Effect of 3D-Substitutional Atoms on the Magnetic and Magnetostrictive Properties in (Tb,Ho)Fe_2 Laves Phase

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***Abstract*—The crystal structure, magnetic and magnetostrictive properties of the Laves phases $Tb_{0.16}Ho_{0.84}Fe_{2-x}T_x$ (T = Co, Ni, x = 0.1, 0.2, 0.3, 0.4) are investigated. Polycrystalline samples were prepared by induction melting with subsequent homogenizing annealing. The studied compounds crystallize in a structure of the $MgCu_2$ type. Partial substitution of iron by Co and Ni leads to a decrease in the parameter and volume of the unit cell by ~3%. The Curie and spin reorientation temperatures of the studied compounds were determined, magnetic phase diagrams were constructed. Compounds with high values of saturation magnetostriction and magnetostrictive susceptibility in the temperature range of 190-320 K were found.**

Keywords—soft magnetic materials, magnetostriction, laves phases, spin reorientation effect

I. Introduction

Rare earth metals, their alloys and compounds based on them are widely used in various fields of modern industry, such as electronics, metallurgy, oil refining, nuclear power, glass, ceramics, optical industry, and medicine [1-2]. It is well known [3-4] that highly magnetostrictive materials are used as actuators and sensors operating in a wide range of environments and temperatures due to their ability to convert electrical energy into mechanical energy. Rare earth Laves phases form an important family of magnetic materials exhibiting large magnetostriction values (of the order of 10^{-3}) in the technically useful temperature range. Some Laves phases near the Curie temperature can exhibit both high magnetostriction values and a large magnetocaloric effect, which may also have potential practical applications [5-8].

It is known that intermetallic compounds with the Laves phase structure RFe_2 (R = rare earth (R) elements) crystallize in a cubic structure of the $MgCu_2$ type of the Fd3m space group and exhibit giant anisotropic magnetostriction at room temperature [9-10]. Significant spontaneous magnetostriction caused by magnetic ordering in these compounds leads to deformation of the cubic crystal lattice along the easy axis. The best known rare earth Laves phases are $TbFe_2$ and $SmFe_2$, which have close in magnitude but opposite in sign magnetostriction values at room temperature. Their saturation magnetostriction at room temperature reaches values of $+1.7\cdot10^{-3}$ and $-1.5\cdot10^{-3}$, respectively [11-12].

For the technical use of such materials, it is important to achieve high magnetostriction in weak magnetic fields, i.e. high values of magnetostrictive susceptibility. Another significant parameter is the saturation magnetostriction, which is usually achieved in a strong magnetic field. Achieving high magnetostrictive susceptibility and reducing the saturation field values can be achieved by partial substitution of both rare earth elements and iron. This leads to compensation of magnetocrystalline anisotropy (MCA). The best known composition with MCA compensation is (Tb,Dy)Fe_2 (Terfenol-D), widely used as ultrasound sources, displacement sensors, level sensors, etc. [13-15]. $HoFe_2$ also exhibits significant saturation magnetostriction at room temperature. However, the value required to achieve saturation is significant, exceeding 17 kOe [16]. $HoFe_2$ and $TbFe_2$ have opposite signs of the first MCA constant. Ternary compounds like (Tb,Ho)Fe_2 can have low MCA and, as a consequence, high magnetostrictive susceptibility [12,17]. Partial substitution of Fe by Co or Ni also improves the magnetic and magnetostrictive properties of pseudobinary alloys (R,R')Fe_2 [6, 18-24]. The aim of this work was to study the effect of partial substitution of iron by Co and Ni in the compound (Tb,Ho)Fe_2 on the structure and magnetic properties and to search for new materials with high magnetostrictive characteristics.

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II. Experimental Details

Synthesis of $Tb_{0.16}Ho_{0.84}Fe_{2-x}T_x$ (T =Co, Ni, x = 0.1, 0.2, 0.3, 0.4) compounds was carried out by induction melting in purified argon atmosphere at excess pressure of 1.1 atm using ultra-pure rare earth metals. Homogenization annealing at 900 °C for two weeks was used to obtain the most homogeneous single-phase alloy. X-ray structural analysis was carried out at room temperature on an Ultima IV powder diffractometer (Rigaku, Japan) with CuKα radiation.

