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The magnetocrystalline anisotropy in Y(Fe,Co)$_{11}$TiH single crystals

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Abstract

The influence of hydrogen on the magnetocrystalline anisotropy of YFe$_{11-x}$Co$_x$Ti ($0 \le x \le 3$) single crystals has been studied in magnetic fields of up to 60 kOe in a temperature range of 4.2–700 K using torque and capacitance magnetometry. In the entire investigated temperature range the easy magnetization direction of YFe$_{11-x}$Co$_x$TiH single crystals with $x \le 3$ was found to coincide with the c-axis. It was established that hydrogenation leads to an increase in the anisotropy field of the transition-metal sublattice; the magnetic anisotropy constants K_1 were determined. The concentration dependencies of K_1(x) for the YFe$_{11-x}$Co$_x$TiH single crystals shows a maximum around $x = 2$, whereas, for the initial YFe$_{11-x}$Co$_x$Ti alloys the maximum was observed at $x = 1$. At $x > 2$, the K_1 value rapidly decreases with increasing of Co content.

Keywords: Hydrides; Ferromagnetism; Magnetic anisotropy

1. Introduction

The results of intensive and systematic studies of the permanent magnet material Nd$_2$Fe$_{14}$B have drawn attention to other ternary intermetallics [1]. Among them, the RFe$_{11}$Ti compounds provide a focus of interest [2,3]. These compounds have high Curie temperatures ranging from 490 to 610 K. Some of them, for example SmFe$_{11}$Ti, which has a large magnetocrystalline anisotropy (MKA) and a high saturation magnetization, can be considered as potential candidates for permanent magnet materials. The relatively simple tetragonal crystalline structure is an attractive feature of the RFe$_{11}$Ti compounds which have a structure of the ThMn$_{12}$ type (space group $I4/mmm$) and a body-centered tetragonal Bravais cell ($a \sim 8.5$ Å, $c \sim 4.8$ Å, and $c/a \sim 0.56$).

A standard approach to improve the magnetic properties of iron-based permanent magnet materials is to add small amounts of cobalt. This was found to increase both the magnetization and Curie temperature of magnetic materials. It is also known that the hydrogenation of RFe$_{11}$Ti compounds leads to an increase of the magnetization and Curie temperature [4–7]. The insertion of hydrogen has been shown to increase the uniaxial anisotropy for YFe$_{11}$Ti [8]. However, no studies of the magnetic anisotropy variations caused by simultaneous substitution of Fe for Co and hydrogen insertion have been performed. The aim of the present work is to study the effect of hydrogenation on the magnetic anisotropy of YFe$_{11-x}$Co$_x$Ti ($0 \le x \le 3$). These compounds are of interest both from a fundamental physics point of view and because of their potential application. In this work MCA values of single crystal samples were studied.

2. Experimental details

The YFe$_{11-x}$Co$_x$Ti samples, prepared as described in [9], were used to synthesize the hydride samples by a gas–solid

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reaction. Procedures for preparing the hydrides of these compounds also were described in [10]. The hydrogen content, one H atom per formula unit, was determined by a volumetric method.

The $YFe_{11-x}Co_xTi$ ($0 \leq x \leq 3$) single crystals were oriented along the principal symmetry axes using a back Laue method. The magnetization measurements were carried out on the single crystals using an automated capacitance magnetometer. All measurements were performed in a temperature range from 4.2 K to room temperature in magnetic fields of up to 60 kOe applied along and perpendicular to the *c*-axis. Torque measurements were done using an anisometer in a temperature range of 77–700 K in a field of 16 kOe. In the magnetic properties measurements the applied magnetic field H was corrected with respect to an inner demagnetizing field.

3. Results and discussion

In an earlier paper [9], we have reported the structural and magnetic properties of $YFe_{11-x}Co_xTi$ compounds. All the $YFe_{11-x}Co_xTiH$ hydrides are found maintain the $ThMn_{12}$-type tetragonal structure and no formation of the α-Fe phase is observed. The incorporation of hydrogen atoms in the crystalline lattice of these compounds results in changes of the lattice parameters (a and c) and unit cell volume V (see Table 1). The increase of the unit cell volume caused by hydrogenation is 0.8–0.9%.

