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Magnetic properties of the new compounds $RFe_{11.5}Ta_{0.5}$ ($R \equiv$ Tb, Dy, Ho, Er and Lu)

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Abstract. A new series of compounds with the $ThMn_{12}$-type structure $RFe_{11.5}Ta_{0.5}$ ($R \equiv$ Tb, Dy, Ho, Er and Lu) has been synthesized and studied by x-ray diffraction, ac susceptibility and magnetization versus temperature and field measurements. The maximum Curie temperature is for the Tb compound ($T_C = 576$ K). Spin reorientation transitions have been observed for the compounds $DyFe_{11.5}Ta_{0.5}$ and $ErFe_{11.5}Ta_{0.5}$. A first order magnetization process has been observed at low temperature in $DyFe_{11.5}Ta_{0.5}$ and $HoFe_{11.5}Ta_{0.5}$. The importance of the metallic radius of M in the magnetic properties of $RFe_{12-x}M_x$ (for the same rate of M substitution) is discussed.

1. Introduction

Since the discovery of $Nd_2Fe_{14}B$, more than 200 binary and ternary iron rich intermetallic compounds have been synthesized looking for better permanent magnet performances or to deepen in the understanding of their magnetic properties. The rare earth (R) iron rich intermetallic compounds with $ThMn_{12}$ structure (space group $I4/mmm$) are specially interesting in this respect since, in addition to having relatively high Curie temperatures and magnetization, they crystallize in a system of high symmetry, the R atom is located in just one site (2a) and the number of atoms in the unit cell ($Z = 2$) is considerably lower than in the $R_2Fe_{14}B$ or R_2Fe_{17} compounds [1, 2].

Irrespective of the rare earth involved, the binary RFe_{12} compounds are not stable. However, the introduction of a third element $M \equiv$ Al, Si, Ti, V, Cr, Nb, Mo, W or Re stabilizes the pseudo-binaries $RFe_{12-x}M_x$ with the $ThMn_{12}$ structure. Though the number of elements capable of substituting for iron in the lattice is very large, the range of compositional variation is element dependent. It may vary from as wide as $0.5 \leqslant x \leqslant 4$ for $M \equiv$ Mo [3] to as narrow as $0.65 \leqslant x \leqslant 0.8$ for $M \equiv$ Nb [4]. The inclusion of a non-magnetic element M has a detrimental influence in the magnetic properties of the $RFe_{12-x}M_x$ compounds. It is known that the Curie temperature and the saturation magnetization M_s decrease as the x concentration is increased [1]. Therefore, it is important to synthesize new compounds with the minimum stabilizing element concentration.

The similarity of the chemical properties of elements fulfilling the diagonal position relationship in the periodic table has been invoked to explain the success in stabilizing the Nb series [4]. On the other hand, the similarity in chemical properties of isoelectronic elements seems to be effective for the VIa group since the Cr, Mo and W substitutions have

stabilized the structure. However, it remains to be proved that all the Va group elements also stabilize it. Until now the V and Nb substitutions existed, but it remains to verify whether the Ta element would also stabilize the pseudo-binary compounds.

In this paper we report on the magnetic properties of the new series $RFe_{12-x}Ta_x$ ($R \equiv Tb$, Dy, Ho, Er and Lu) compounds in which the minimal amount of stabilizing agent, $x = 0.5$, is needed.

2. Experimental details

Only the heavy rare earth $RFe_{11.5}M_{0.5}$ ($R \equiv Tb$, Dy, Ho, Er and Lu) alloys could be synthesized as homogeneous compounds, while the attempts to prepare compounds with either yttrium or rare earths lighter than terbium were not successful. The synthesis was done in a high-frequency induction furnace, using the cold crucible method, where the stoichiometric amounts of the constituent elements were melted. The samples were studied in the as-cast state. The crystallinity of the samples and their homogeneity was assessed from x-ray diffraction (XRD) on powdered samples. XRD analysis was performed at room temperature by using a Rigaku diffractometer and Cu K_α radiation. All samples investigated were found to be nearly single phase, with minor amounts of free α-iron and $TaFe_2$. The unit cell parameters and volume are given in table 1. We can observe that the lattice parameter a decreases with increasing atomic number of the R atom owing to the lanthanide contraction, whereas c remains almost constant. Essentially the reason is that the a dimension is related to the R–T chains in the 1:12 structure and is sensitive to the R atomic size, while the c dimension is related just to the Fe sublattice and it is only slightly modified by the R substitution [5]. We have determined that the Ta atom enters only at the Bi site.