The magnetization measurements in the temperature range 300–700 K were carried out on a vibrating-sample magnetometer in an applied field up to 10 kOe. The samples magnetization was measured using an induction magnetometer (insert to the MagEq MMS 901 setup, AMT&C, (Troitsk, Moscow)) (magnetic fields up to 12 kOe in the temperature range from 90 to 350 K). To measure magnetostriction, a strain gauge method was used, with a sensor with a 5 mm base and a resistance of 120 Ω (in the temperature range of 80-350 K in magnetic fields up to 12 kOe). Measurements were carried out along (longitudinal magnetostriction, $\lambda_{||}$) and perpendicular (transverse magnetostriction, $\lambda_{\perp}$) to the direction of the applied magnetic field. Anisotropic magnetostriction was calculated as the difference between the longitudinal and transverse magnetostriction ($\lambda_a = \lambda_{||} - \lambda_{\perp}$).

III. Results and Discussion

The Rietveld method was used to determine the lattice parameter for the main phase in the alloys under investigation. As shown in Table I, the compounds $Tb_{0.16}Ho_{0.84}Fe_{2-x}T_x$ (T = Co, Ni) exhibit variations in the unit cell parameter. When iron is partially substituted with Co or Ni ($x \leq 0.4$) the unit cell parameter and volume of the compound $Tb_{0.16}Ho_{0.84}Fe_2$ decrease by ~3%. This trend was also observed when iron was partially replaced by Co in $Tb_{0.27}Dy_{0.73}Fe_2$ compound [20], as well as in other rare-earth alloys with the Laves phase crystal structure [25-26].

The field dependencies of longitudinal ($\lambda_{||}$) and transverse ($\lambda_{\perp}$) magnetostriction of $Tb_{0.16}Ho_{0.84}Fe_{2-x}T_x$ (T = Co, Ni) compounds were studied at different temperatures. In our previous work, we showed that, at room temperature, in Co-substituted compounds, $\lambda_{||}$ was positive while transverse magnetostriction was negative. However, the transverse magnetostrictive effect weakly depended on Co concentration and required high fields greater than 5 kOe to saturate.

In Ni-substituted alloys the transverse magnetostriction decreases with increasing Ni content. The alloy $Tb_{0.16}Ho_{0.84}Fe_{1.8}Ni_{0.2}$ shows the lowest values of $\lambda_{||}$. The absolute value of the longitudinal magnetostriction is greater than that of the $\lambda_{\perp}$ for Ni-substituted compositions. However, the value of $\lambda_{\perp}$ in the $Tb_{0.16}Ho_{0.84}(Fe,Ni)_2$ system exceeds the value of $\lambda_{||}$ in a system with iron replaced with cobalt.

Fig. 1 shows the field dependence of the anisotropic magnetostric effect in $Tb_{0.16}Ho_{0.84}(Fe,Ni)_2$ alloys. We found that these alloys exhibited significant positive anisotropic magnetization at room temperature and the highest values of λ_a were 700 ppm for Ni substitution and 500 ppm in the case of Co substitution. It has been shown that the volume magnetostriction is significantly less than anisotropic as usual for most RFe_2 compounds [27-29], for example, $\omega = -160$ ppm at $Tb_{0.16}Ho_{0.84}Fe_{1.9}Co_{0.1}$.

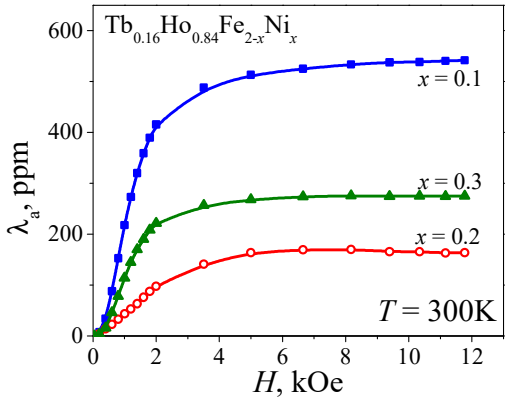

Fig. 1. Anisotropic magnetostriction field dependences of the $Tb_{0.16}Ho_{0.84}Fe_{2-x}Ni_x$ compound in temperature T = 300 K.

