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X-ray and Mössbauer Studies of the $Tb_{0.3}Dy_{0.7}Fe_{2-x}Co_x$ System Alloys

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Abstract—A complex study of the phase composition, atomic and crystalline structure, magnetic properties, and superfine interactions of the $Tb_{0.3}Dy_{0.7}Fe_{2-x}Co_x$ system alloys (x = 0–1.3) has been performed. The synthesized alloys are isotypic with the cubic Laves phase (C15 type), their cell parameter monotonically decreases with an increase in the Co content and, hence, the saturation magnetostriction also does. However, the concentration dependences of the Curie temperature, saturation magnetization, and superfine magnetic field strength measured in the Mössbauer experiment display a nonmonotonic (dome-like) character.

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Alloys and compounds of the rare-earth 4*f* metals with transient 3*d* metals form a special group of new magnetic materials for radio engineering and electronics. Of most interest are the rare-earth Laves phases RT_2 (T = Fe, Co), which have a giant magnetostriction [1–4]. Pseudobinary alloys $(R_{1-x}R'_x)Fe_2$ can be giant magnetostrictive at room temperatures in relatively weak magnetic fields. This can be achieved by using the *R* components with magnetocrystalline anisotropy (MCA) of opposite signs so that the resultant MCA will be diminished owing to their mutual compensation. On this way, the presently popular $Tb_{0.27}Dy_{0.73}Fe_2$ composition called Terfenol has been fabricated. In our opinion, a still deeper compensation of MCA is possible by substituting Co atoms for Fe atoms because the MCA constants of the ions of these elements in the RFe_2 crystalline structure have opposite signs.

The aim of this work is a complex study of the structure and magnetic properties of the $Tb_{0.3}Dy_{0.7}Fe_{2-x}Co_x$ system alloys (x = 0–1.6).

The investigation was based on the data of X-ray phase analysis, Mössbauer spectroscopy, and magnetization and magnetostriction measurements.

RESULTS AND DISCUSSION

The $Tb_{0.3}Dy_{0.7}Fe_{2-x}Co_x$ samples (where x = 0, 0.2, 1.0, 1.1, 1.3, 1.6) were synthesized by the method described in [5] (arc melting followed by homogenizing annealing at T = 750°C for 200 h). The phase composition of the alloys was determined from the X-ray diffraction data. All the diffraction patterns obtained demonstrate the peaks that are definitely induced in the cubic system of the rare-earth Laves phase of the C15 type. According to the X-ray data, the alloys are single-phase. The cell parameter of the $Tb_{0.3}Dy_{0.7}Fe_{2-x}Co_x$ system alloys was calculated and the corresponding concentration dependence $a(x)$ was plotted (Fig. 1). It is seen that, as the Co content in the system increases from x = 0 to x = 1.6, parameter a decreases monotonically but nonlinearly because the metallic radius of the Co atom is smaller than that of the Fe atom. This result correlates with calculations using Vegard's law.

The techniques used for measuring magnetization and magnetostriction were described in detail in [5].

The Curie temperature (T_C) was determined from temperature dependences $\sigma(T)$ measured in a weak magnetic field H = 500 Oe as the temperature of the sharpest drop of magnetization during the ferromagnetic–paramagnetic transition. Figure 2 shows the concentration dependence of T_C. The curve $T_C = f(x)$ is

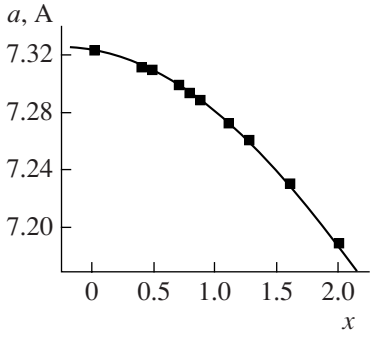

Fig. 1. Concentration dependence of the cell parameter, $a(x)$.

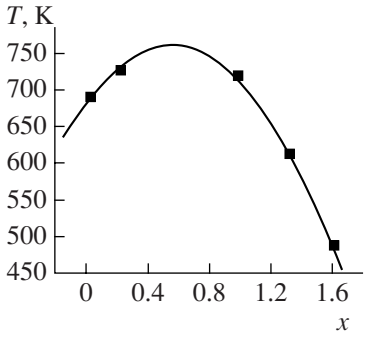

Fig. 2. Curie temperature as a function of concentration.