Table 1. Structural parameters and magnetic properties of the $RFe_{11.5}Ta_{0.5}$ compounds.

Compound	a (Å)	c (Å)	V (Å^3)	T_C (K) ± 2 K	T_{SR} (K) ± 5 K	EMD (300 K)	M_s (μ_B fu^{-1}) 5 K	300 K	H_a (T) (300 K)
$TbFe_{11.5}Ta_{0.5}$	8.5051(3)	4.7787(3)	345.67(3)	576	—	$\perp c$	12.4	13.2	3
$DyFe_{11.5}Ta_{0.5}$	8.4963(3)	4.7780(3)	344.91(3)	550	185, 265	$\parallel c$	11.7	14.2	2
$HoFe_{11.5}Ta_{0.5}$	8.4828(3)	4.7739(2)	343.52(3)	541	—	$\parallel c$	12.3	14.5	3.5
$ErFe_{11.5}Ta_{0.5}$	8.4777(1)	4.7744(1)	343.14(1)	532	40	$\parallel c$	12.5	15.2	3.5
$LuFe_{11.5}Ta_{0.5}$	8.4721(4)	4.7784(3)	342.97(4)	499	—	$\parallel c$	20.9	18.3	3

Oriented samples for magnetic measurements and the study of the magnetocrystalline anisotropy were prepared by mixing powders (sieved to give a particle size lower than 20 μm) with epoxy resin, and then curing the resin at room temperature under an applied field of about 0.5 T.

The easy magnetization direction (EMD) at room temperature was deduced from the XRD patterns recorded on field-aligned samples. In the x-ray patterns of the samples with $R \equiv Dy$, Ho, Er and Lu, only the Bragg reflections with Miller indices $[0, 0, l]$ are observed, indicating that these compounds have uniaxial magnetocrystalline anisotropy at room temperature, with the EMD being parallel to the crystallographic c axis. The pattern of the Tb based compound shows exclusively reflections with Miller indices $[h, k, 0]$, which reveals that this compound has basal anisotropy, with the EMD lying on the basal plane at room temperature. For magnetic measurements, the powder of the Tb based compound was oriented using a rotating device [6].

The Curie temperature T_C was determined from the magnetization curves obtained in a Faraday balance. The thermomagnetic scans were carried out between room temperature and 1073 K. The samples, in the form of small crushed ingots, were enclosed in silica-glass sample holders sealed under argon.

Magnetic measurements were carried out in a commercial Quantum Design SQUID magnetometer. The complex ac susceptibility, χ_{ac}, was measured covering a temperature range from 5 K to room temperature, with an a.c. exciting field of 5×10^{-4} T and 90 Hz. The magnetization against temperature was measured between 5 and 300 K, with an applied field of 0.05 T, and versus field between zero and 5 T. We have measured these magnitudes both in the direction parallel ($\chi_{\parallel}$, $M_{\parallel}$) and perpendicular ($\chi_{\perp}$, $M_{\perp}$) with respect to the alignment axis of the epoxy-bonded samples; i.e. with respect to their EMD at room temperature.

The saturation magnetization M_s was deduced from the $M_{\parallel}(H)$ curves measured at 1.7 K by extrapolating $M_{\parallel}(1/H^2)$ to $1/H^2 \to 0$. The anisotropy field, H_a, was determined from the intersection of $M_{\perp}(H)$ with $M_{\parallel}(H)$ for increasing field, at a given temperature.

3. Results and discussion

In this section we present the main magnetic features of the RFe$_{11.5}$Ta$_{0.5}$ compounds (they are summarized in table 1). A simple molecular field model has been used to analyse the exchange interactions in this series [7].

Assuming a negligible R–R interaction ($T_{RR} = 0$), the exchange coefficients n_{FeFe} and n_{RFe} can be deduced in the molecular field approximation from the expressions:

$$n_{FeFe} = \frac{T_{Fe}}{C_{Fe}}$$

where

$$C_{Fe} = 4N_{Fe}S^*(S^* + 1)\mu_B^2/3k_B$$

and

$$n_{RFe} = \frac{\sqrt{T_C(T_C T_{Fe})}}{2|\gamma|\sqrt{C_R C_{Fe}}}$$

where $C_R = N_R g_J^2(J + 1)\mu_B^2/3k_B$.

