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Large (H,D) isotope effect on the metamagnetic transition in $Y_{0.9}R_{0.1}Fe_2(H,D)_{4.3}$ compounds

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The influence of the (H,D) isotope substitution on the metamagnetic transition in $(Y_{1-x}R_x)Fe_2(H_{1-y}D_y)_{4.2}$ (R=Tb, Er, x=0, 0.1, and y=0, 1) compounds has been studied by x-ray diffraction and magnetic measurements. All these compounds crystallize in the same monoclinic structure with larger cell volume for the hydrides compared to the deuterides. The metamagnetic transition temperature T_M is not only field dependent but also strongly related to the volume change. The increase of T_M is more strongly related to the (H,D) isotope substitution than by the rare earth substitution on the Y site. © *2006 American Institute of Physics.* [DOI: 10.1063/1.2172204]

I. INTRODUCTION

The YFe_2D_x deuterides are ferromagnetic with an increase of the mean Fe moment and a decrease of T_C for $x \leq 3.5$ D/mol.[1] For x=4.2, the monoclinic compound is ferromagnetic at low temperature then undergoes a sharp first-order magnetovolumic transition towards an antiferromagnetic structure at 84 K.[2] This transition has been attributed to an itinerant electron metamagnetic (IEM) behavior of one of the Fe sites which is surrounded by about 5 D atoms.[3] Surprisingly this transition is very sensitive to the H for D substitution, which increases the mean Fe moment at 4.2 K and shifts the transition temperature to 130 K (50% increase). Since the cell volume of the hydride is 0.78% larger than the deuteride such a giant isotope effect has been attributed to the strong influence of the volume on the IEM behavior.[4] In order to understand better the interplay between magnetic and elastic energies on this transition, we have undertaken the study of $Y_{0.9}R_{0.1}Fe_2(H,D)_{4.2}$ compounds (R=Er,Tb). R for Y substitution introduces both volume changes and internal molecular field of the rare earth. The influence of these different parameters on the first order metamagnetic transition, in particular, its sensitivity to the large (H,D) isotope effect, will be presented and discussed.

II. EXPERIMENT

The $(Y_{1-x}R_x)Fe_2$ intermetallic compounds were prepared by induction melting of the pure elements followed by 3 weeks annealing treatment at 1100 K. Their composition and homogeneity were checked by x-ray diffraction (XRD) and electron probe microanalysis as described in Ref. 2. The $(Y_{1-x}R_x)Fe_2(H_{1-y}D_y)_{4.2}$ compounds were prepared by solid-gas reaction using a Sievert apparatus and their homogeneity was checked by XRD. XRD measurements were carried out at 300 K using a Bruker D8 diffractometer (Cu $K\alpha$ radiation). The XRD patterns were refined using the FULLPROF code.[5] The magnetization measurements were performed using a physical properties measurement system (PPMS) magnetometer from Quantum Design operating up to 9 T and from 2 to 300 K for all samples and a high-magnetic-field magnetometer of the LCMI laboratory using extraction method ($B \leq 230$ kG) for the $YFe_2(H,D)_{4.2}$ samples.
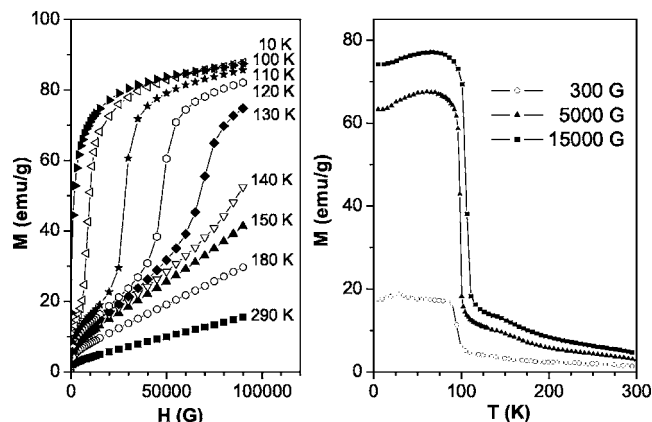
III. RESULTS

A. X-ray diffraction

The $Y_{0.9}R_{0.1}Fe_2$ (R=Tb,Er), alloys crystallize in the same cubic $C15$ structure as YFe_2 with slightly smaller cell parameters [a=7.355(1) Å for Tb, a=7.348(1) Å for Er, and a=7.356(1) Å for YFe_2]. These cell parameter variations are in agreement with the lanthanide contraction. These alloys were progressively loaded with H (D) until a concentration close to 4.3 and the observation of a sharp pressure increase, indicating that a single hydride (deuteride) phase was obtained. All these samples crystallize in the same monoclinic structure refined in the $C2/m$ space group[2] and their cell parameters are reported in Table I. The compounds have been sorted by increasing cell volume. These results show that for all samples the hydrides have larger cell parameters than the corresponding deuterides, but this is not related to a larger H/D content since the opposite is observed. In addition whereas the hydrides (deuterides) of the $Y_{0.9}Er_{0.1}Fe_2$ compounds have a lower cell volume than the YFe_2 ones, the $Y_{0.9}Tb_{0.1}Fe_2$ hydrides (deuterides) have a larger cell volume. This means that H (D) absorption in the substituted alloys

TABLE I. Cell parameters of the $Y_{1-x}R_xFe_2$ deuterides and hydrides in the monoclinic ($C2/m$) structure.

