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ORIGINAL PAPER

Hyperfine interactions in $Tb_{0.27-x}Y_xDy_{0.73}Fe_2$ compounds at 295 K

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Abstract. This paper concerns the synthesis of yttrium-dilute $Tb_{0.27-x}Y_xDy_{0.73}Fe_2$ intermetallic series with a borderline compound $Tb_{0.27}Dy_{0.73}Fe_2$ known as Terfenol-D by arc melting, XRD analysis and ^{57}Fe Mössbauer effect studies at 295 K. XRD analysis (295 K) shows that all samples crystallize in a cubic, *Fd*3*m*, $MgCu_2$-type structure. The lattice parameter increases along with yttrium content. Mössbauer effect spectra for the $Tb_{0.27-x}Y_xDy_{0.73}Fe_2$ series collected at 295 K consist of two subspectra due to easy direction of magnetization <111>. Hyperfine interaction parameters: isomer shift, magnetic hyperfine field and a quadrupole splitting were obtained from the fitting procedure of the spectra.

Key words: hyperfine interaction • intermetallics • Laves phase • Mössbauer effect

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Introduction

Magnetostrictive heavy rare earth (R) – transition metal (M) compounds with formula RM_2 have been widely studied for scientific and practical reasons [3–5]. Therefore, the magnetic properties of RM_2 intermetallics strongly depend on exchange interactions between the 4*f*(5*d*) electrons of the rare earth sublattice and the 3*d* band-type electrons of the transition metal sublattice [10, 11].

From the practical point of view, RFe_2-type materials have been studied for their strong magnetostriction [6, 7, 16]. It has been found that in $Tb_{1-x}Dy_xFe_2$ intermetallics with Tb/Dy substitution in the R-sublattice especially strong magnetostriction has been derived for the $Tb_{0.27}Dy_{0.73}Fe_2$ compound, often called Terfenol-D [6, 7]. Recently RM_2-type compounds have been intensively tested as strongly magnetostrictive constituents of composites with piezoceramics in order to obtain magnetoelectric composites [9, 13].

Due to the lack of a well localized magnetic shell, yttrium is often used in intermetallics instead of rare earths [4, 5]. Yttrium is a non-magnetic metal with the valence electron configuration $4d^5s^2$. We thought it would be revealing to test the influence of the Y/Tb substitution on the 4*f*-4*d*(5*d*)-3*d* magnetism of the $Tb_{0.27-x}Y_xDy_{0.73}Fe_2$ series.

At this stage, it was interesting to study the influence of Tb/Y substitution on the physical properties of Terfenol-D type compounds. Specifically, it was interesting to study the $Tb_{0.27-x}Y_xDy_{0.73}Fe_2$ intermetallics by the ^{57}Fe Mössbauer effect.