In these expressions N_{Fe} and N_R are the number of Fe and R atoms per unit volume, respectively, $2[S^*(S^* + 1)]^{1/2}\mu_B$ is the Fe atomic effective moment in the paramagnetic state, which is taken as 3.7 μ_B by comparison to other related compounds [1, 7], and $\gamma = 2(g_J - 1)/g_J$.

3.1. LuFe$_{11.5}$Ta$_{0.5}$

For the compound with non-magnetic rare earth (LuFe$_{11.5}$Ta$_{0.5}$), the magnetic moments are aligned along the c axis at room temperature, as deduced from XRD. In figure 1 we show the ac magnetic susceptibility measured in the direction perpendicular to the c axis where no anomaly could be detected. Moreover, neither in the magnetization measurements against temperature nor field any change in the EMD could be observed.

The Curie temperature measured was $T_C = 499$ K. Equating T_C with $T_{Fe} = n_{FeFe}C_{Fe}$, the average Fe–Fe exchange interaction $n_{FeFe} = 211$ μ_0 has been obtained.

The value $M_s = 20.9$ μ_B fu^{-1} was obtained at 1.7 K which, in turn, led to the average Fe magnetic moment of 1.82 μ_B. From the $M(H)$ curves at various temperatures M_s and H_a were deduced. The M_s values are displayed in figure 2. The anisotropy field increases
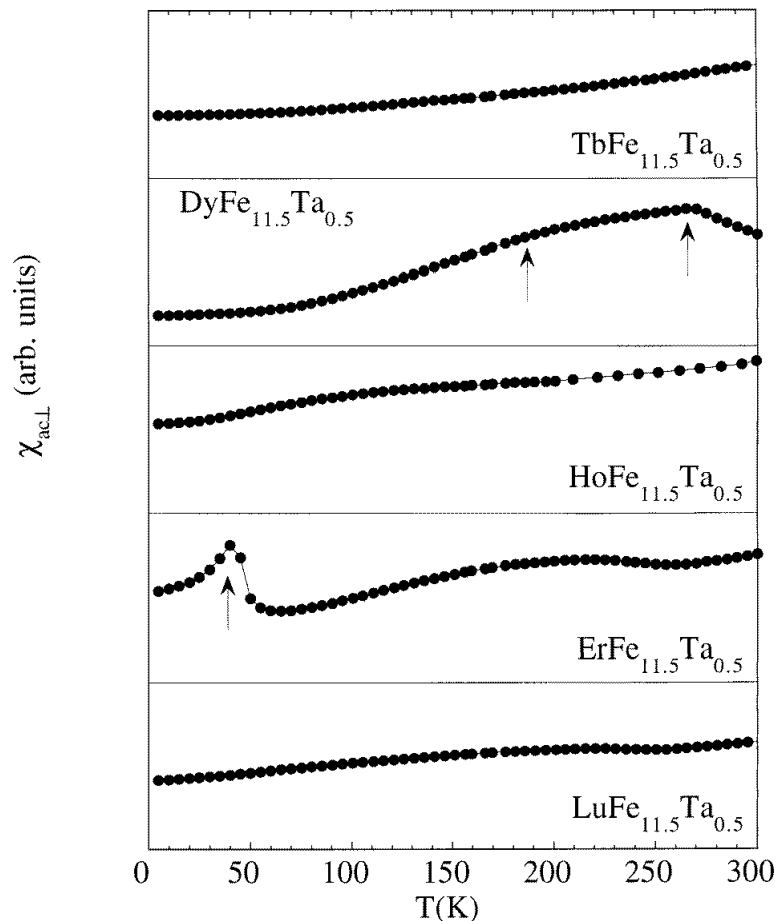
Figure 1. Temperature dependence of the ac susceptibility of $RFe_{11.5}Ta_{0.5}$ measured in the direction perpendicular to the EMD at 300 K. SRTs are marked by arrows.

for decreasing temperature, and ranges between 3 T at 300 K up to 5 T at 100 K, the highest field we could reach.

These parameters are considered to represent the contributions due to the Fe sublattice in the analysis below.

3.2. $ErFe_{11.5}Ta_{0.5}$

The Curie temperature of this compound is $T_C = 532$ K. The average R–Fe interaction, n_{RFe}, deduced by using the molecular-field approximation, yields the value $n_{RFe} = 170\ \mu_0$.