It is known that at the spin-reorientation (SR) transition temperature, the crystal lattice of the studied compounds is deformed and its structural type changes [26-31]. In this case, near the T_{SR}, the maximum and minimum values of anisotropic magnetostriction should be observed. Fig. 2 shows the temperature dependences of the anisotropic magnetostriction of $Tb_{0.16}Ho_{0.84}Fe_{2-x}T_x$ (T = Co, Ni) compounds in a magnetic field of 2 kOe. With an increase in the Co concentration, the maximum magnetostriction in the studied alloys shifts toward low temperatures, and the minimum does not change. In the composition with Ni substitution, with an increase in the Ni concentration, the maximum magnetostriction significantly shifts toward high temperatures (by ~40 K). The minimum magnetostriction shifts toward low temperatures. It should be noted that if T_{SR1} decreases with increasing cobalt concentration, then when replacing Fe with Ni, T_{SR1}, on the contrary, increases. The temperature of another transition T_{SR2} slightly decreases with increasing nickel content, while T_{SR2} for the system with cobalt are practically the same. Table I shows the temperatures T_{SR1} and T_{SR2}, taken as the temperatures of the minimum and maximum of the anisotropic magnetostriction obtained in a magnetic field of 2 kOe.

TABLE I. Unit Cell Parameters, Magnetic and Magnetostrictive Characteristics of the $Tb_{0.16}Ho_{0.84}Fe_{2-x}T_x$ (T = Co, Ni) Compounds

Compounds	a, Å (T = 300 K)	V, Å³ (T = 300K)	T_C, K	T_{SR1}, K	T_{SR2}, K	λ_{111}, Oe^{-1} 10^{-6} (T = 300 K)
x = 0.1 (T = Co)	7.312(1)	390.94	610	112	250	368.1
x = 0.2 (T = Co)	7.309(5)	390.46	638	115	229	585.6
x = 0.3 (T = Co)	7.307(7)	390.14	645	125	232	469.2
x = 0.4 (T = Co)	7.305(3)	389.82	648	113	234	654.7
x = 0.1 (T = Ni)	7.315(6)	391.42	565	246	134	697.5
x = 0.2 (T = Ni)	7.312(1)	390.94	538	275	115	237.3
x = 0.3 (T = Ni)	7.308(4)	390.30	527	271	113	423.3

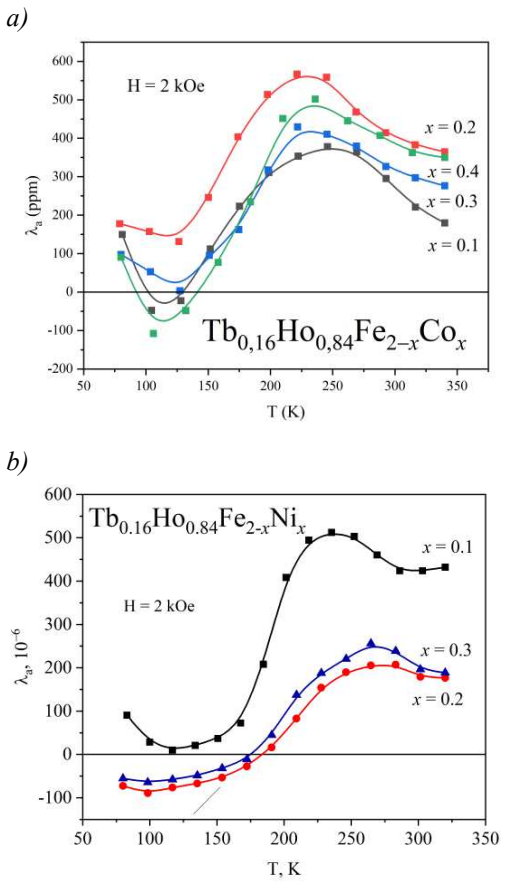

Fig. 2. Anisotropic magnetostriction temperature dependences of the $Tb_{0.16}Ho_{0.84}Fe_{2-x}T_x$ compound (T = Co (a), Ni (b)) in field of 2 kOe.