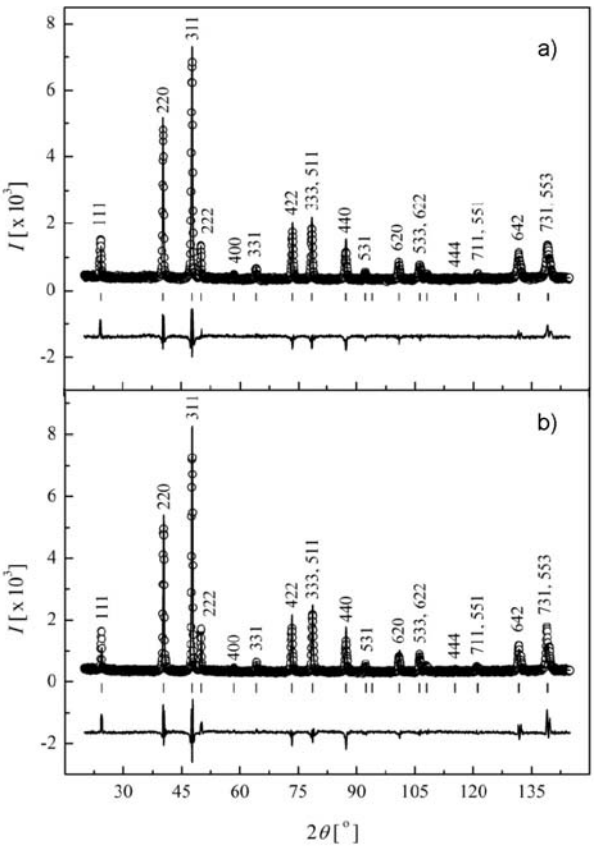

Fig. 1. Exemplary X-ray powder diffraction patterns observed for the compounds: $Tb_{0.27}Dy_{0.73}Fe_2$ (a) and $Y_{0.27}Dy_{0.73}Fe_2$ (b) (295 K). Fitted differential pattern is added below each diffractogram.

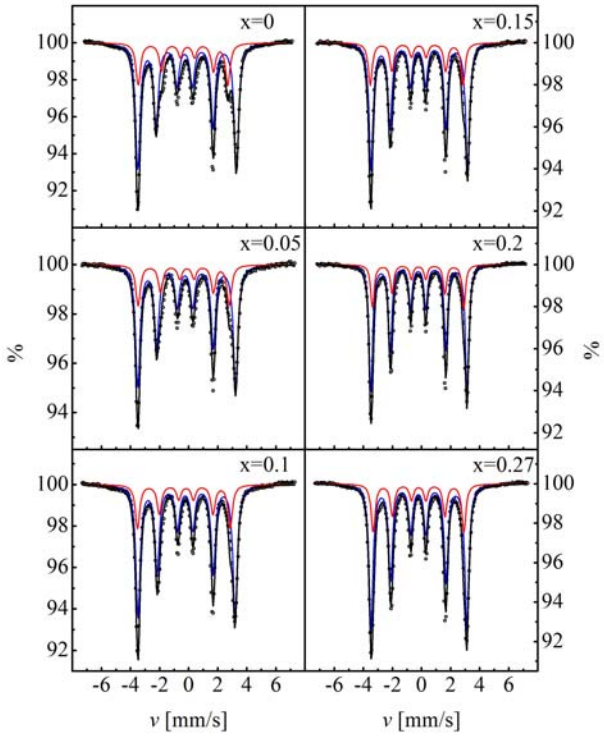

Fig. 2. ^{57}Fe Mössbauer effect spectra of the $Tb_{0.27-x}Y_xDy_{0.73}Fe_2$ intermetallics (295 K).

Materials and studies

Polycrystalline compounds $Tb_{0.27-x}Y_xDy_{0.73}Fe_2$ (x = 0, 0.05, 0.1, 0.15, 0.2 and 0.27) were synthesized by arc melting. The synthesized ingots were then annealed in a vacuum (10^{-6} Pa) at 850°C for 10 h and then allowed to cool down along with the furnace at approximately 250°C/h.

The crystal structures were tested with a standard X-ray powder diffraction procedure (TUR-M61, E. Germany) using CoKα radiation. The exemplary diffractograms for starting and ending compounds are presented in Fig. 1. A cubic, *Fd3m*, $MgCu_2$-type, C15 crystal structure was observed for all studied compounds. It can be added that in the C15 crystal structure each transition metal atom is surrounded by six transition metal atoms as the nearest neighbours and six rare earth metal atoms as the next nearest neighbours [12]. The lattice parameter equals 7.332(1) Å for the $Tb_{0.27}Dy_{0.73}Fe_2$ compound (x = 0) and increases linearly across the series to 7.377(1) Å for the $Y_{0.27}Dy_{0.73}Fe_2$ compound (x = 1).

The ^{57}Fe Mössbauer effect patterns characteristic of the <111> easy axis of magnetization collected at 295 K by using a standard transmission technique with a ^{57}Co in Pd source are presented in Fig. 2. These spectra can be compared because all the samples have the high Curie temperature (above 650 K).