The value of the saturation magnetization at 4.2 K is $M_s = 12.5\ \mu_B$ fu^{-1}, much lower than for the Lu compound, which is consistent with the expected antiferromagnetic (ferrimagnetic) coupling of the heavy R and the Fe sublattice. The M_s thermal evolution is shown in figure 2. Subtracting the value $\mu_{Fe} = 1.82\ \mu_B$ taken from the Lu compound, we obtain the value of the Er moment $\mu_{Er} = 8.4\ \mu_B$, lower than the free-ion value $\mu_{Er} = g_J J \mu_B = 9\ \mu_B$.
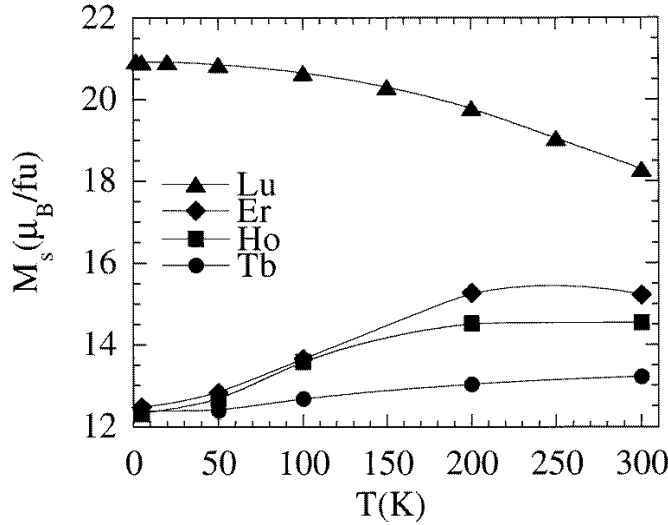
Figure 2. Saturation magnetization as a function of temperature for RFe$_{11.5}$Ta$_{0.5}$.

At room temperature the EMD is aligned along the c axis, as determined from XRD. As the temperature is decreased, no anomalous behaviour is detected from 300 K down to 40 K where a sharp peak appears in $\chi'_{ac}(T)$ (see figure 1). This is evidence of the presence of a thermally activated spin reorientation transition (SRT) at $T_{SR} \approx 40$ K. This SRT is also clearly detected as an increase in $M_{\perp}(T)$ measurements below T_{SR} (figure 3). From the difference between the $M_{\parallel}$ and $M_{\perp}$ versus field measurements we conclude that the low temperature phase is conical. In the axial phase ($T > T_{SR}$), the anisotropy field increases for decreasing temperature, and ranges between 3.6 T at 300 K and 4.8 T at 200 K.

In the RFe$_{12-x}$M$_x$ compounds, the first order crystal electric field (CEF) coefficient A_{20} is negative. Consequently, for the Er case (Stevens factor $\alpha_J > 0$) the first order contribution of the crystal field should add uniaxial anisotropy to the Fe sublattice one. Therefore, the SRT found at low temperatures must be due to an increase of the effect of the CEF terms of order higher than two as temperature is lowered. This is consistent with the quenching of the angular momentum by the crystal field found at 5 K.

3.3. HoFe$_{11.5}$Ta$_{0.5}$

From the value of $T_C = 541$ K the value of $n_{RFe} = 183$ μ_0 was deduced. At room temperature the EMD is aligned along the c axis. It remains so down to the lowest temperature, as proved by the lack of any anomaly in the $\chi'_{ac}(T)$ measurements (figure 1). The value $M_s = 12.3$ μ_B fu^{-1} at 5 K yields to $\mu_{Ho} = 8.6$ μ_B, significantly lower than the free-ion value (10 μ_B). The thermal evolution of M_s is displayed in figure 2.