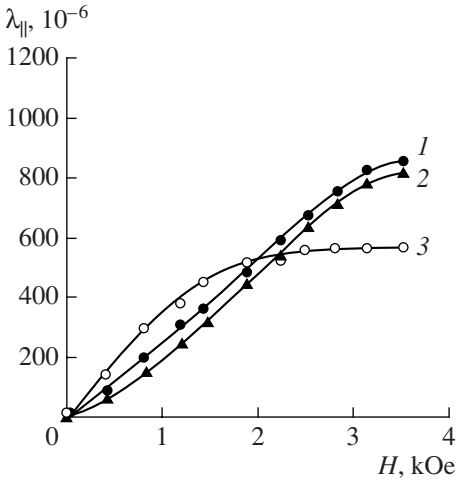

Fig. 3. Field dependence of longitudinal magnetostriction λ for the $Tb_{0.3}Dy_{0.7}Fe_{2-x}Co_x$ compounds at $x = (1)$ 0, (2) 0.2, and (3) 1.3 at $T = 300$ K.

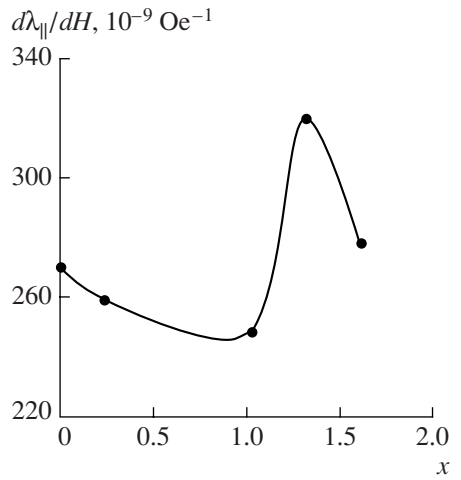

Fig. 4. Concentration dependence of $\partial\lambda_{\parallel}/\partial H$ for the $Tb_{0.3}Ho_{0.03}Dy_{0.67}Fe_{2-x}Co_x$ system alloys at $T = 300$ K.

dome-shaped with a maximum falling at the intermediate concentration range $0.2 < x < 1.0$.

Figure 3 plots the field dependences of the longitudinal magnetostriction of the alloys at $T = 300$ K in the range from 0 to 3.5 kOe. It is clearly seen that the magnetostriction decreases with an increase in the Co content. However, as mentioned above, the technical applicability of the alloys depends on whether it is possible to achieve high magnetostriction values at low magnetic fields (i.e., to obtain high values of magnetostriction susceptibility). Figure 3 demonstrates that, in the region of weak field $H \le 2$ kOe, the values of magnetostriction for the $Tb_{0.3}Dy_{0.7}Fe_{0.7}Co_{1.3}$ composition (curve *3*) are higher than those for the compositions with a high Fe content: $Tb_{0.3}Dy_{0.7}Fe_2$ (curve *1*) and $Tb_{0.3}Dy_{0.7}Fe_{1.8}Co_{0.2}$ (curve *2*). Figure 4 shows the appearance of magnetostriction susceptibility $\partial\lambda_{\parallel}/\partial H$ as a function of the Co concentration in the $Tb_{0.3}Dy_{0.7}Fe_{2-x}Co_x$ system at $T = 300$ K. It is seen that the maximal value of $\partial\lambda_{\parallel}/\partial H$ is obtained for the composition with $x = 1.3$.

Mössbauer spectra of the $Tb_{0.3}Dy_{0.7}Fe_{2-x}Co_x$ alloys were measured using an $Ms1104E_m$ spectrometer. Figure 5 presents the Mössbauer spectrum of the $Tb_{0.3}Dy_{0.7}Fe_2$ alloy. The Mössbauer spectra were processed by a UNIVEM MS program.

A specific feature of the crystallographic structure of the RFe_2 alloys is that, for three out of four Fe atoms, the electric field gradient (EFG) axis makes the same angle of 72° with the easy magnetization axis ([111]). It is only for the fourth atom of the cell that the EFG and magnetization axes coincide. Therefore, in processing the Mössbauer spectra, a two-sextet model was used (one sextet for three Fe atoms and the other, for one atom in the unit cell). As a result, we obtained the result that (i) the isomer shifts and quadrupole splits are almost invariable from alloy to alloy; (ii) the intensity ratio of the two sextets varies near 3 : 1; and (iii) the line widths vary from 0.28 mm/s for the alloy with $x = 0$ to 0.48 mm/s for the alloy with $x = 1.3$ (this points to the local irregularities arising as the Co atoms are substituted for Fe atoms).

Much information was extracted from measurements of superfine field strength H_{eff}. Figure 6 plots the concentration dependence of superfine magnetic fields H_1 and H_2 ($H_{\text{eff}} = f(x)$). These dependences are seen to be dome-like.

