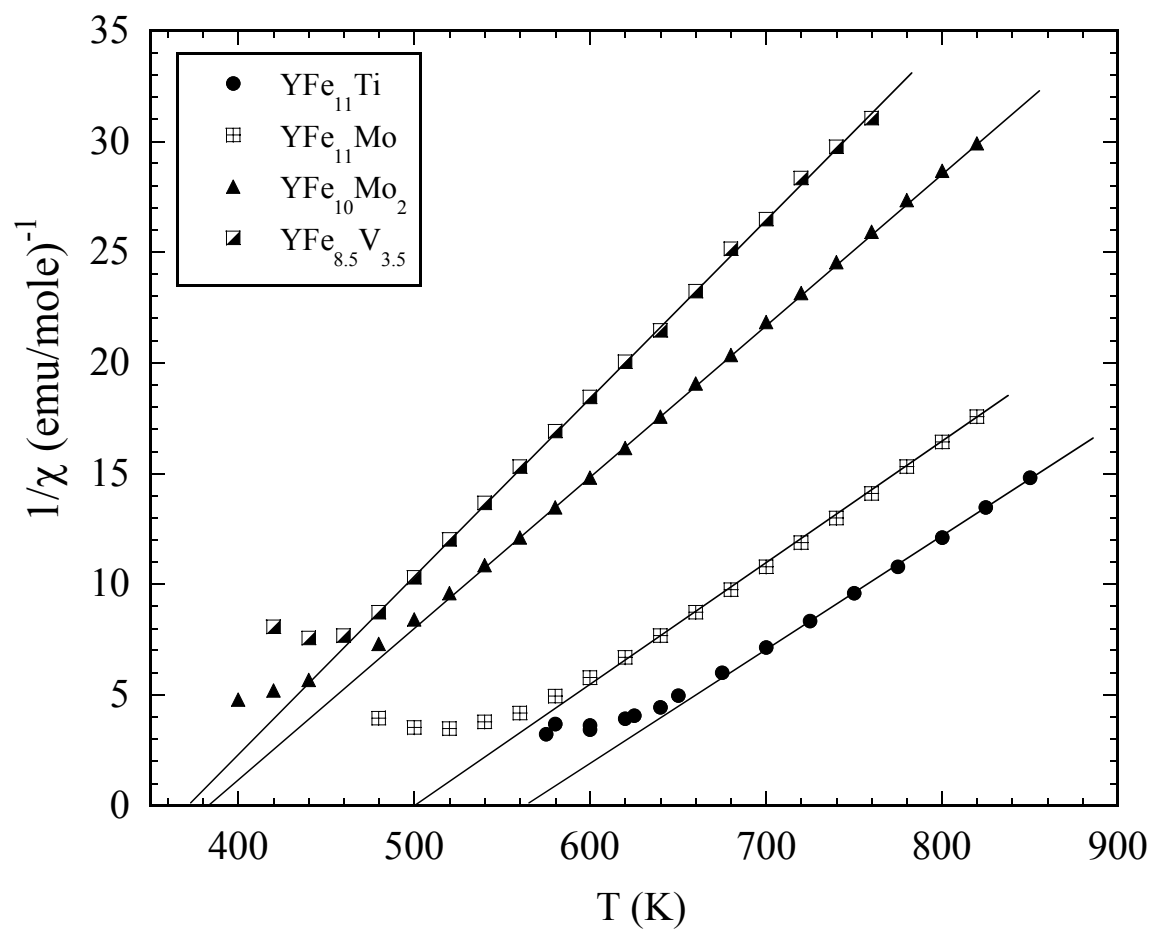


Figure 5