The $M_{\perp}(H)$ measurements show a first order magnetization process (FOMP) below 125 K, with a maximum critical field $H_c = 1.7$ T (at $T = 5$ K), as deduced from the field of maximum slope in the curve. The EMD turns from the c axis to a conical direction above H_c (figure 4(a)). The value of H_c decreases very slowly for increasing temperature, and becomes unobservable above the threshold temperature $T_{FOMP} \approx 125$ K (see figure 4(b)). In contrast, the anisotropy field, for $T > T_{FOMP}$, decreases for increasing temperature (figure 4(b)).
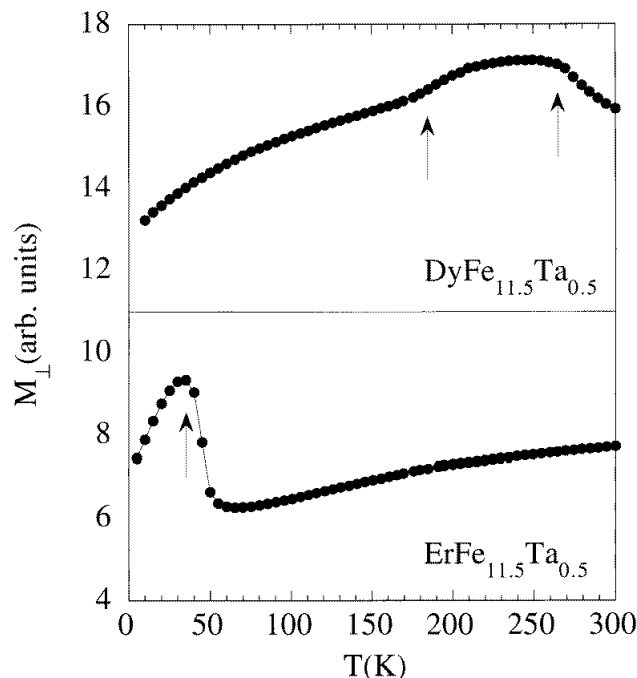
Figure 3. Magnetization versus temperature curves measured in the direction perpendicular to the *c* axis for R ≡ Dy and Er. SRTs are marked by arrows.

In the Ho case α_J is negative, so it is expected that the R sublattice favours a planar anisotropy. However, the HoFe$_{11.5}$Ta$_{0.5}$ compound exhibits uniaxial magnetocrystalline anisotropy in the whole temperature range. The contribution to the anisotropy stemming from the Ho sublattice shows up when, at relatively low temperature and field, the FOMP to conical phase takes place. In this thermal range ($T < 150$ K), the CEF terms of order higher than two may be important, as indicates the very low value of the R moment at 5 K, so they would give a significant contribution to the existence and character of the FOMP.

3.4. DyFe$_{11.5}$Ta$_{0.5}$

In this compound $T_C = 550$ K and $n_{RFe} = 176$ μ_0. The value $M_s = 11.7$ μ_B fu^{-1} yields $\mu_{Dy} = 9.2$ μ_B, also lower than in the free state (10 μ_B). In the $\chi'_{ac}(T)$ measurements a peak is detected at $T_{SR1} = 265$ K and a change in slope at $T_{SR2} = 185$ K (figure 1). These two transitions are confirmed in the $M_\perp(T)$ measurements (figure 3). They correspond, as temperature is reduced, to an SRT from axial to conical phase at T_{SR1}, and probably from conical to basal phase below T_{SR2}, as we may conclude from our $M_\parallel(H)$ and $M_\perp(H)$ measurements and from the similarity to the χ'_{ac} and $M_\perp(T)$ data in DyFe$_{11.35}$Nb$_{0.65}$ [4].