It is known that the most reliable data on the MCA can be obtained from magnetization and torque curves measured on single-crystal samples. Fig. 1 shows experimental torque curves $L(\theta)$ measured with $H = 16$ kOe for the (1 1 0) single-crystal $YFe_{10}CoTiH$ disk; Fig. 2 demonstrates magnetization curves $M(H)$ measured for a YFe_8Co_3TiH single crystal along the principal crystallographic axes. All measurements were performed at different temperatures. The shape of $L(\theta)$ and $M(H)$ curves are typical of high uniaxial anisotropy magnets. The easy magnetization direction (EMD) for $YFe_{10}CoTiH$ and YFe_8Co_3TiH single crystals coincides with the [0 0 1] direction (*c*–axis).

In order to determine the magnetic anisotropy constants at each temperature, the M versus H variations along [1 1 0] were analyzed using a Sucksmith–Thompson method [11]. At high temperatures, when the external magnetizing field was exceeded the anisotropy field, the magnetic anisotropy constants were obtained from the torque curves by separation of successive harmonics using a least-squares method. Only the first anisotropy constant K_1 needs to be considered for an adequate description of the experimental data. The temperature dependence of K_1 for $YFe_{11-x}Co_xTiH$ ($0 \leq x \leq 3$) is represented in Fig. 3. It is seen that K_1 decreases continuously with temperature increase for all the investigated compounds.

Table 1
Structural data for $YFe_{11-x}Co_xTiH_y$ ($y = 0$ and 1) single crystals

Compounds	a (Å)	c (Å)	V (Å^3)	$\Delta V/V$ (%)
$YFe_{10}CoTi$	8.535	4.799	349.6	–
$YFe_{10}CoTiH$	8.562	4.809	352.5	0.8
YFe_9Co_2Ti	8.529	4.793	348.7	–
YFe_9Co_2TiH	8.555	4.804	351.6	0.8
YFe_8Co_3Ti	8.519	4.785	347.2	–
YFe_8Co_3TiH	8.548	4.794	350.3	0.9

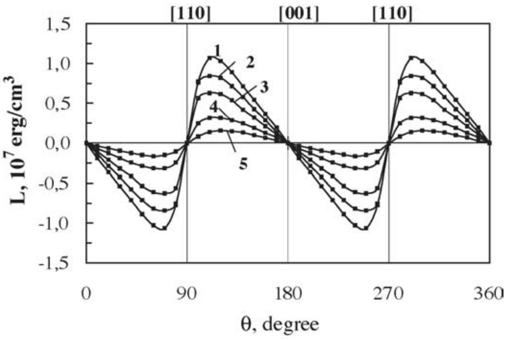

Fig. 1. The torque of the $YFe_{10}CoTiH$ sample ((1 1 0) disk) measured at different temperatures: (1) 150 K; (2) 250 K; (3) 350 K; (4) 550 K; (5) 650 K.

The composition dependencies of the anisotropy constant $K_1(x)$ demonstrate a more complex behavior. Fig. 4 shows the $K_1(x)$ curves for $YFe_{11-x}Co_xTi$ [9] and $YFe_{11-x}Co_xTiH$ compounds at $T = 300$ K. For the initial compounds, the first anisotropy constant reaches a weak maximum value at $x = 1$ and then decreases with increase of the Co content. Since the Fe local anisotropy is uniaxial, whereas the Co's is planar, the change of the K_1 sign takes place within the composition range $5 < x < 6$ [12]. The concentration dependence of $K_1(x)$ for the $YFe_{11-x}Co_xTiH$ single crystals shows a strong maximum near $x = 2$. At $x > 2$, K_1 decreases rapidly with an increase of the Co content. It is likely that the change of the K_1 sign of the compounds can be observed as the x value increases from 3 to 4; it is shown by a dotted curve in Fig. 4. Hydrogenation leads to an increase of the K_1 anisotropy constant of all the compounds explored in this work.

