Structural features and magnetic properties of Laves phases (Tb,Y,Sm)Fe_2

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Abstract. The paper presents the results of investigation of structural, magnetic and magnetostrictive properties of alloys of multicomponent system $(Tb_{1-x}Y_x)_{0.8}Sm_{0.2}Fe_2$ with substitution parameter x = 0, 0.2, 0.4, 0.6, 0.8, 1. Structural, magnetisation and magnetostriction studies were carried out in a wide temperature range. The presence of compensation points on the magnetisation curves for both concentration and temperature was found. Inversion of the sign of magnetostriction constants in the region of values of $0.8 \leq x \leq 0.9$. Key words: Laves phases, magnetisation, Curie temperature, magnetostriction.

1 Introduction

One of the most important scientific and technical tasks of modern condensed state physics is the search for new magnetic materials with high magnetostrictive parameters for modern innovative equipment. In this connection, intermetallic compounds of rare-earth metals with 3d - transition metals have become widespread in science and technology due to their outstanding physical and chemical properties (Mn, Fe, Co and Ni) type RFe_2. The above compounds possess the structure of Laves C15 or C14 phases [1-4]. The most interesting properties are those that depend on the structural features of RFe2 compounds, in other words, they are structure-sensitive properties.

It is known [5-6] that Laves phases are convenient model objects for studying a number of fundamental problems of modern condensed state physics, including the establishment of the relationship between the electronic characteristics of atoms or ions constituting a solid and its physical properties. They combine a relatively simple crystalline structure and unique magnetic properties such as giant magnetostriction and large magnetocaloric effect arising under the action of an external magnetic field. Therefore, the study of the structure of Laves phases and physical properties is an urgent task and allows us to obtain and predict materials with a given complex of properties.

Connections of the type RFe_2 with cubic structure like $MgCu_2$ (C15), can be considered as two-sublattice lattices. Two lattices - of iron ions and rare-earth metal ions - are inserted into each other.

Connections RFe_2 with light REMs have ferromagnetic ordering, namely magnetic moments of the iron sublattice are orientated to the moments of the rare-earth sublattice.

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The purpose of this work is to investigate the structural features and magnetic properties in alloys of the system of $(Tb_{1-x}Y_x)_{0.8}Sm_{0.2}Fe_2$.

2 Samples and experimental techniques

Synthesis of alloys $(Tb_{1-x}Y_x)_{0.8}Sm_{0.2}Fe_2$ (x = 0, 0.2, 0.4, 0.6, 0.8, 1) was carried out in a Leybold-Heraeus electric arc furnace (ingot weight 20 g) with a non-consumable tungsten electrode on a specially designed copper water-cooled bed in an argon atmosphere at normal pressure. Metals of high purity (99.978 wt.%) were taken as initial components. The samples were subjected to subsequent homogenising annealing in vacuum sealed quartz ampoules at temperature 850-900°C within 2 weeks.

Certification of alloys carried out by X-ray diffractometer phase analysis method «DRONE-2» (CuKα-radiation) described in References [6].

Magnetostriction measurements were carried out in the region of low and room temperatures in magnetic fields up to 12 kE using the strain gauge method. The magnetostriction measurement error did not exceed 6%. Field and temperature dependences of magnetisation were measured using standard equipment in the temperature range from 4.2 to 300 K. In addition, the temperature dependences of ac susceptibility in the temperature range from 300 to 800 K were obtained.

3 Results and discussion

Fig. 1 shows the concentration dependences of the pseudocubic cell of the compounds of the system $(Tb_{1-x}Y_x)_{0.8}Sm_{0.2}Fe_2$, obtained at different temperatures. It can be seen that at low temperatures with increasing yttrium concentration a monotonic, close to linear, growth of the unit cell parameter is observed. At temperatures above room temperature, the dependence is not monotonic, a minimum is observed at yttrium concentrations x = 0.4 – 0.6.