The hyperfine interactions in the studied series depend on both the direction of the easy axis of magnetization [1, 8] and the random distribution of the (Tb,Dy)/Y atoms in the rare earth sublattice. The presence of a magnetic axis renders them distinguishable, because the angle θ between the axis of the electric field gradient and the magnetic field is not the same for all iron atoms. In general, for the direction [uvw] of the easy axis of magnetization there is four magnetically inequivalent sites with different hyperfine interactions due to different angles θ. For the <110> direction of the easy axis, two equivalent sites exist with a ratio 1:1. For the direction <100> of the easy axis, all iron sites have the same θ. If the crystal magnetized along one of the <111> directions, then for some of the iron atoms the directions of the local magnetic field and electric field gradient will not coincide. It can easily be shown that the angle θ between the magnetic and electric axes is zero for one of the iron atoms in a tetrahedron and 70°32' for the other three. Thus, the presence of a non-zero quadrupole interaction would cause the ^{57}Fe Mössbauer spectrum to split into two six-line patterns with intensities in the ratio 1:3. For this reason, all obtained spectra were fitted as a superposition of two six line patterns of intensity ratio 1:3 concerning the <111> easy axis [1, 8].

The average hyperfine interaction parameters weighted by subspectra area with a 75% contribution and subspectra with a 25% contribution, i.e. the isomer shift IS (with respect to iron metal, at 295 K), the magnetic hyperfine field $\mu_0 H_{hf}$ and the quadrupole interaction parameter QS [15], are presented in Fig. 3 close together with the known literature data [2, 14].

The isomer shift IS increases slightly with x, according to the numerical formula $IS(x) = (0.214x^2 - 0.032x - 0.103)$ mm/s (Fig. 3, line 1). The reduction of the $\mu_0 H_{hf}$ field follows the formula $\mu_0 H_{hf}(x) = (-1.525x^2 - 1.028x + 20.541)$ T (Fig. 3, line 2). The quadrupole parameter

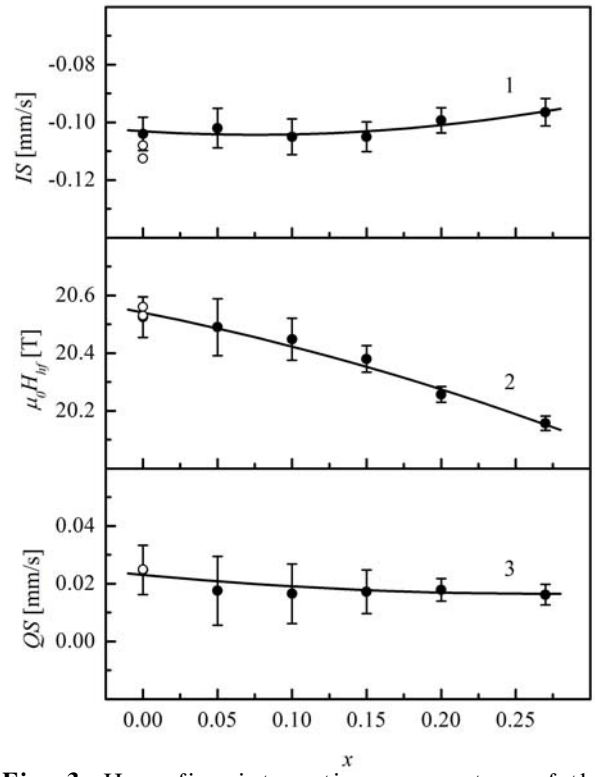

Fig. 3. Hyperfine interaction parameters of the $Tb_{0.27-x}Y_xDy_{0.73}Fe_2$ series. Open points denote the literature data [10, 11].

QS reduces slightly with x as described by the numerical formula $QS(x) = (0.091x^2 - 0.049x + 0.023)$ mm/s (Fig. 3, line 3).

Summary

The $Tb_{0.27-x}Y_xDy_{0.73}Fe_2$ compounds are found to stabilize in the cubic $MgCu_2$ Laves phase structure. The lattice parameter linearly increases with yttrium content.

Yttrium substitution in the $Tb_{0.27-x}Y_xDy_{0.73}Fe_2$ series reduces the average value of magnetic hyperfine field $\mu_0 H_{hf}$. The isomer shift and quadrupole interaction parameter change only slightly with x.

This result can be helpful both in further band type calculations and in practical applications of the Terfenol-D type $Tb_{0.27-x}Y_xDy_{0.73}Fe_2$ compounds, treated potentially as particulate constituents of the magnetoelectric composites.

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