In $RFe_{11}Ti$ compounds with the $ThMn_{12}$-type structure, the rare-earth atoms (R) occupy equivalent sites with the [0 0 0] and [1/2 1/2 1/2] coordinates; iron and titanium atoms occupy three general positions—8*f*, 8*i*, and 8*j* [13]. These positions of iron atoms are nonequivalent due to the difference in their symmetries and the predominant occupation of the 8*i* positions with titanium atoms due to the valence configuration of Ti atoms which differs from that of Fe atoms and the difference in the ion radii. Upon substitution of the Fe atoms, the Co atoms were observed to predominantly occupy the 8*f* and 8*j* positions [14].

The rather high uniaxial magnetocrystalline anisotropy of the investigated compounds with Y (nonmagnetic element) can be explained, in part, by the "defreezing" of the orbital angular momentum L of the 3d-ions in the anisotropic local crystal field. A small "defrozen" orbital angular momentum

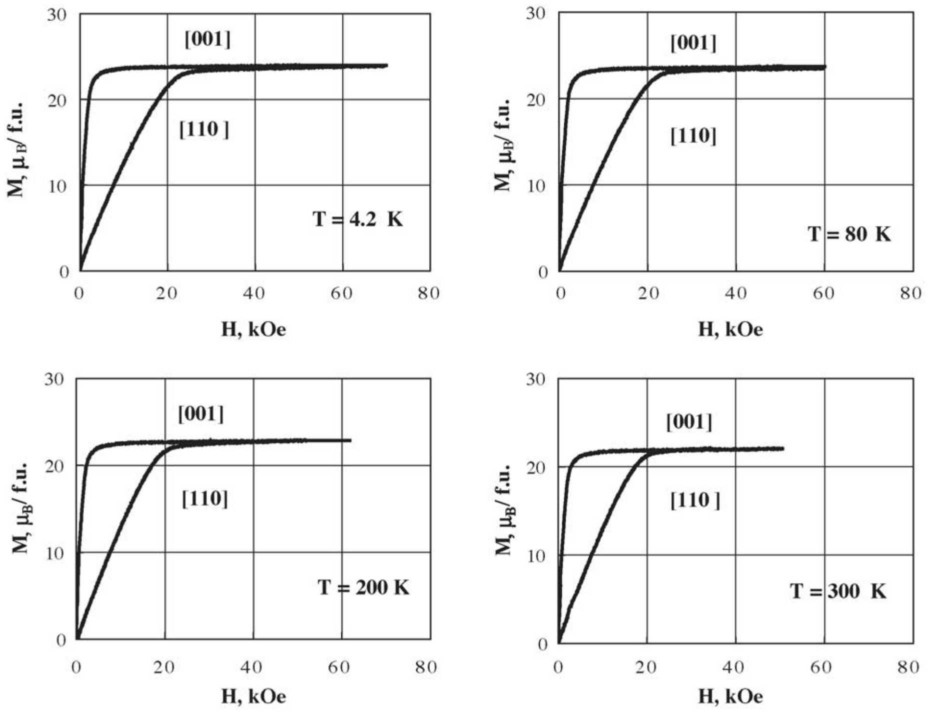

Fig. 2. Field magnetization curves of YFe_8Co_3TiH single crystal measured at different temperatures in the magnetic field applied along [0 0 1] and [1 1 0] directions.

(the component of the orbital angular momentum L) is aligned with the easy magnetization direction and, in turn, orients the total spin angular momentum at the expense of spin-orbital interaction.