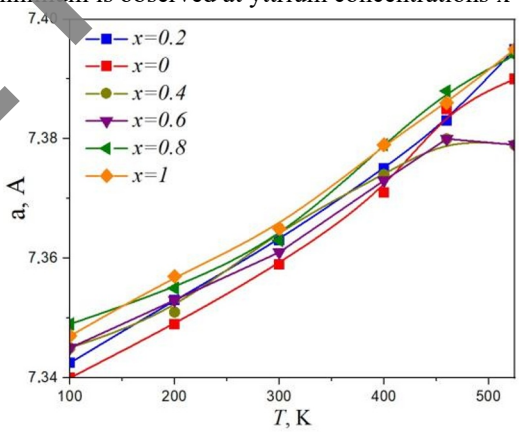

Fig. 1. Dependence of the unit cell parameter of the system compositions $(Tb_{1-x}Y_x)_{0.8}$ $Sm_{0.2}Fe_2$ from temperature.

According to the above, in compounds with x = 0, 0.2 and 0.4 at room temperature, a splitting of peaks is observed, indicating rhombohedral distortions of the cubic crystal lattice.

Fig. 2 shows the dependence of thermal expansion (TE) $(Tb_{1-x}Y_x)_{0.8}$ $Sm_{0.2}Fe_2$ alloys from temperature in the temperature range from 80 to 300 K, in this temperature range the compounds are magnetically active, since the Curie temperatures of all these alloys exceed 600 K (Table 1).

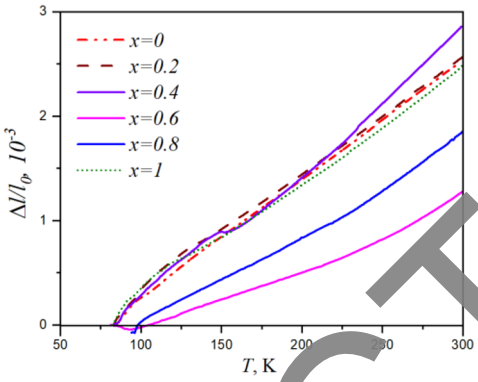

Fig. 2. Dependence of thermal expansion of alloys on system temperature $(Tb_{1-x}Y_x)_{0,8}Sm_{0.2}Fe_2$

In the investigated temperature region TR is caused by both phonon contribution and contribution from spontaneous magnetostriction. The figure shows that for alloys with concentrations x=0; 0.2; 0.4, 1 TR have similar behaviour. The composition with x=0.6 has the lowest thermal expansion value among the compositions. Magnetic measurements of this composition showed that the magnetic moment values of samarium, terbium and iron sublattices are compensated. The results for TR suggest that these concentrations of terbium and samarium ions allowed to compensate the negative and positive contributions to the spontaneous magnetostriction from samarium and terbium.

To determine the Curie temperature, ac susceptibility measurements were performed at temperatures above 300 K for all initial compositions. The Curie temperature was determined from the peak in the temperature dependence of the susceptibility. Values T_C for connections $(Tb_{1-x}Y_x)_{0.8}Sm_{0.2}Fe_2$ are given in Table 1. The dependence of the Curie temperature on the concentration of the substituent element is shown in Fig. 3. As it is known [8, 9], the Curie temperature of compounds RFe_2 is determined primarily by the contribution of the integral of the exchange interaction within the Fe-Fe iron sublattice and the contribution of the exchange integral of the R-Fe antilattice interaction. Value T_C for $Tb_{0.8}Sm_{0.2}Fe_2$ amounts to 670 K, which is less than the Curie temperatures of the compounds $TbFe_2$ (T_C = 682 K) and $SmFe_2$ (T_C = 700 K). At dilution of the rare sublattice of compounds $Tb_{0.8}Sm_{0.2}Fe_2$ by nonmagnetic yttrium ions there is a monotonic decrease in the Curie temperature with increasing Y concentration, caused by a decrease in the integral of the R-Fe exchange interaction. Curie temperature of ferromagnetic compound $Y_{0.8}Sm_{0.2}Fe_2$ almost identical to T_C connections YFe_2 [10] and significantly lower than T_C connection $SmFe_2$ (700 K). This fact allows us to conclude that the strong dilution of the rare-earth sublattice of the compound $SmFe_2$ by yttrium ions suppresses the Sm-Fe exchange interaction.