Besides, at $T < T_{SR2}$, when the EMD lies on the basal plane, the $M_\parallel(H)$ curves show a FOMP type transition to a conical phase (figure 5(*a*)). Such a type of transition has also been detected in the DyFe$_{11}$Ti compound [8, 9], which undergoes two spin reorientation transitions ($T_{SR1} \approx 200$ and $T_{SR2} \approx 50$ K) and a FOMP at low temperature and field. In the DyFe$_{11}$Ti case, when $T < 50$ K, the FOMP takes place, as the field increases, from the basal phase corresponding to $T < T_{SR2}$ to the conical phase characteristic of $T > T_{SR2}$.
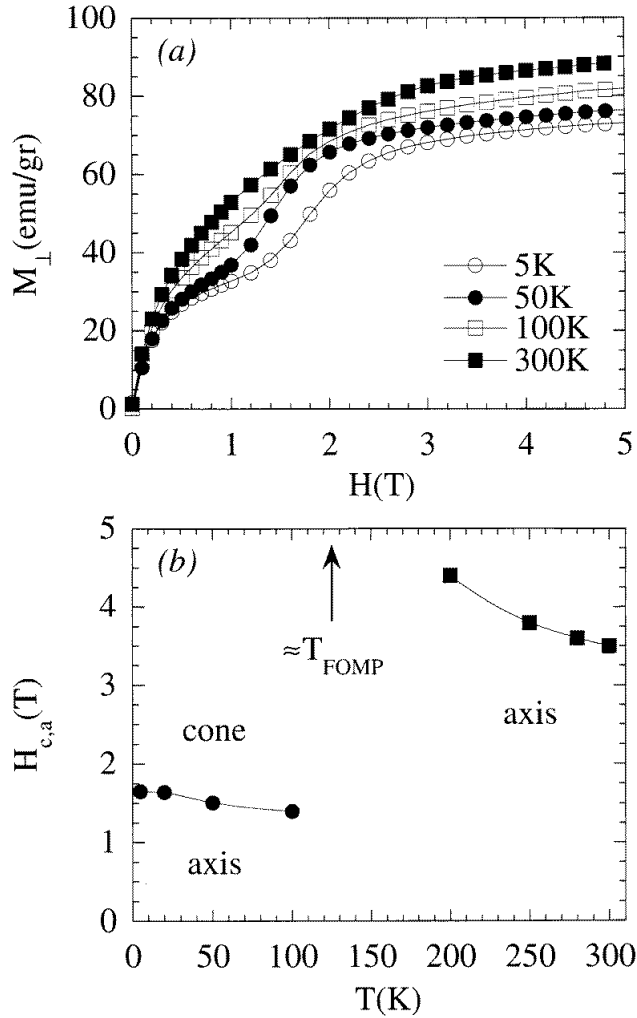
Figure 4. (*a*) Magnetization versus field curves measured at various temperatures in the direction perpendicular to the *c* axis. (*b*) Thermal evolution of H_c (full circles) and H_a (full squares) of HoFe$_{11.5}$Ta$_{0.5}$. The FOMP temperature is marked by an arrow.

We conjecture by similarity that DyFe$_{11.5}$Ta$_{0.5}$ undergoes a FOMP from basal to a conical phase similar to its zero field phase at temperatures between T_{SR1} and T_{SR2}.

The anisotropy field H_a could be determined for temperatures above T_{SR2} and below T_{SR1} (figure 5(*b*)); H_a increases when approaching T_{SR1} from above, and decreases below, tending to zero as T tends to T_{SR2}.

3.5. TbFe$_{11.5}$Ta$_{0.5}$

For the Tb compound $T_C = 576$ K, yielding to the value of $n_{RFe} = 171$ μ_0. As in the previous compounds, the value $M_s = 12.4$ μ_B fu^{-1} yields a μ_{Tb} value lower than in the free state, $\mu_{Tb} = 8.5$ μ_B, against $g_J J \mu_B = 9$ μ_B. The compound is basal at room temperature, as evidenced in the XRD results, and remains so at all measured temperatures down to 5 K.
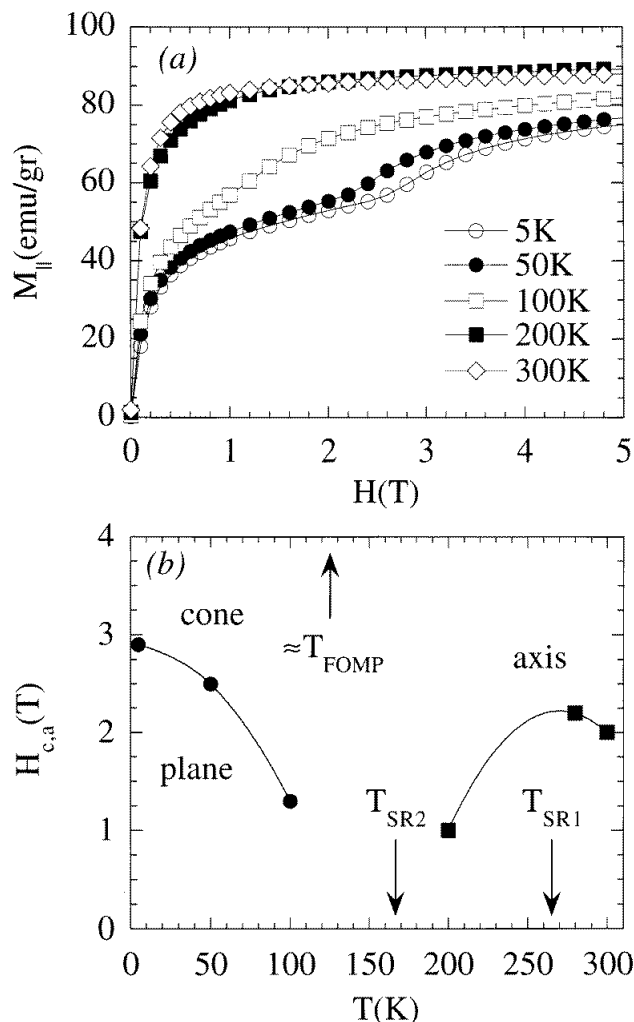
Figure 5. (*a*) Magnetization versus field curves measured at various temperatures in the direction parallel to the *c* axis. (*b*) Thermal evolution of H_c (full circles) and H_a (full squares) of DyFe$_{11.5}$Ta$_{0.5}$. SRTs and FOMP temperatures are marked by arrows.