Hydrogenation brings an increase in the magnetocrystalline anisotropy of the $YFe_{11-x}Co_xTi$ ($0 \leq x \leq 3$) compounds. It can be related to either the increase in the orbital angular momentum L of the 3d-ions or to the change in the local crystal field upon incorporation of hydrogen atoms within the first coordination shell of the 3d-ions. The magnetic anisotropy constant K_1 for the initial compounds can be represented as the sum of three partial constants corresponding to the contributions from the 3d-ions located at three crystallographically nonequivalent positions (8*i*, 8*j*, and 8*f*); it is [15]:

$$K_1(T) = K_{1i} + K_{1j} + K_{1f}$$

Upon introduction of the hydrogen atoms into the $ThMn_{12}$-type crystal structure, light interstitial atoms are located in the closest proximity to the 8*j* positions. Hence, they strongly affect the iron ions (in compounds with the low cobalt concentration) occupying these positions. The experimental data indicates that, the presence of H atoms in these positions leads to an increase in the total K_1 anisotropy constant. Therefore, a significant increase in the partial constant K_{1j} can be noted. As the Co content increases, Co atoms begin to occupy predominantly the 8*f* positions and this results in an abrupt decrease of K_1.

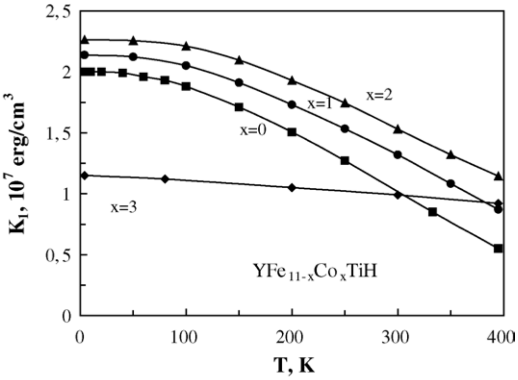

Fig. 3. Temperature dependences of the magnetic anisotropy constant $K_1(T)$ for $YFe_{11-x}Co_xTiH$ with $0 \leq x \leq 3$.

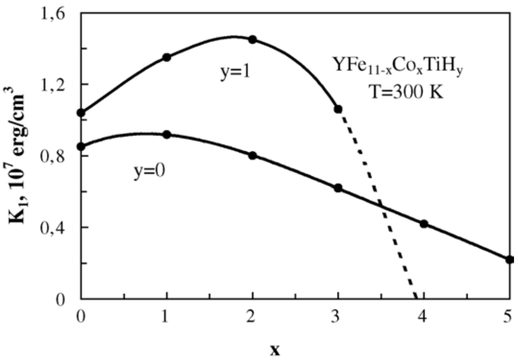

Fig. 4. Composition dependences of the magnetic anisotropy constant $K_1(x)$ for $YFe_{11-x}Co_xTiHy$ ($y = 0; 1$) at $T = 300$ K.

Upon hydrogenation of $YFe_{11-x}Co_xTi$ an increase in the interatomic spacings and changes in the electron structure of the compounds take place. Experimental data on the pressure effect on the anisotropy constant K_1 of $YFe_{11}Ti$ show that $\partial K_1/\partial p$ is close to zero [16], i.e. the magnetovolume effect is negligible for the transition-metal sublattice. Thus, in this case, it can be concluded that the change in the electron structure of the $YFe_{11-x}Co_xTiH$ compounds is a determining factor in the variations in the magnetocrystalline anisotropy.

4. Conclusion

The insertion of hydrogen atoms in the crystal lattice of the $YFe_{11-x}Co_xTi$ ($0 \leq x \leq 3$) compounds does not change their crystal structure and results in the increase of the unit cell volume. Upon hydrogenation, a significant increase of K_1 of the investigated compounds was observed. It occurs because of an increase in the 3d orbital moment or a change in the crystal field induced by the interstitial H atoms. It was found that the composition dependence of the anisotropy constant $K_1(x)$ of the hydrogenated compounds exhibited a maximum at $x = 2$. The complex dependence of $K_1(x)$ for $YFe_{11-x}Co_xTiH_y$ with $y = 0, 1$ results from the partial contributions of different positions of the transition-metal atoms and preferential occupation of only some of the positions by the Co atoms.

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