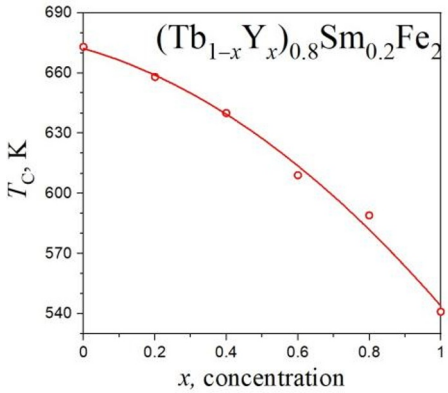

Fig. 3. Concentration dependence of the Curie temperature of the system $(Tb_{1-x}Y_x)_{0.8}Sm_{0.2}Fe_2$.

Table 1. Magnetic characteristics of compounds $(Tb_{1-x}Y_x)_{0.8}Sm_{0.2}Fe_2$: Curie temperature ($T_C$), saturation magnetisation (M_{sat}) and spontaneous magnetisation (M_S) at a temperature of 4.2 K

x	T_C, K	μ_{sat} (μ_B)	μ_S(μ_B)
0	670	3.57	3.2
0.2	658	2.20	2.2
0.4	640	1.47	1.2
0.6	609	0.48	0.3
0.8	589	2.02	1.6
1	541	2.44	1.6

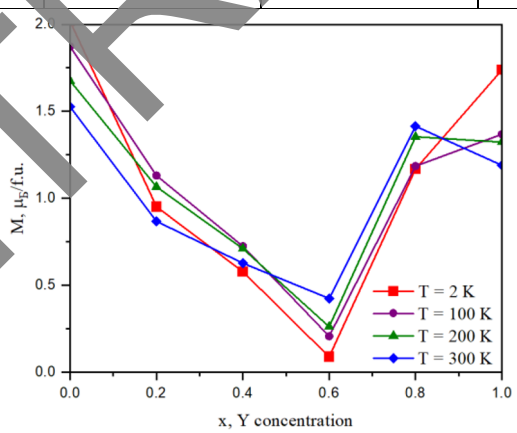

Fig. 4. Dependence of magnetization of alloys $(Tb_{1-x}Y_x)_{0.8}Sm_{0.2}Fe_2$ of the substitution parameter x in the temperature range from 2 to 300 K.

Fig. 4 shows the concentration dependence of the magnetisation in a 2 kE field for temperatures from 2 to 300 K. It can be seen that in the region of the parameter x = 0.6 in the system $(Tb_{1-x}Y_x)_{0.8}Sm_{0.2}Fe_2$ magnetic compensation of the rare-earth and iron sublattices is observed. For the connection $(Tb_{0.4}Y_{0.6})_{0.8}Sm_{0.2}Fe_2$ magnetisation practically drops to zero compared to the other compounds, i.e., the sum of magnetic moments of the sublattices (directed antiparallel to each other) compensate each other.

The composition and temperature at which the compensation effect occurs are called the compensating composition and compensation temperature, respectively [17, 19]. The most important factors for the transformation of this effect are atomic concentration and temperature. This phenomenon has an important practical significance; by varying the concentration of substitutional atoms near the compensating composition, the magnitude of the spontaneous magnetisation can be controlled.

Temperature studies of the magnetization of the composition $(Tb_{0.4}Y_{0.6})_{0.8}Sm_{0.2}Fe_2$ (x = 0.6) show that at the temperature T_{comp} = 136 K full compensation of magnetisation is observed for the given composition.