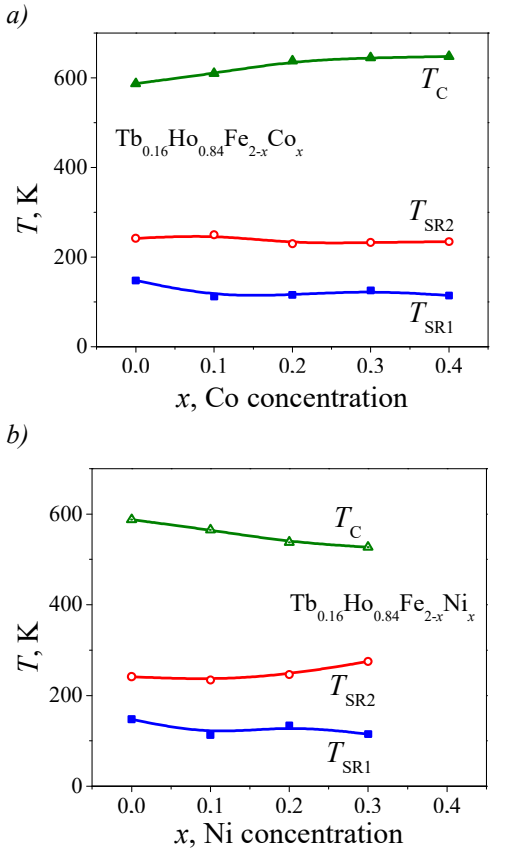

Fig. 3. Magnetic phase diagram of $Tb_{0.16}Ho_{0.84}Fe_{2-x}T_x$ compound (T = Co (a), Ni (b)) in field of 2 kOe.

Table I presents the temperatures of SR phase transitions of all $Tb_{0.16}Ho_{0.84}Fe_{2-x}T_x$ (T = Co, Ni) systems. Using the Curie temperatures found from thermomagnetic measurements and the SR transition temperatures obtained from magnetostriction studies, a magnetic phase diagram of the studied systems was constructed (see Fig. 3). According to the crystal field theory, as a rule, an approximation is used in which magnetic phase transitions are not accompanied by a change in cubic symmetry (Fd3m). However, numerous studies have been shown that in case of the Laves phase's one can speak of magnetostructural phase transitions, the set of which can be considered as a morphotropic phase boundary in the magnetostructural phase diagram.

One of the defining characteristics that determines the practical application of magnetostrictive alloys is the achievement of high values of magnetostrictive deformations in weak magnetic fields, that is, magnetostrictive susceptibility. The field dependence of the reduced magnetostriction $\lambda/\lambda s$ at room temperature in moderate magnetic fields for Co- and Ni-substituted compounds is shown in Fig. 4, a and Fig. 4, b respectively. It is evident from the figure that among the studied compositions, the compound $Tb_{0.16}Ho_{0.84}Fe_{1.8}Co_{0.2}$ has the highest magnetostrictive susceptibility ($d\lambda/dH = 34.4 \cdot 10^{-8}$ Oe^{-1}). The magnetostrictive susceptibility was determined as the maximum slope angle of the linear portion of the field dependence of the anisotropic magnetostriction in fields from 0.6 to 1.6 kOe for Co-substituted compounds.