This is in contrast to other TbFe$_{12-x}$M$_x$ compounds in which one or two SRTs have been detected [10–13]. No FOMP has been detected either. Finally, the anisotropy field also increases for decreasing temperature, and ranges between 3 T at 300 K and 5 T at 200 K, the highest field we could reach.

In the introduction we proposed that the electronic similarities of atoms of the same group were, to our understanding, the paramount feature in the stabilization of the 1:12 compounds, rather than the diagonal position relationship. The fact that we could synthesize the Ta compound proves it, since:

(a) it completes the Va group series (V, Nb, Ta);
(b) the substitution following the diagonal relation, Zr, does not stabilize homogeneous

1:12 compounds. Indeed, in the literature there are no reports on Zr compounds, and in spite of efforts on our side, we always found other compositions. Moreover, the VIa group substitutions (Cr, Mo and W) also exist.

It is noteworthy that the Va group Nb and Ta series, with $x \approx 0.5$, have almost identical magnetic properties. The Tb compounds are basal in the thermal range studied, 5–300 K, the Dy compounds undergo two SRTs ($T_{SR1} = 245$ K and $T_{SR2} = 160$ K, for Nb and $T_{SR1} = 265$ K and $T_{SR2} = 185$ K for Ta), the Ho compounds remain axial and the Er based compounds show an SRT at $T \approx 40$ K. Besides, the Fe sublattice magnetic properties derived from the Lu compounds are almost the same (see table 2). Consequently, Ta and Nb causes the same perturbation in the Fe sublattice. We note that the Ta and Nb ions not only belong to the same group and enter in the lattice with the same rate of M substitution, but they also have the same atomic radius ($r = 1.63$ Å). In fact, we think that the value of the atomic radius plays an important role in the properties of the RFe$_{12-x}$M$_x$ series.

Table 2. Comparison of the parameters T_C, n_{FeFe} and μ_{Fe} among different RFe$_{12-x}$M$_x$ compounds with non-magnetic rare-earth (R ≡ Lu or Y) (in all cases, n_{RFe} has been calculated using the same value of $2[S^*(S^*+1)]^{1/2}\mu_B = 3.7$ μ_B, see details in the text).

Compound	T_C (K)	n_{FeFe} (μ_0)	μ_{Fe} (μ_B)	Ref.	Compound	T_C (K)	n_{FeFe} (μ_0)	μ_{Fe} (μ_B)	Ref.
LuFe$_{11.5}$Ta$_{0.5}$	499	211	1.82		YFe$_{11}$Mo	472	207	2.1	[3]
LuFe$_{11.65}$Nb$_{0.35}$	489	210	1.81	[4]		480	212	2.0	[15]
YFe$_{11.65}$Nb$_{0.35}$	526	226	1.88	[4]	YFe$_{10.8}$W$_{1.2}$	525	235	2.1	[16]
YFe$_{11.5}$Mo$_{0.5}$	491	205	2.14	[3]		500	224	1.9	[17]
YFe$_{10}$Cr$_2$	514	246	1.88	[13]		510	230	1.8	[18]
YFe$_{10}$Mo$_2$	317	156	1.40	[14]	YFe$_{10.8}$Re$_{1.2}$	460	206	1.8	[18]

We can extend the study of the effect of the atomic radius of M by comparing the magnetic properties of the RFe$_{12-x}$M$_x$ series. It is important to keep in mind that the magnetic properties are strongly dependent on the x value [1], and we must compare compounds with the same rate of substitution.