From the results of these studies, the saturation magnetisation, spontaneous magnetisation at 4.2 K, and Curie temperature for each composition were determined. To calculate the saturation magnetisation, the dependences of the magnetisation on the inverse of the magnetic field M(1/H) were plotted. The value of saturation magnetisation (M_{sat}) is determined by extrapolation of the M(1/H) dependence to the maximum field (1/H→0). The value of spontaneous magnetisation (M_S) was determined by linear extrapolation of the magnetisation curve to the zero field. Values M_{sat} and M_S connection $(Tb_{1-x}Y_x)_{0.8}Sm_{0.2}Fe_2$ at a temperature of 4.2 K are shown in Table for compositions $(Tb_{1-x}Y_x)_{0.8}Sm_{0.2}Fe_2$ values of the spontaneous magnetisation and saturation magnetisation at low temperatures differ insignificantly, which is caused by the small value of the paraprocess susceptibility. As can be seen from Table 1, the minimum value of saturation magnetisation and spontaneous magnetisation is observed for the composition with x = 0.6, which is the closest to the compensating composition.

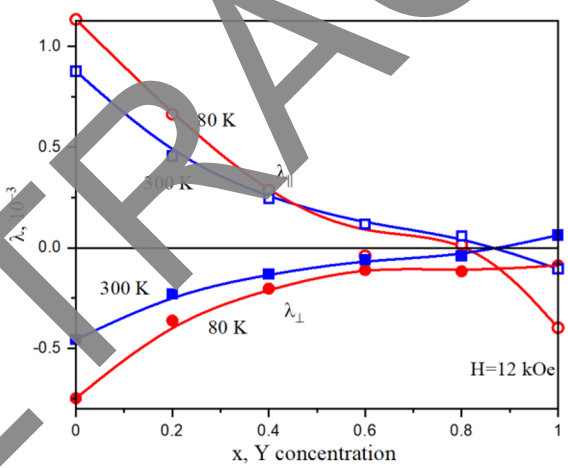

Fig. 5. Longitudinal $\lambda_{||}$ and trasverse $\lambda_{\perp}$ magnetostriction of alloys $(Tb_{1-x}Y_x)_{0.8}Sm_{0.2}Fe_2$ depending on the substitution parameter x.

In Fig. 5 shows the values of longitudinal and transverse magnetostriction of alloys of the system $(Tb_{1-x}Y_x)_{0.8}Sm_{0.2}Fe_2$ depending on the substitution parameter x. It is found that longitudinal magnetostriction of all alloys of the system $(Tb_{1-x}Y_x)_{0.8}Sm_{0.2}Fe_2$ with substitution parameter x ≤ 0.8 in an external magnetic field H = 12 kE is positive, and transverse magnetostriction is negative. In alloys with a value of x ≥ 0.9, on the contrary, longitudinal magnetostriction is negative and transverse magnetostriction is positive. Thus, in this system in the region of the parameter x = 0.8, the sign of the magnetostriction constants is reversed.

4 Conclusions

A special feature of the selected compositions is the fact that, the compound $TbFe_2$ demonstrates positive spontaneous and induced magnetostriction, while the compound $SmFe_2$ – negative. In the system $(Tb,Y,Sm)Fe_2$ sign inversion of the magnetostriction constants occurs in the region of values of the parameter of substitution of terbium by yttrium $0.8 \leq x \leq 0.9$.

Dilution of the rare-earth sublattice with nonmagnetic yttrium and increasing the distance between magnetically active terbium and samarium atoms leads to a change in the sign of the Tb-Fe and Sm-Fe exchange interactions competing with each other at a certain yttrium concentration.

Moreover, by varying the concentration of the rare-earth component (terbium) it is possible to obtain a composition with full magnetic compensation of magnetisation in the whole temperature range of magnetic ordering.

combination of ions of light and heavy elements (terbium and samarium) in one and the same rare-earth sublattice allowed to reveal compositions of such alloys of multicomponent system $(Tb,Y,Sm)Fe_2$, which have fully or almost fully compensated magnetocrystalline anisotropy and, therefore, can be expected to have the composition with the most optimal magnetostrictive characteristics [11]. This is an alloy with the substitution parameter xcomp = 0.6. The combination of the above two factors at once has not been investigated before.

Compositions with the specified features of magnetic and magnetostrictive properties can be used in such fields as robotics.

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