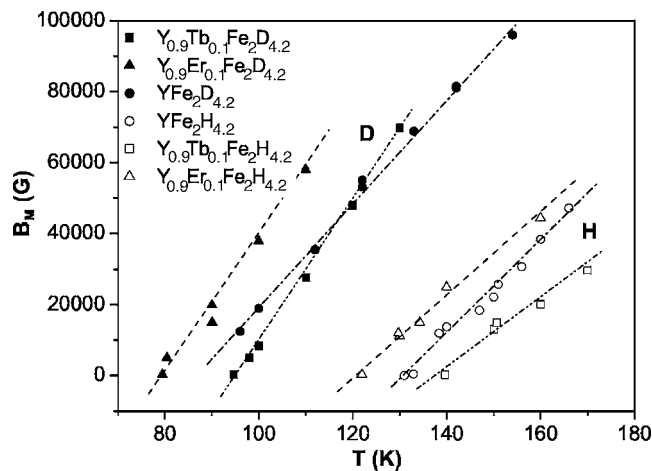| Compounds	a (Å)	b (Å)	c (Å)	β (°)	V (Å^3)
$Y_{0.9}Er_{0.1}Fe_2D_{4.55}$	9.429(2)	5.734(1)	5.505(1)	122.397(2)	251.34(1)
$YFe_2D_{4.30}$	9.430(1)	5.735(2)	5.506(2)	122.351(1)	251.48(2)
$Y_{0.9}Tb_{0.1}Fe_2D_{4.50}$	9.436(2)	5.743(1)	5.510(1)	122.338(1)	252.26(1)
$Y_{0.9}Er_{0.1}Fe_2H_{4.26}$	9.440(2)	5.745(2)	5.515(2)	122.361(1)	252.65(2)
$YFe_2H_{4.24}$	9.451(2)	5.745(2)	5.517(1)	122.350(2)	253.40(1)
$Y_{0.9}Tb_{0.1}Fe_2H_{4.24}$	9.455(2)	5.755(2)	5.523(1)	122.348(2)	253.90(2)

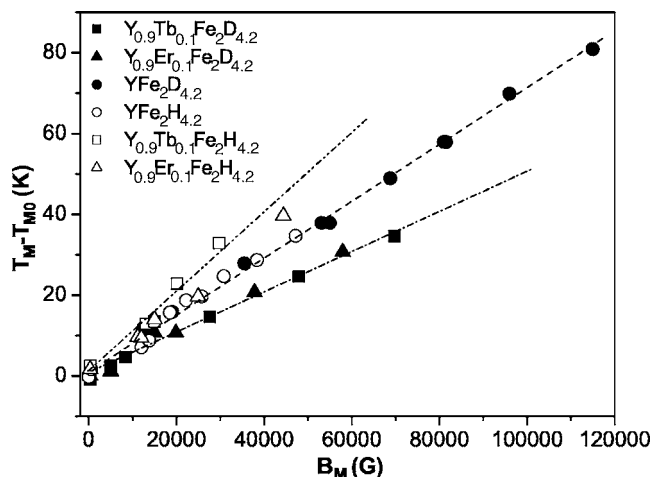[a)]Electronic mail: mguillot@grenoble.cnrs.fr

FIG. 1. Magnetization isofields (a) and isotherms (b) of $Y_{0.9}Tb_{0.1}Fe_2D_{4.2}$.

FIG. 3. Evolution of the difference T_M-T_{M0} vs the applied magnetic field for all compounds.

leads to a larger volume expansion than in pure YFe_2. For the deuterides this can be related to a larger D solubility, but such argument seems not true for the hydrides. Note that the monoclinic β angle remains very similar for all samples, indicating that no strong variation of the monoclinic distortion occurs.