In the case $x \approx 0.5$, unfortunately there are no reports on the full series RFe$_{11.5}$M$_{0.5}$; only the compounds with M ≡ Mo and R ≡ Y, Nd and Sm have been described. So, we can only compare the compounds YFe$_{11.35}$Nb$_{0.65}$ [4] and YFe$_{11.5}$Mo$_{0.5}$ [3]. In table 2 we can see that in this case the T_C, n_{FeFe} and μ_{Fe} values are quite different between them and from the values obtained in the Nb and Ta based compounds.

If we compare element substitution such as Cr [13] and Mo [14] with the same value of x ($x = 2$) belonging to the same periodic group (VIa) but with different atomic radius (1.42 Å for Cr and 1.55 Å for Mo) along the heavy rare earths, we can observe several differences. In the Cr case, the Tb, Dy and Er compounds undergo an SRT at 298, 190 and 25 K respectively. In the Mo based compound, the Tb compound remains axial and the Dy and Er ones undergo SRTs at 137 and 180 K respectively. The properties of the non-magnetic rare-earth based compounds are also fairly different among them, as we can see in table 2.

A further comparison may be drawn from compounds with $x \approx 1$ substitution, with M ≡ Mo, W and Re, all of which have very similar radius ($r = 1.55$, 1.56 and 1.52 Å, respectively), although there is a certain scatter of published data. The values of T_C, n_{FeFe} and μ_{Fe} for the compounds YFe$_{11}$Mo [3, 15], YFe$_{10.8}$W$_{1.2}$ [16–18] and YFe$_{10.8}$Re$_{1.2}$ [18] are displayed in table 2. In Mo, W and Re the differences are less pronounced than in the

Nb, Ta and Mo case, in concordance with the fact that the atomic radius is more similar. (For the sake of comparison, in all cases, n_{RFe} has been calculated using the same value of $2[S^*(S^* + 1)]^{1/2}\mu_B = 3.7~\mu_B$, to avoid discrepancies derived from the fact that some authors take $S^* = 1$ when they calculate n_{RFe}.)

Therefore, the comparison suggest that the value of the atomic radius of the M element plays an important role in the magnetic properties of the $RFe_{12-x}M_x$ compounds.

4. Conclusions

Compounds of the type $RFe_{11.5}Ta_{0.5}$ ($R \equiv$ Tb, Dy, Ho, Er and Lu) isotypic with $ThMn_{12}$ have been synthesized with a very narrow Ta stability region.

The Fe sublattices have c-axis easy anisotropy in the thermal range 5–300 K. All the $RFe_{11.5}Ta_{0.5}$ with magnetic rare-earth elements show easy c-axis anisotropy at room temperature, except for $R \equiv$ Tb.

In all compounds the anisotropy field at 300 K has been found to be somewhat higher than the values found in the similar compounds $RFe_{11.35}Nb_{0.65}$ [4]. In all cases, the increase of H_a with increasing temperature is also higher than in the Nb series. Thus, the anisotropy seems to be larger in the Ta based compounds. However, caution must be paid to this result since the experimental technique employed in the Nb series is singular point detection (SPD), different from ours.

SRTs have been observed in $DyFe_{11.5}Ta_{0.5}$ and $ErFe_{11.5}Ta_{0.5}$ compounds.

In the $DyFe_{11.5}Ta_{0.5}$ and $HoFe_{11.5}Ta_{0.5}$ compounds a FOMP has been observed at low temperatures. With increasing temperature the FOMP becomes less pronounced and disappears around 125 K.

The molecular field approximation applied to the $RFe_{11.5}Ta_{0.5}$ compounds yields the average value of the exchange interaction $\langle n_{RFe} \rangle = 172~\mu_0$ and $n_{FeFe} = 211~\mu_0$ as derived from the Lu based compound. For the different R compounds n_{RFe} is nearly constant. This corroborates that the R–Fe exchange interaction varies little along the heavy R lanthanide series, as has been found in the $R_2Fe_{14}B$, R_2Fe_{17} and, indeed, in the $RFe_{12-x}M_x$ compounds.

The comparison among the different series of the $RFe_{12-x}M_x$ compounds seems to indicate that the value of the atomic radius, for the same rate of substitution, has an important role in the magnetic properties of these compounds, particularly in the perturbation of the Fe sublattice caused by the M substitution.

Acknowledgments

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