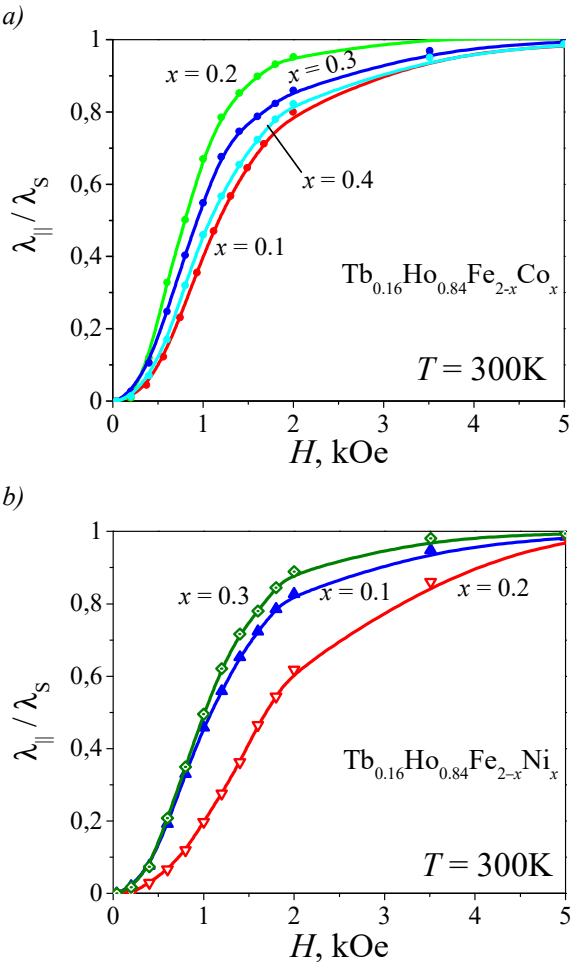

Fig. 4. Field dependencies of reduced magnetostriction of $Tb_{0.16}Ho_{0.84}Fe_{2-x}T_x$ compound (T = Co (a), Ni (b)) in room temperature in magnetic field up to 5 kOe.

In cubic crystals, anisotropic magnetostriction is usually characterized by two main constants λ_{100} and λ_{111}, i.e. the relative change in the crystal size in the [100] and [111] directions upon magnetization. In our case, the distortions are rhombohedral, so $\lambda_{100} \ll \lambda_{111}$ [3]. Knowing the saturation values of longitudinal magnetostriction from experimental data, we can calculate the magnetostriction constant λ_{111}, which is responsible for the rhombohedral distortion of the cubic crystal lattice, according to the formula:

$$\lambda_{\parallel s} = \frac{3}{5}\lambda_{111} - \frac{2}{5}\lambda_{100}.$$

The magnetostriction constants λ_{111} are shown in Table I. The obtained magnetostriction constants λ_{111} of the studied alloys are significantly lower than the magnetostriction constant of Terfenol-D (1640 ppm) [9, 32]. However, magnetostrictive susceptibility, $d\lambda/dH$ of the studied alloys in weak fields (~1 kOe) exceeds its value for Terfenol-D ($20 \cdot 10^{-8}$ Oe^{-1}) and some substituted alloys based on it ($27 \cdot 10^{-8}$ Oe^{-1} [33]). The values of the Curie temperature of $Tb_{0.16}Ho_{0.84}(Fe,Co)_2$ compounds are close to T_C of Terfenol-D (T_C = 650 K) [8], which is important for technical use.

IV. Conclusion

In this work, the effect of partial substitution of Fe by Co and Ni on the magnetic properties of the $Tb_{0.16}Ho_{0.84}Fe_2$ compound is investigated. Magnetic and magnetostrictive properties of $Tb_{0.16}Ho_{0.84}Fe_{2-x}T_x$ (T = Co, Ni, x = 0.1, 0.2, 0.3, 0.4) polycrystals are studied in a wide range of temperatures and magnetic fields. The temperatures of magnetic phase transitions are determined, and a magneto phase diagram is constructed. It is found that partial substitution of Fe by Co and Ni in both systems leads to an increase in anisotropic magnetostriction. The highest value of anisotropic saturation magnetostriction (λ_a = 500 ppm) of the studied systems, as well as the highest value of magnetostrictive susceptibility ($d\lambda/dH = 34.4 \cdot 10^{-8}$ Oe^{-1}), is possessed by the $Tb_{0.16}Ho_{0.84}Fe_{0.8}Co_{0.2}$ compound, which is therefore the most promising for practical application.

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