B. Magnetization measurements

Isotherm and isofield magnetizations were measured for all the hydrides and deuterides at selected fields and temperatures. As shown in Fig. 1 for $Y_{0.9}Tb_{0.1}Fe_2D_{4.2}$, the isotherm curves display a metamagnetic behavior above a critical temperature, which corresponds to a steep decrease of the magnetization in the isofield measurements. This magnetization step, which has been studied in detail for $YFe_2D_{4.2}$, corresponds to a transition from a ferromagnetic to an antiferromagnetic structure related to the IEM transition of one of the Fe sites.[3] In Fig. 2, the critical field B_M is plotted versus the temperature for all the samples. The linear extrapolation of the curves to $B=0$ leads to transition temperatures $T_M(B=0)$ denoted as T_{M0}, which for YFe_2 were found in good agreement with neutron diffraction and differential scanning calorimetry measurements.[4] The variation of $T_M(B)-T_{M0}$ vs B_M ($\Delta T/B$) is linear for each sample (Fig. 3),

but with slightly different slopes for the substituted compounds ($R=$Er, Tb): the slopes of the corresponding hydrides are larger than those of $YFe_2H_{4.2}$ whereas they are smaller for the deuterides compared to $YFe_2D_{4.2}$. The variation of T_{M0} versus the cell volume (Fig. 4) clearly indicates that the larger the volume the higher the transition temperature. Nevertheless this variation is not linear for all the studied compounds since the hydrides and the deuterides are aligned on two different lines as indicated by dash lines.

IV. DISCUSSION

In Ref. 4 the giant isotope effect observed for $YFe_2(H,D)_{4.2}$ has been attributed, in relation to other experimental and theoretical works related to the IEM effect, to the volume change caused by the H/D substitution. This volume difference, currently observed in metal-hydride systems, can be related to the isotope mass influence on the amplitude of the zero point vibration.[6] With the present work, we can conclude that cell volume variation has truly a major influence on the IEM transition. But, as seen in Fig. 4, the variation of T_{M0} versus cell volume is different for the hydrides and the deuterides. Moreover the analysis of the magnetic

FIG. 2. Evolution of the transition field B_M vs temperature for all compounds.
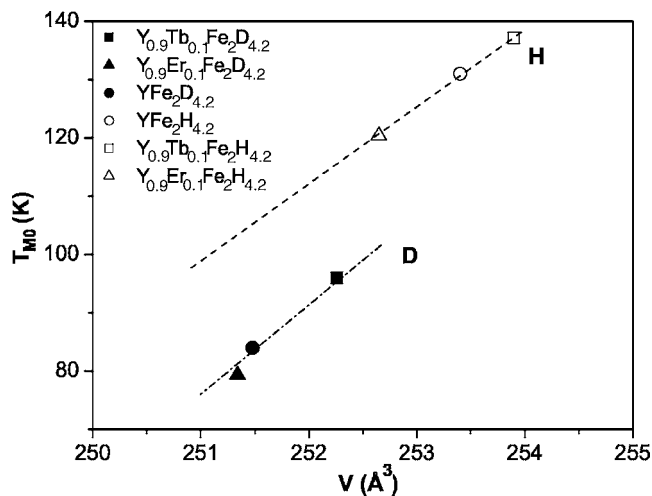
FIG. 4. Evolution of T_{M0} vs the cell volume for all compounds.

data indicated slightly different $\Delta T/B$ slopes for hydrides and the deuterides of the substituted compounds (Fig. 3). In $R\mathrm{Co}_2$ compounds, which display an IEM effect, a linear relation between B and T^2 was generally observed[7] whereas in the present systems B is linear versus T. This difference may be related to the fact that in $\mathrm{YFe}_2(\mathrm{H,D})_{4.2}$ compounds the field dependent transition is not only due to an IEM behavior of one of the Fe sublattices but also to the associated ferro-antiferromagnetic transition.[3,4] Nevertheless, these results mean that although the cell volume is a critical parameter for the IEM transition, additional effects have to be taken into account. Since the IEM effect is strongly related to the density of states (DOS) at the Fermi level any change of the electronic structure, such as the contribution of additional $4f$ electrons in the valence band, can influence the metamagnetic transition.[8] The presence of the molecular field created by the magnetic substituted rare earth can also modify the stability of the Fe moment as it has been observed for Co in $R\mathrm{Co}_2$ compounds.[9]

These results already confirm the strong interplay between elastic and magnetic energies for this intriguing $R\mathrm{Fe}_2(\mathrm{H,D})_{4.2}$ system. In order to get a more complete understanding of these competitive effects, further studies with larger R substitutions or other R elements are currently under progress.

V. CONCLUSION

The large influence of the (H,D) isotope effect on the metamagnetic transition previously observed in $\mathrm{YFe}_2(\mathrm{H,D})_{4.2}$ is maintained upon 10 at. % of Y by Er and Tb substitutions. Volume difference between hydrides and deuterides is clearly a key parameter to explain the large difference in the transition temperature T_{M0}, but additional effects related to changes of the electronic structure and internal molecular field of the rare earth should also be probably considered.

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