It is interesting to compare the properties of these alloys with the properties of the Fe–Co and Fe–Ni alloys and other transient metal alloys. The values of the saturation magnetization per atom measured in Bohr magnetons are maximal for the Fe–Co alloy and decrease linearly both toward Mn, Cr and to Ni, Cu (the Slater-Pauling curve). The comparison allows us to conclude that the iron-based compounds with the Laves phase structure have simple magnetic properties because the magnetic properties of the Fe sublattice are almost independent of the rare-earth ions. The latter

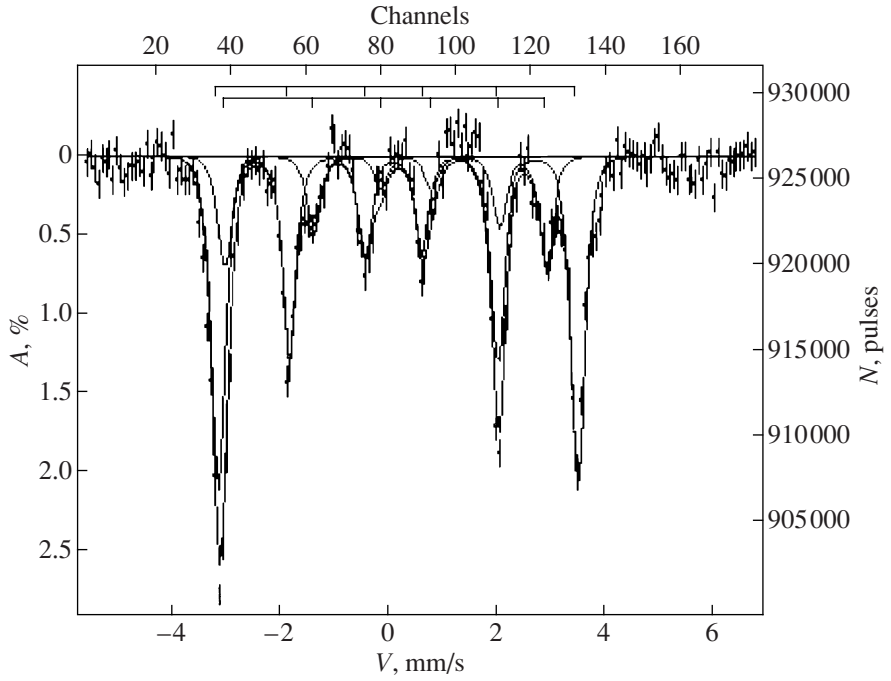

Fig. 5. Mössbauer spectrum of the $Tb_{0.3}Dy_{0.7}Fe_2$ alloy.

behave almost like paramagnetic ions arranged by the field exerted by the Fe sublattice.

Thus, the results of the complex study of the $Tb_{0.3}Dy_{0.7}Fe_{2-x}Co_x$ system can be summarized as follows: (1) nearly single-phase compounds isotypic with the Laves cubic phase C15 have been synthesized in the concentration range $0 \le x \le 1.6$; (2) it has been shown that, as the Co content increases from $x = 0$ to $x = 1.3$, the unit cell parameter decreases monotonically from $a = 7.328$ Å to $a = 7.295$ Å; (3) the Curie temperature has been found to increase in the concentration range $0.2 \le x \le 1.0$ and then decrease, featuring a dome shape; (4) the superfine field strengths H_1 and H_2 at the Fe^{57} nuclei were determined in two positions of the Fe atom, and their concentration dependences also appear to be dome-shaped; (5) it has been found that the easy magnetization axis coincides with the ⟨111⟩ direction in all the alloys, thus, letting the Fe atoms be in two magnetically nonequivalent positions; and (6) it has been shown that the high Co concentration in these alloys makes it possible to achieve high magnetostriction values in weak (to 2 kOe) magnetic fields. The composition with the Co content $x = 1.3$ has the maximal value of $\partial\lambda_{\parallel}/\partial H$ and, therefore, holds much promise for practical applications in the field range from 1 to 2 kOe.

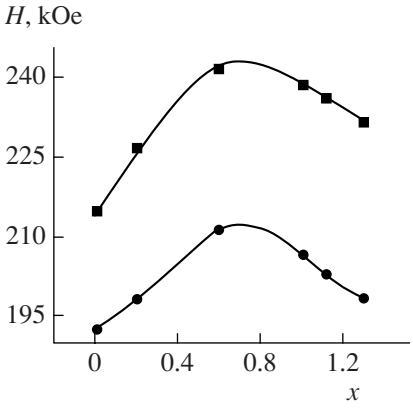

Fig. 6. Superfine field in the $Tb_{0.3}Dy_{0.7}Fe_2$ alloy as a function of x.

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